



Cite this: *Environ. Sci.: Processes Impacts*, 2020, 22, 2291

## Modeling of $^{210}\text{Pb}$ and $^{210}\text{Po}$ radionuclide emissions from local power plants in central Poland

Magdalena Długosz-Lisiecka \* and Damian Perka

Due to the more volatile nature of  $^{210}\text{Po}$  in relation to  $^{210}\text{Pb}$ , an imbalance of activity concentrations in high temperature combustion processes can be observed, especially in fine particulates (diameter < 0.1, 0.2 and 0.5  $\mu\text{m}$ ). In the atmosphere and in the soil around coal-fired power plants,  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  concentrations are a combination of activities from natural and anthropogenic sources. In this study only portions of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  radionuclides resulting from energy production activities were analyzed. Due to the high mobility of fine particles, a surface area of 172 km  $\times$  140 km in central Poland was chosen for simulation. For validation of the modeling approach, three grid versions were applied: 1 km, 2 km and 4 km. Simulated results confirmed experimental–computational values of an excess of both radionuclides in the atmosphere in 2017 in the city of Lodz. Different aerosol fractions, seasons and various grids in the selected area were subjected to 36 individual simulations. The  $^{210}\text{Po}$  activity concentrations measured in winter and summer 2017 were 42.5 and 8.99  $\mu\text{Bq m}^{-3}$ , respectively. Simulated and measured values of artificial  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities are well correlated.

Received 29th March 2020

Accepted 29th September 2020

DOI: 10.1039/d0em00141d

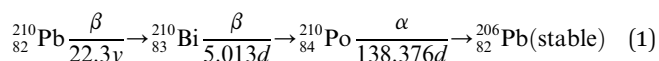
rsc.li/epi

### Environmental significance

Fossil fuel power plants emit significant portions of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  radionuclides. Fine aerosols ( $d < 0.1 \mu\text{m}$ ) contain the highest concentration of both radionuclides. The CALPUFF/CALMET modeling system is a useful tool for air pollution prediction. In central Poland a half of total  $^{210}\text{Po}$  activity and only ~5% of  $^{210}\text{Pb}$  activity in the atmosphere result from local power supply activities. Comparison of simulation data with data from stations can have a considerable impact on the validation and development of dispersion models. From the images received, it can be stated that in winter, areas exposed for additional portions of radionuclides are located around emitters.

## Introduction

The major source of  $^{210}\text{Pb}$  radionuclides in the atmosphere is  $^{222}\text{Rn}$  exhalation from the ground. Radionuclide  $^{210}\text{Po}$  arises from  $^{210}\text{Pb}$  as a result of two successive beta decays (eqn (1)):



$^{210}\text{Pb}$  and  $^{210}\text{Po}$  are radionuclides with both radioactive  $\beta$  and  $\alpha$  emitters, respectively. Inhalation can be harmful to human health; however, the inhalation dose depends on the size distribution of the aerosols. In several studies  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  radionuclides are effective indicators of atmospheric dynamic processes like removal or washout ratios,<sup>1–3</sup> residence time<sup>4–7</sup> and exchange in air masses within or between the troposphere and stratosphere.<sup>8</sup> In recent years both isotopes were applied for tracing airborne pollutants and have been widely used in investigating their dispersion in urban air.<sup>9–13</sup> Numerical modeling is a new investigative approach for

determining the distribution of the tracer over long distances, even hundreds of kilometers. The National Atomic Law (November 29, 2000, *Journal of Law* 2018, item 792) defines and classifies levels of activity of radioactive sources. Natural isotopes like  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  are classified in the first group of the most toxic elements, exactly like the isotopes of heavy elements with the U-type nuclear designation. In addition, the content of  $^{210}\text{Pb}$  together with derivatives at the level of total activity 10 kBq or a concentration of 10 kBq  $\text{kg}^{-1}$  classifies a given material as a radioactive source. It turns out that reaching a concentration level of 10 kBq  $\text{kg}^{-1}$  for  $^{210}\text{Pb}$  fractionated dust is possible.<sup>14</sup> The results obtained in a previous study<sup>15</sup> indicate high  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  activity concentrations in aerosols collected next to the coal power plant located near the city center. In fine fraction of aerosols <0.58  $\mu\text{m}$ , the average activity concentration of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  can reach values of over 23 kBq  $\text{kg}^{-1}$  and 3.6 kBq  $\text{kg}^{-1}$ , respectively<sup>15</sup>

Polonium emitted from anthropogenic sources at a high temperature usually forms volatile compounds and quickly condenses into fine dust which easily enters the atmosphere despite the use of multilevel dust extraction systems. Point source emission from fossil fuel power plants has considerable input to the level of the  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  radionuclides in the

Lodz University of Technology, Faculty of Chemistry, Institute of Applied Radiation Chemistry, Wroblewskiego 15, 90-924 Lodz, Poland. E-mail: mdlugosz@mitr.p.lodz.pl



air.<sup>16,17</sup> Radioactive isotopes deposited on the surface of dust or ash can freely move long distances, increasing the radioactive background in the region of the emission source.<sup>5,9,14,18–20</sup>

Modeling and validation of radionuclide distribution results are indispensable stages for isotope concentration and exposure assessment, especially in cases of radiation events, e.g. nuclear power plant accidents and failures at fuel processing plants or radioactive waste storage facilities. The prediction of radionuclide distribution in the environment helps in decision-making when dealing with radiation incidents and ensuring radioactive protection of the local population (including total or temporary evacuation). The assessment of the concentration of radionuclides in the air in several selected fractions is an indispensable step for the assessment of inhalation doses for people living directly in the region of emission. Energy supplies based on coal or lignite power plants are a significant source of the gases and dusts emitted which contain volatile radioactive elements in varying concentrations throughout the year.<sup>9,19</sup> Several important radionuclides, especially those with high vapor pressure, leave the power plant chimneys very effectively even in systems equipped with improved filters or dedusting systems. The reduction of dust emissions, unfortunately, does not guarantee an effective reduction of emissions of volatile elements, including heavy metals, which often have the ability to concentrate on the smallest fractions with a diameter of the order of nanometers.<sup>6,7</sup> Several studies confirm the presence of radioactive <sup>210</sup>Po and <sup>210</sup>Pb contaminations in surface soil, mosses and lichens around fossil fuel power plants.<sup>18,21–23</sup>

Each element has different preferences in creating clusters or the rate of condensation on fine dust. However, a very strong relationship between the concentration of the radioactive element and the diameter (due to the specific surface area) of the aerosol fraction, to which the radionuclides attach, is observed. The determination of isotope concentrations in the smallest fractions of <1 μm seems to be of particular importance, as these are the sources of industrial human activity, especially energy generation. According to the estimation described in detail in previous studies,<sup>8</sup> in 2017, approximately 50% of the <sup>210</sup>Po and 5% of the <sup>210</sup>Pb activities contained in coal were not retained in the slag or ash caught by using electrofilters. Polonium and lead compounds partially leave the dust extraction system and are lifted *via* the smallest fly ash with a diameter of nanometers.<sup>24,25</sup> Fly ash, especially with the smallest fractions, is often transported many kilometers from the emission sources, causing an increase in the content of these isotopes in relation to <sup>210</sup>Po and <sup>210</sup>Pb in urban air. Several studies based on radioecological monitoring methods applied to power plants in the region confirm a higher content of volatile compounds and radionuclides in the environment.<sup>7,18,21,24</sup> There are limited available data on radionuclide transport in the size distribution of the radioactive particles and their interaction with natural aerosols in the troposphere.

Previous studies on the distribution of <sup>210</sup>Pb and <sup>210</sup>Po activity in fractionated dust confirmed that concentrations of 10 kBq kg<sup>-1</sup> or 10 Bq g<sup>-1</sup> are already obtained for the fraction of *d* < 1 μm aerodynamic diameter.<sup>6,7,15</sup> This study focuses on materials of natural origin that emit ionizing radiation, so-called

NORM (Naturally Occurring Radioactive Materials), which is concentrated by human activity, but the physicochemical processes in the atmosphere magnify the problem, and these cannot be limited. Therefore, it would be highly recommended to monitor the radiation situation around industrial plants, foundries or heat and power plants, where natural radionuclides such as volatile elements can be freely emitted along with off-gases. In addition, similar to the environmental regulations limiting the content of harmful substances type B(a)P, nitrogen oxides and sulfur, or dust, there should be provisions limiting the content of radioactive elements in the atmosphere or limitations relating to their emission.

Fossil fuels account for about 50.5% of the energy supply in Poland, compared to their average energy mix of 16.2% in the EU.<sup>26</sup> The lack of appropriate regulations limiting the content of natural radioactive elements around industrial complexes does not exempt the need to monitor the radiation situation, for example around the largest heat and power plants in Poland. The most important factors in simulating the spread of pollutants in the atmosphere, including radioactive elements, are meteorological conditions, which are assigned to each field in the computational grid, *i.e.* the direction and wind speed, the height of the mixing layer and the class of the atmosphere balance. There is a large number of models to simulate the air pollution propagation.

The CALMET/CALPUFF tool model advanced non-steady-state meteorological systems for predicting air quality and simulate observations.<sup>27</sup> A new version of these tools allows for particulate matter and long-range transport of selected radionuclides and their decay products in several aerosol fractions to be assessed.

Mathematical modeling allows additional radionuclide activity in the air to be estimated, and therefore it will estimate possible additional radiological exposure to the local population.<sup>25,28,29</sup>

The aim of this work is to model the propagation of radionuclides from point and surface sources over central Poland. The simulation method allows reconstruction of the anthropogenic emission of <sup>210</sup>Po and <sup>210</sup>Pb radionuclides from various heating systems, taking into account meteorological conditions and landforms.

## Materials and methods

To simulate the propagation of radionuclides, we applied a CALPUFF atmospheric dispersion model with meteorological data derived from an external CALMET model to a set of three power plants in central Poland to evaluate <sup>210</sup>Pb and <sup>210</sup>Po isotope impacts. This set is recommended by the U.S. EPA. There is a lack of similar studies for these isotopes. However, in separate studies, the CALPUFF model was applied to a set of nine power plants in Illinois to evaluate the impact of primary and secondary particulate matter<sup>30</sup> and the specific case of the <sup>137</sup>Cs isotope released in the atmosphere during the 1986 Chernobyl Nuclear Power Plant accident.<sup>31</sup>

Modeling of the spread of pollutants in urban air (Lodz metropolitan area) was conducted using an advanced multi-layer



cloud CALPUFF Gaussian model, which has algorithms including diversity of terrain. For modeling the spread of contamination in the urban atmosphere, the CALPUFF pollutant dispersion model was used, including the CALMET meteorological model. To run a set of models, they were obtained through several preprocessors that process data from public databases.

The output obtained from the CALMET meteorological model was implemented as an input file for the CALPUFF model in version 7.2.1 (version form 2019/01/04), which simulate release of radionuclides and its decay products.

In addition, a method for extracting land cover data and conversion has been developed to a format compatible with the preprocessor using a spreadsheet. As part of the work, a statistical analysis of selected calculation grids: 1 km, 2 km and 4 km, was made. In this study an area (size: 172 km × 140 km) in central Poland, including Lodz and Belchatow metropolitan areas, was analyzed. The computational grid was chosen to cover most of the territory of the Lodz Voivodeship.

Estimated data of emissions were included in Długosz-Lisiecka<sup>7</sup> which amount to 6.28 and 0.59 GBq per year respectively for <sup>210</sup>Po and <sup>210</sup>Pb radionuclides emitted by two similar coal power plants located in the city of Lodz. A lignite power plant located in Belchatow emitted 171.51 and 62.82 GBq per year for <sup>210</sup>Po and <sup>210</sup>Pb, respectively. In the assessment, modeling has also been applied to about 20 000 domestic heating systems which are a surface source of emission for the Lodz metropolitan area. Each year about 2.76 and 0.47 GBq of <sup>210</sup>Po and <sup>210</sup>Pb respectively were emitted from this source. Comparing power plants and domestic heating systems we can show differences in chimney height used by those examples and the diameter of emitted dust. The modeled dust fractions are: 0.1 μm, 0.2 μm and 0.5 μm, which together account for about 72% of total emissions. In contrast, the smallest fraction modeled is about 40% of the emitted radionuclides. The assumed weights of specific fractions are 40%, 22% and 10% for fraction 0.1 μm, 0.2 μm and 0.5 μm, respectively. Distribution of radionuclides depends on local and domestic heating activities, which vary according to the season. The results of modeled <sup>210</sup>Pb and <sup>210</sup>Po activity concentration are provided in μBq m<sup>-3</sup>.

The selected time period was a week, similar to the period of dust collection by Air Aerosol Sampling Stations (ASSs) working within the national program of monitoring radionuclides in the atmosphere. In this study, three points and one area source were selected as sources of radionuclide emission that have the most impact on emission rates in Lodz Voivodeship: two coal-fired power plants located in Lodz, one lignite power plant located in Belchatow and approx. 20 000 domestic heating systems within the Lodz metropolitan area. One coal-fired power plant is in the north-western part of the city, the second is in the central-eastern part, and the lignite power plant is located about 60 km away, in the south of Poland. The location of the three fossil fuel plants has a notable impact on the air quality in central Poland.

## Results

The model manufacturer, Exponent, on the website <http://www.src.com>, shows links to compatible databases with

preprocessors, but most of them have upgraded versions which are not specified. The calculation area around the emitters should include at least 50 times the height of the emitter according to the general guidelines for mathematical modeling, therefore an area of 172 km × 140 km was chosen. The choice of grid was because the city of Lodz is one of the largest emitters of pollution in the Lodz Voivodeship, and a substantial one also in the surrounding areas. In this study a rectangular grid, with areas of 1 km, 2 km or 4 km, was selected. The model used in this work is complex enough to require delivery information about the release for more accurate result simulations close to the values observed in reality. There are several databases providing such information: GTOPO30 and GMTED2010 from 2010, containing more information or SRTM3 in a format compatible with preprocessors TERREL and MAKEGEO. The classification of land cover on the given computational grids was obtained from the Corine Land Cover (CLC) base with a resolution of about 250 m. The database covers the countries of the European Union.

To implement the simulation, it was also necessary to determine the meteorological conditions on the given computational grids, originating from ground and aerological stations. For this purpose, SMERGE and READ62 preprocessors were used to prepare data for a compatible format with a meteorological CALMET model.<sup>32</sup> Data from ground stations came from the main station from the National Oceanic & Atmospheric Administration National Climatic Data Center database (NOAA NCDC) which include wind speed and direction, air temperature, ceiling height, cloud cover, surface pressure, relative humidity and precipitation type code. Data from aerological stations came mainly from National Oceanic & Atmospheric Administration Earth System Research Laboratory (NOAA ESRL).

**Table 1** Wind direction for four seasons (a) spring (b) summer, (c) autumn and (d) winter

(a) Wind-direction-spring		(b) Wind-direction-summer	
N	6%	N	6%
NE	5%	NE	7%
E	23%	E	19%
SE	9%	SE	7%
S	9%	S	6%
SW	23%	SW	14%
W	23%	W	29%
NW	3%	NW	11%
(c) Wind-direction-autumn		(d) Wind-direction-winter	
N	6%	N	0%
NE	4%	NE	1%
E	23%	E	10%
SE	7%	SE	9%
S	7%	S	14%
SW	18%	SW	25%
W	28%	W	37%
NW	7%	NW	3%



The data collected reflected the meteorological conditions measured by the Lodz station in 2017, from the free database of Weather Online Meteorological Services (Table 1).

Based on the numerical geophysical data computing area and atmospheric conditions, calculations of meteorological parameters were performed to define the grid, as the input data for the CALPUFF model of version 7.2.1. CALMET creates a three-dimensional mesh, where the height of layers and their number can be modified by the user. The frequency of winds in particular directions formed the basis on which wind roses were developed for the four seasons. The ground layers of the winds are analyzed because they are subject to the largest fluctuations and most often result in scattered results. However, the upper layers are characterized by the uniformity of meteorological conditions. Table 1 shows data containing average wind directions for four seasons.

Radioactive emission rates of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  radionuclides were estimated for 2017 based on data included in Długosz-Lisiecka,<sup>7</sup> in GBq per year (Table 2). In 2013, about 10.9 GBq and 0.9 GBq of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  radionuclides were possibly released into the atmosphere in Lodz city center from local coal power plants.

The calculation was done based on the amount of combusted coal, slag and its radioactive analysis. For 2017, the estimation of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  isotope emissions was based on the reduction of  $\text{SO}_2$  and  $\text{PM}_{10}$  emissions, respectively. A previous study<sup>8</sup> confirms the considerable linear correlation between the unsupported  $^{210}\text{Po}$  activity concentration and  $\text{SO}_2$  emission from energetic coal combustion.

Modeled dust fractions are: 0.1  $\mu\text{m}$ , 0.2  $\mu\text{m}$  and 0.5  $\mu\text{m}$ , which together account for about 72% of the total emitted particles. This value is the average from previous experiments. With this approach, the weights of emission for each season were assumed to be equal to 50%, 20%, 10% and 20% of the total activity (for both isotopes) for winter, spring, summer and autumn respectively. This assumption was established by the energy consumption needed for heating by season.

The simulation of radionuclide propagation in various dust fractions was performed using the CALPUFF model, while the results were isolated using the CALPOST preprocessor, which allows the specification of the average or maximum values in a given interval time.

Fig. 1 shows the chosen distribution of the  $^{210}\text{Po}$  radionuclide concentration in the 0.1  $\mu\text{m}$  fraction in different seasons in a grid of 4 km.

Fig. 1 mainly includes the 0.1  $\mu\text{m}$  fraction containing  $^{210}\text{Po}$ , because other particles have less effect on living/hazardous

organisms than the particle size spread shown. It is also the most affected by meteorological conditions, because it depends on the size and weight of particles; the lower the size and weight, the further the transport range. Therefore, seasonal variation of emission is mainly caused by industrial production of electricity, which is variable in Poland by season. The direction of plumes depends mainly on wind direction and speed or land diversity. The biggest impact on emitted dust is power plants. Power plants have the biggest impact on emitted dust on a large area, but domestic heating systems pollute locally.

The modeled results have been compared with experimental data obtained from the Aerosol Sampling Station ASS-500 (Lodz city, 51°47' N and 19°28' E) during all seasons in 2017. The average value of measured activity concentration for the  $^{210}\text{Po}$  isotope in 2017 was equal to 24.9  $\mu\text{Bq m}^{-3}$  (range of the value was from 7.8 to 42.5  $\mu\text{Bq m}^{-3}$ ), 12.9, 3.6 and 11.1  $\mu\text{Bq m}^{-3}$ , respectively for winter, spring, summer and autumn.

In this study,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity concentrations have been modeled for 0.1, 0.2 and 0.5  $\mu\text{m}$  aerosol fractions, depending on the grid size, e.g. 1 km, 2 km and 4 km and for different seasons. In practice, 36 individual simulations were done. The modeled results of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activity concentrations confirm the measured values. The maximum values of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities for the 0.1  $\mu\text{m}$  fraction (Fig. 1) obtained in winter, for the region close to the emission sites, can reach over 50 and about 10  $\mu\text{Bq m}^{-3}$ , respectively. Artificial  $^{210}\text{Po}$  activity concentrations in various fractions are over two times higher than those of the  $^{210}\text{Pb}$  isotope. The activity concentration of the  $^{210}\text{Po}$  isotope in the 0.1  $\mu\text{m}$  fraction can even reach a four times higher value compared to the 0.5  $\mu\text{m}$  fraction (Table 3). The simulated  $^{210}\text{Po}$  activity concentration in total dust in summer is 24% of the winter result. However, in the experimental approach, the summer  $^{210}\text{Po}$  activity concentration is 14% of the winter result. The difference may result from the fraction shares in different seasons and the different inflow of artificial  $^{210}\text{Po}$  from different emission sources.

Analysis of meteorological conditions showed that the occurrence of southern winds was dominant for each of the given resolutions and seasons, which is not conclusive with the direction of the dispersion of the streak, because the force of the wind also plays a substantial role. Meteorological conditions in the upper layers are usually more stable than in layers at ground level. Polonium radioactive decay generates several orders of magnitude lower, subject to, among other factors, dilution of pollutants in the air, without causing any visible differences in streaked images.

Table 2 Estimated values of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  radionuclide emission rates in 2017

Point source emitters	Surface source		
	Lodz city (GBq per year)	Belchatow city (GBq per year)	20 000 domestic heating systems (GBq per year)
Isotopes			
$^{210}\text{Po}$	6.28	171	2.76
$^{210}\text{Pb}$	0.59	62.8	1.47



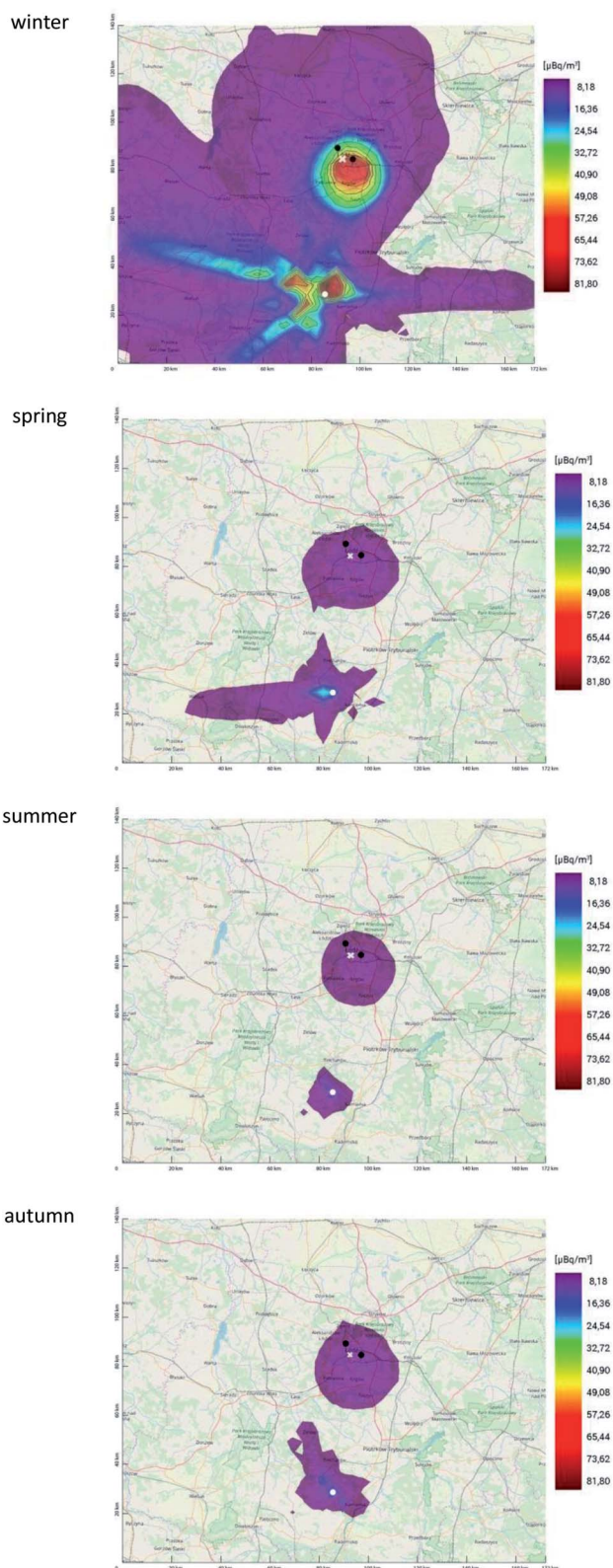


Fig. 1 Maximal  $^{210}\text{Po}$  activity distributions in the air [ $\mu\text{Bq m}^{-3}$ ] for 0.1  $\mu\text{m}$  fraction over central Poland obtained by simulation for a 4 km grid (white points represent power plants). Black circles—two similar coal power plants in Lodz city, white circle—lignite power plant in Belchatow city, and white cross—sampling point.

Table 3 Average concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in fractions [ $\mu\text{Bq m}^{-3}$ ]

	Winter	Summer	Autumn	Spring
<b>Average concentration of <math>^{210}\text{Po}</math> in fractions</b>				
0.5 $\mu\text{m}$	4.46	1.06	1.84	1.84
0.2 $\mu\text{m}$	9.76	2.33	4.04	4.04
0.1 $\mu\text{m}$	17.86	4.27	7.27	7.27
<b>Average concentration of <math>^{210}\text{Pb}</math> in fractions</b>				
0.5 $\mu\text{m}$	2.19	0.52	8.91	0.92
0.2 $\mu\text{m}$	4.80	1.14	1.95	2.03
0.1 $\mu\text{m}$	8.59	2.05	3.51	3.65

Average annual activity was calculated based on point and surface receptors introduced in the CALPUFF model using the BOOT software. Statistical indicators were compared to received values of the 1 km grid, because this computational grid is more precise than others. Validation procedures of dispersion models were determined (Table 4).

The values of average concentrations for the whole computational area, for each cell of the grid, are 4.94 and 2.31  $\mu\text{Bq m}^{-3}$  respectively, for unsupported  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities for all seasons in 2017. The average annual value of emissions based on point source and surface receptors is more consistent than an annual value based on activity in a given area (Table 4). Differences in average activity of radionuclides depend on the computational grid. A 1 km grid is more detailed than others, so it was selected to compare 2 km and 4 km grids, because in Lodz Voivodeship there are not enough ASSs to compare many records. The values of activity for 1 km and 4 km are similar, therefore, comparing the average activity of 1 km and 4 km to a 2 km computational grid of simulations shows a non-linear correlation between scale and activity. In fact, ASSs should be treated like point receptors, which could provide authoritative and repeatable results of radioactivity emitted by radionuclides, but using an average area value exposes a defect in the CALPUFF model: if the territory is larger, the radioactivity is lower. The average area value does not provide repeatable results, which cannot be considered correct, but values based on receptors can be seen dispersed in the air after collecting and analyzing the

Table 4 Selected statistical indicators calculated for discrete receptors located in the Lodz metropolitan area, depending on the computational grid (Rzeszutek, 2019)<sup>27</sup>

Grid area	1 km	2 km	4 km
Average activity of $^{210}\text{Po}$ [ $\mu\text{B m}^{-3}$ ]	4.80	5.17	4.86
Average activity of $^{210}\text{Pb}$ [ $\mu\text{Bq m}^{-3}$ ]	2.24	2.43	2.27
NMSE	0	0.01	0
R	1	1	1
FAC2	1	1	1
FB	0	-0.075	-0.013
FB <sub>fn</sub>	0	0	0
FB <sub>fp</sub>	0	0.075	0.013
MG	1	0.961	1.079
MG <sub>fn</sub>	1	1	1.079
MG <sub>fp</sub>	1	1.041	1



dust. The relative standard deviation (NMSE) for the grid areas of 1 km, 2 km and 4 km equals 0, 0.01 and 0, respectively. The linear correlation  $R$  takes the number one as the ideal value and is observed for all grid variants. The indicator characterizing the quality of the model (FAC2) shows that despite the use of different grids, the area remains constant and takes the value of one, so this means that more than half of the modeling results do not exceed double underestimation and overestimation. The standard deviation FB is within  $-0.075 \leq FB \leq 0$ , but FB is a component of  $FB_{fn}$  and  $FB_{fp}$ . The indicator  $FB_{fn}$  shows that with decreasing areas in the computational grid, underestimation of statistical error does not occur, while the  $FB_{fp}$  indicates a smaller overestimation of results with a non-linear decreasing grid area. The geometric mean MG is within  $0.961 \leq MG \leq 1.079$ , which theoretically indicates a relatively correct model. Simulation of a 4 km grid needs less time and is more economical than a 1 km grid. According to average activity calculated for discrete receptors, favorable options vary with a 4 km grid. The BOOT statistical model evaluation software package contains a technical and more detailed description of the indicators used.

## Conclusions

The content of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the ground-level atmosphere depends mainly on the  $^{222}\text{Rn}$  gas concentration emanating from the soil, but also on the share of anthropogenic sources. In aerosols, both isotopes never reach radioactive equilibrium. A simulation was carried out to obtain the distribution activity of artificial  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  occurring in central Poland. These simulated values were confirmed by using an experimental method. At a single receptor point located in Lodz, the average measured excess of  $^{210}\text{Po}$  was  $24.9 \mu\text{Bq m}^{-3}$  and  $3.6 \mu\text{Bq m}^{-3}$  respectively in winter and summer. The average (for all seasons in 2017) of the excess of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities was 49.5% and 4.99% relative to measured values for both isotopes. This means that half of the total  $^{210}\text{Po}$  activity and only ~5% of  $^{210}\text{Pb}$  activity in the atmosphere result from local power supply activities. Simulated and measured values of artificial  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities are well correlated. The maximal excess of the  $^{210}\text{Po}$  activity concentration measured in winter and summer 2017 was  $42.5$  and  $8.99 \mu\text{Bq m}^{-3}$ , respectively. The maximal simulated value of the  $^{210}\text{Po}$  activity concentration for the same seasons reaches ~50 and ~10  $\mu\text{Bq m}^{-3}$ , respectively (Fig. 1).

The results obtained relate only to the additional radioactivity associated with industrial emissions. The results, to a small degree, depended on the choice of the calculation grid and the existing differences were of a statistical nature. It is worth noting that the assumptions regarding the emission of individual dust fractions do not fully reflect real conditions, but an approximation. In this case, choosing a 4 km grid results in similar results to the more detailed 1 km grid. This is satisfactory because the difference in calculation time is remarkable. On the other hand, the estimated values of the average annual activity are part of the assumptions concerning the share of the combined heat and the power plant's observed activity of dust fractions, collected by ASSs,<sup>6,7,14</sup> which may indicate the

usefulness of the model in practice. Comparison of simulation data with data from stations can have a great impact on the validation and development of dispersion models. Grid resolution has no influence on dust dispersion, whereas the data extraction method has a direct impact on the estimation of the annual average value and on validating the model for targeted needs.

## Conflicts of interest

There are no conflicts to declare.

## Acknowledgements

This research work was supported by the National Science Centre under SONATA grant no. UMO-2012/07/D/ST10/02874.

## References

- 1 M. Baskaran, Po-210 and Pb-210 as atmospheric tracers and global atmospheric Pb-210 fallout: a review, *J. Environ. Radioact.*, 2011, **102**(5), 500–513, DOI: 10.1016/j.jenvrad.2010.10.007.
- 2 B. R. Persson and E. Holm, Polonium-210 and lead-210 in the terrestrial environment: a historical review, *J. Environ. Radioact.*, 2011, **102**(5), 420–429, DOI: 10.1016/j.jenvrad.2011.01.005.
- 3 A. Baeza, J. A. Corbacho, J. Guillén, A. Salas, J. C. Mora, B. Robles and D. Cancio, Enhancement of natural radionuclides in the surroundings of the four largest coal-fired power plants in Spain, *J. Environ. Monit.*, 2012, **14**, 1064–1072, DOI: 10.1039/C2EM10991C.
- 4 D. McNeary and M. Baskaran, Residence times and temporal variations of  $^{210}\text{Po}$  in aerosols and precipitation from southeastern Michigan, United States, *J. Geophys.*, 2007, **112**(D4), 1–11.
- 5 C. Papastefanou, Residence time of tropospheric aerosols in association with radioactive nuclides, *Appl. Radiat. Isot.*, 2006, **64**, 93–100.
- 6 M. Długosz-Lisiecka, Excess of Polonium-210 activity in the surface urban atmosphere. Part 1. Fluctuation of the  $^{210}\text{Po}$  excess in the air, *Environ. Sci.: Processes Impacts*, 2015, **17**(2), 458–464.
- 7 M. Długosz-Lisiecka, Excess of Polonium-210 activity in the surface urban atmosphere. Part 2. Origin of  $^{210}\text{Po}$  excess, *Environ. Sci.: Processes Impacts*, 2015, **17**(2), 465–470.
- 8 M. Długosz-Lisiecka, Chemometric methods for source apportionment of  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  for 10 years of urban air radioactivity monitoring in Lodz city, Poland, *Chemosphere*, 2019, **220**, 163–168.
- 9 A. Ugur, B. Özden and I. Filizok, Determination of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations in atmospheric deposition in Izmir (Aegean Sea-Turkey), *Atmos. Environ.*, 2011, **45**, 4809–4813.
- 10 K. Ram and M. M. Sarin, Atmospheric  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$  and  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratio in urban aerosols: temporal variability and impact of biomass burning emission, *Tellus B*, 2012, **64**, 17513.



- 11 Y. M. Amin, S. Uddin, M. Khandaker, A. K. Shyen, R. H. Mahat, R. M. Nor and D. A. Bradley, Radionuclide emissions from a coal-fired power plant, *Appl. Radiat. Isot.*, 2013, **80**, 109–116.
- 12 J. L. Guerrero, I. Gutiérrez-Álvarez, F. Mosqueda, M. JesúsGázquez and J. P. Bolívar, Evaluation of the radioactive pollution in the salt-marshes under a phosphogypsum stack system, *Environ. Pollut.*, 2020, **258**, 113729.
- 13 A. Boryło, G. Olszewski and B. S. kwarzec, A study on lead ( $^{210}\text{Pb}$ ) and polonium ( $^{210}\text{Po}$ ) contamination from phosphogypsum in the environment of Wiślinka (northern Poland), *Environ. Sci.: Processes Impacts*, 2013, **15**, 1622–1628, DOI: 10.1039/C3EM00118K.
- 14 M. Długosz-Lisiecka, The sources and fate of  $^{210}\text{Po}$  in the urban air: a review, *Environ. Int.*, 2016, **94**, 325–330.
- 15 M. Długosz, P. Grabowski and H. Bem, 2010.  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  radionuclides in the urban air of Lodz, Poland, *J. Radioanal. Nucl. Chem.*, 2010, **283**, 719–725, DOI: 10.1007/s10967-009-0407-x.
- 16 Ö. Kılıç, M. Belivermiş, E. Sıkdokur, *et al.*, Assessment of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  by moss biomonitoring technique in Thrace region of Turkey, *J. Radioanal. Nucl. Chem.*, 2019, **322**, 699–706, DOI: 10.1007/s10967-019-06721-4.
- 17 I. Filizok, A. Uğur and B. Ozden, Local Enhancement of  $^{210}\text{Po}$  Atmospheric Flux at a Site in Izmir, Turkey, *Water*, 2014, **225**, 1823, DOI: 10.1007/s11270-013-1823-7.
- 18 J. Suhana and M. Rashid, Naturally occurring radionuclides in particulate emission from a coal fired power plant: A potential contamination?, *J. Environ. Chem. Eng.*, 2016, **4**(4), 4904–4910.
- 19 M. S. Kh. Al-Masri, A. W. Haddad Doubal, I. Awad and Y. Al-Khatib, Assessment of soil contamination by  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  around heavy oil and natural gas fired power plants, *J. Environ. Radioact.*, 2014, **132**, 89–93.
- 20 G. Jia, G. Torri, D. Centioli and L. Magro, A radiological survey and the impact of the elevated concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  released from the iron- and steel-making plant ILVA Taranto (Italy) on the environment and the public, *Environ. Sci.: Processes Impacts*, 2013, **15**, 677–689, DOI: 10.1039/C2EM30784G.
- 21 M. Długosz-Lisiecka and J. Wróbel, Use of moss and lichen species to identify  $^{210}\text{Po}$  contaminated regions, *Environ. Sci.: Processes Impacts*, 2014, **16**(12), 2729–2733.
- 22 M. Długosz-Lisiecka, Kinetics of  $^{210}\text{Po}$  accumulation in moss body profiles, *Environ. Sci. Pollut. Res.*, 2017, **24**, 20254–20260, DOI: 10.1007/s11356-017-9659-0.
- 23 L. Al Attar, B. Safia and B. Ghani, Chemical composition of scales generated from oil industry and correlation to radionuclide contents and gamma-ray measurements of  $^{210}\text{Pb}$ , *Environ. Pollut.*, 2016, **210**, 246–252.
- 24 T. Vaasma, J. Loosaar, F. Gyakwaa, M. Kiisk, B. Ozden and A. H. Tkaczyk, Pb-210 and Po-210 atmospheric releases via flyash from oil shale-fired power plants, *Environ. Pollut.*, 2017, **222**, 210–218.
- 25 T. Vaasma, M. Kaasik, J. Loosaar, M. Kiisk and A. H. Tkaczyk, Long-term modelling of fly ash and radionuclide emissions as well as deposition fluxes due to the operation of large oil shale-fired power plants, *J. Environ. Radioact.*, 2017, **178–179**, 232–244.
- 26 Report, Energy Union Factsheet Poland, Brussels, 23.11.2017 SWD, 2017, 407.
- 27 M. Rzeszutek, Parameterization and evaluation of the CALMET/CALPUFF model system in near-field and complex terrain - Terrain data, grid resolution and terrain adjustment method, *Sci. Total Environ.*, 2019, **689**, 31–46.
- 28 A. Leelőssy, I. Lagzi, A. Kovács and R. Mészáros, A review of numerical models to predict the atmospheric dispersion of radionuclides, *J. Environ. Radioact.*, 2018, **182**, 20–33.
- 29 M. Jabłońska and J. Janeczek, Identification of industrial point sources of airborne dust particles in an urban environment by a combined mineralogical and meteorological analyses: A case study from the Upper Silesian conurbation, Poland, *Atmos. Pollut. Res.*, 2019, **10**(3), 980–988.
- 30 J. I. Levya, J. D. Spengler, D. Hlinka, D. Sullivan and D. Moon, Using CALPUFF to evaluate the impacts of power plant emissions in Illinois: model sensitivity and implications, *Atmos. Environ.*, 2002, **36**, 1063–1075.
- 31 D. Gaiotti, D. Oshurok and O. Skrynyk, The Chernobyl nuclear accident  $^{137}\text{Cs}$  cumulative depositions simulated by means of the CALMET/CALPUFF modelling system, *Atmos. Pollut. Res.*, 2018, **9**(3), 502–512.
- 32 D. Tartakovsky, D. M. Broday and E. Stern, Evaluation of AERMOD and CALPUFF for predicting ambient concentrations of total suspended particulate matter (TSP) emissions from a quarry in complex terrain, *Environ. Pollut.*, 2013, **179**, 138–145.

