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Radiological Hazard Related with Natural Radioactivity in Natural Gas Transportation—A Case Study from Poland

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Abstract: Activity concentration of ^{210}Pb in black powder and gamma radiation dose rate related to natural gas transportation were discussed. As part of the research, the content of radiolead (^{210}Pb) in black powder, spent filter containers, radon (^{222}Rn) activity concentration in natural gas and gamma radiation dose were measured around selected points of natural gas transportation infrastructure in Poland. The content of some heavy metals was also analyzed. The average concentrations of ^{222}Rn and ^{210}Pb ranged from 30 to 1400 Bq/m^3 and from around 450 to 16,000 Bq/kg, respectively. The external exposure to gamma radiation was low; the gamma radiation dose rate was at the level of background radiation or slightly exceeded it. The research demonstrated that the content of ^{210}Pb in black powder is strongly related to radon concentration in natural gas, the volumetric flow rate of the transported natural gas and solid fraction content. Some black powder samples should be classified based on ^{210}Pb content as low-radioactive waste.

Keywords: ^{210}Pb—radiolead; gamma radiation dose rate; natural gas; black powder

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Citation: Nowak, J.; Jodłowski, P.; Macuda, J.; Nguyen Dinh, C. Radiological Hazard Related with Natural Radioactivity in Natural Gas Transportation—A Case Study from Poland. *Minerals* **2022**, *12*, 662. https://doi.org/10.3390/min12060662

Academic Editor: Fernando P. Carvalho

Received: 14 April 2022
Accepted: 23 May 2022
Published: 24 May 2022

1. Introduction

In the mining industry, radiation hazard is associated with the occurrence of natural radionuclides and results mainly from the migration of ^{238}U and ^{232}Th decay products, along with a mixture of oil and natural gas extracted from the deposit. In the case of extraction and transportation of natural gas, radon and its decay products, mainly long-lived radioisotope ^{210}Pb ($T_{1/2}$ = 22.3 years) and ^{210}Po ($T_{1/2}$ = 138.4 days), are a significant problem. Radon decay products, due to their chemical properties, are easily adsorbed on various types of surfaces. As a consequence, a thin radioactive film is formed on the surfaces of the compressor cylinders and the walls of the gas pipeline. Furthermore, radon decay products along with aerosols easily hold onto filters, which in turn can lead to the accumulation of significant ^{210}Pb activity on them [1,2].

Previous studies on the subject of natural radioactivity related to natural gas transportation focus mainly on the level of radon activity concentration and correlation of ^{222}Rn content in natural gas with geological conditions. In turn, the studies on black powder focus mainly on its chemical composition and impact on the gas flow in pipeline. Research dealing with the problem of ^{210}Pb is sporadic. Furthermore, the research does not consider the aspect of radiation exposure. For example, studies carried out in other countries show that the concentration of radon in transported natural gas varies in a wide range from several dozen Bq/m^3 to several thousand Bq/m^3, and depends mainly on the proximity of gas mines and the geological structure of the deposit from which the gas is extracted. For instance, in the United Kingdom, the average radon concentration in gas pipelines is 170 Bq/m^3. In the states of Pennsylvania and New York (USA), the concentration of radon in the gas pipeline ranges from approx. 625 Bq/m^3 to over 1600 Bq/m^3. In natural gas from the Marcellus deposit, ^{222}Rn activity concentration ranges from around 40 Bq/m^3 to 2950 Bq/m^3 [3–5]. As a result of the presence of radon in natural gas, the transportation process can cause material known as black powder. The name 'black powder' derives from the color of the material, which changes from brown to black. Black powder is a residue of corrosion process of steel pipes and other steel elements of the pipeline network due to the presence of liquid aerosols, corrosive species (such as CO_2, H_2S, organic acids or O_2) and microorganisms. It can be wet or dry and consists of numbers of chemical forms of iron sulfides, iron oxides and iron carbonates. It is rich with radon progeny, especially radiolead (^{210}Pb) [6–11]. In Brazil ^{210}Pb content in black powder varied from 40 to 8800 Bq/kg. Additionally, Godoy et al. noticed correlation involving ^{210}Pb, zinc and barium [11]. In a recent publication Abbasi et al. [12] presented data on radon content in natural gas from different parts of the globe, taking into account current publications [13–15]. The activity concentration of ^{222}Rn in natural gas varied from 4 to around 53,000 Bq/m^3 [12]. Cowie and El-Sherik from the Saudi Aramco measured ^{210}Pb activity concentration in 65 samples of black powder. The radiolead content in the residue from pipeline ranged from around 15 to 7000 Bq/kg [16]. Due to the properties of the emitted gamma radiation (low-energy gamma radiation—46.5 keV) by the ^{210}Pb, the radiolead accumulated onto the filters is not 'visible' to the gamma radiation dosimeters, which work on the basis of a sodium iodide scintillation crystal or a Geiger counter. On the other hand, ^{210}Pb is well visible in the case of laboratory measurements using gamma spectrometry. Therefore, measuring only the gamma radiation dose rate may lead to erroneous conclusions about exposure to ionizing radiation.

The presence of black powder in the gas pipeline network may cause pressure and flow disturbances. Therefore, in order to ensure the proper functioning of the gas pipeline network, regular cleaning and inspection services known as pipeline pigging are carried out. During the services procedure, vast amount of black powder may be brought with the pig [7,8].

The Polish natural gas pipeline network is around 11,000 km long and it consists of 881 gas pipeline stations, 58 hubs and 14 compressor stations. Every year, around 14 billion cubic meters both of national and imported gas is transported. The black powder is present in the whole pipeline network. For example, the one compressor station can generate around 80 kg per year of black powder, which mainly holds onto filters. The pigging process along 50 km of the pipeline may bring even more than 600 kg of the material. Figure 1 shows the position of measurement points.

The scope of the presented research was selected in such a way that, in contrast to the previous studies, it would address the aspect of radioactivity in a comprehensive manner. In particular, the paper presents the results of measurements of gamma radiation dose rate around selected gas pipeline elements, radon activity concentration in natural gas and ^{210}Pb content in black powder taken from filter stations or immediately after the pigging process. Based on the obtained results, a transportation model of radiolead in the gas pipeline was created, binding the occurrence of ^{210}Pb with the average concentration of radon in the gas, the average concentration of particulate matter and the gas volumetric flow rate. A novel precise methodology of determination of radiolead content in black powder was used. Moreover, in the radon measurements, an important correction was introduced to take into account the increase in the efficiency of the measuring cell in the natural gas measurements. The findings were used to develop a novel methodology of determination of radiolead content in black powder in filter cartridges. In addition, an assessment of the potential risk associated with internal exposure to radiation resulting from direct contact with black powder was carried out, contrary to the previous research.

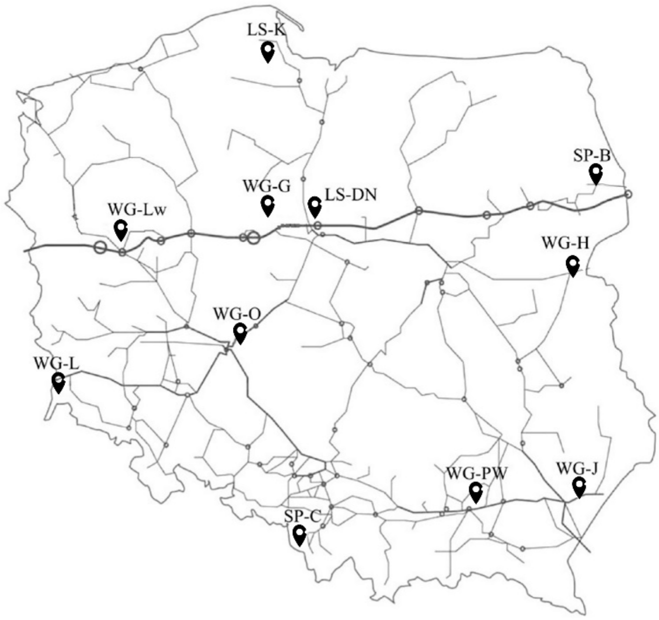

Figure 1. Location of measurement points (SP—a gas station; WG—a gas hub; LS—a launching station).

2. Materials and Methods

2.1. The Gamma Radiation Dose Rate

The gamma radiation dose rate was measured by the Ludlum Model 19 Micro R Meter portable dosimeter from Ludlum Measurements, Inc. The dose rate was performed at a distance of approx. 10 cm from the tested object (gas valves, batteries of filters, transceiver locks). The measurements were carried out during the "normal" operation of the transmission network, i.e., without pigging or replacing the filters, at points where there is the greatest probability of employees' presence and during the service procedure, i.e., the pigging process and the filters replacement at filter stations.

2.2. Radon

Gas samples for analysis were taken directly from sampling valves located at gas mainline into a 2.4 L gas cylinder using a set of high-pressure reducers. The cylinder was filled to a pressure of 10–15 bar. Then, a sample of gas through a set of low-pressure reducers was expanded to the Lucas cell (a Pylon type 300A cell was used). The gas with a volume of about 15 L flowed freely through the Lucas cell for 2–3 min. In order to achieve radioactive equilibrium between ^{222}Rn and short-lived decay products, the measurement was carried out after a minimum of 2 h after filling the chamber with a gas sample. The radon concentration in the gas samples was measured using the Pylon AB5 portable radiation monitor. A detailed description of the methodology was described by Nowak et al. [17].

2.3. ^{210}Pb

Black powder samples were taken from filters and immediately after the pigging process and placed into plastic containers. Then, in the Laboratory of Nuclear Spectrometry of The Faculty of Physics and Applied Computer Science AGH UST, the samples were sealed in cylindrical aluminum containers with a volume of 121 mL. The activity concentration of ^{210}Pb was measured by gamma radiation spectrometry using the HPGe model Canberra GX4020 germanium detector with a detection efficiency of 42%. A detailed description of the methodology was described by Jodłowski and Kalita [18] and Jodłowski [19].

3. Results

3.1. Chemical Composition—Content of Heavy Metals

Chemical analysis shows that iron is the main component of black powder in the gas pipeline network in Poland, and occurs in the form of iron(III) oxide. In addition, black powder was analyzed for the content of heavy metals chromium, cadmium, mercury, nickel and lead, as well as arsenic and barium. The results of the analyses are shown in Table 1.

Table 1. Concentration of selected heavy metals in black powder samples.

Location	Concentration of Heavy Metals [mg/(kg Dry Weight)]						
	As	**Ba**	**Cr**	**Cd**	**Hg**	**Ni**	**Pb**
[1] SP-B	11	140	304	<0.01	0.11	402	112
SP-C	12	220	442	<0.01	0.18	482	278
[2] WG-J	10	126	208	0.4	0.11	342	74
WG-L	18	146	506	<0.01	0.19	856	112
WG-O	11	170	98	1	0.24	88	38
[3] LS-DN	14	144	162	<0.01	0.42	932	388
LS-K	9	182	516	2	0.13	490	60
Limit value for the classification of waste as hazardous	25	300	70	5	2	40	50

[1] SP—a gas station; [2] WG—a gas hub; [3] LS—a launching station.

The content of heavy metals ranges from 98 to 516 mg/(kg dry weight), from 0.01 to 2 mg/(kg dry weight), from 0.11 to 0.42 mg/(kg dry weight), from 88 to 932 mg/(kg dry weight), from 38 to 388 mg/(kg dry weight), from 9 to 18 mg/(kg dry weight) and from 126 to 220 mg/(kg dry weight), respectively for Cr, Cd, Hg, Ni, Pb, As and Ba. Generally, black powder due to the content of heavy metals such as Cr, Ni and Pb should be treated as hazardous waste in accordance with national regulations. The Cr, Ni and Pb content greatly exceeds the limit value for the classification of waste as hazardous [20].

3.2. Activity Concentration of ^{222}Rn in Natural Gas

The average, minimum and maximum measured value of radon activity concentrations in natural gas from selected locations of the gas pipeline network are presented in Table 2.

Table 2. The ^{222}Rn activity concentration in natural gas and the ^{210}Pb activity concentration in black powder.

Location	The Activity Concentration of ^{222}Rn [Bq/m^3]			^{210}Pb [Bq/kg]
	Min.	**Max.**	**Average**	
SP-B	46.9 (4.1) [1]	64.6 (5.6)	56.4 (4.2)	445 (50)
SP-C	<30 [2]	<30	<30	5600 (600)
WG-J(1)	278 (25)	425 (31)	361 (26)	12,500 (1300)
WG-J(2)	-	-	-	16,000 (1500)
WG-L	<30	69.5 (6.1)	55.0 (8.0)	2100 (200)
WG-O(1)	<30	<30	<30	645 (60)
WG-O(2)	-	-	1360 (120)	-
WG-Lw	55.2 (5.2)	79.9 (7.1)	65.3 (6.5)	-
WG-G	63.0 (6.0)	97.1 (8.7)	77.5 (7.8)	-
WG-H	53.2 (4.2)	104 (10)	80 (10)	-
LS-DN (MFL pig)	-	-	-	15,700 (1600)
LS-K (brush cleaning pig)	-	-	-	3300 (350)
LS-K (MFL pig)	-	-	-	2620 (250)

[1] The standard uncertainty was given in the brackets; [2] the detection limit of applied method.

The average activity concentration of ^{222}Rn in natural gas varies, ranging from 30 Bq/m^3 (method detection limit) to approx. 363 Bq/m^3. For comparison, the level of radon concentration in residential buildings in Poland is around 150 Bq/m^3 [21]. Relatively low ^{222}Rn content in natural gas is related to the fact that imported gas was present in almost all studied locations. For imported gas, the approximate time elapsed from the gas extraction to the collection of samples is several weeks. Therefore, as a result of radon decay, the ^{222}Rn activity concentration ($T_{1/2}$ = 3.8 days) in the gas decreased significantly during its transportation. At the location of WG-J, the average measured radon concentration is 363 Bq/m^3 and is much higher than the average radon concentrations for other points. This result is related to the fact that the measured gas in one-third was gas from local Polish mines. For the gas samples from WG-O(2), the activity concentration of ^{222}Rn around 1360 Bq/m^3 was measured. In the location WG-O(2), the natural gas came directly from a nearby gas mine.

3.3. Activity Concentration of ^{210}Pb in Black Powder

The activity concentration of ^{210}Pb in black powder samples both from the filter stations and taken after the pigging process is significant and ranges from 445 to 16,000 Bq/kg and from around 2600 to 15,700 Bq/kg, respectively (see Table 2). For comparison, the activity concentration of ^{210}Pb in the surface layer of soil (3 cm) in the vicinity of Krakow is around 100 Bq/kg [22]. Additionally, the highest content of ^{210}Pb in black powder is around twice higher than reported in Brazil and in the Kingdom of Saudi Arabia [11,16].

The highest concentration of ^{210}Pb (16,000 Bq/kg) was measured for the sample from WG-J(2), where transported gas was a blend of gas from nearby mines with imported one. A high activity concentration of ^{210}Pb about 5600 Bq/kg, was also measured for the sample from SP-C. In other locations, the ^{210}Pb activity concentration is much lower and varies in the range of around 450—2100 Bq/kg. The results show that the activity concentration of ^{210}Pb in black powder samples from filter stations is closely related to the total activity of ^{222}Rn that flows through the filter. The ^{210}Pb activity concentration in black powder exceeds the value at which, according to Polish law, waste is classified as radioactive waste [23].

3.4. Gamma Radiation Dose Rate

During normal operation of the gas pipeline for all measurement points, the gamma radiation dose rate emitted from selected elements of the gas pipeline was at the level of background radiation or slightly exceeded it. Background radiation at the analyzed points fluctuated in the range of 0.044–0.070 (see Table 3).

Table 3. Gamma radiation dose rate around selected gas pipeline elements.

Location	Gamma Radiation Dose Rate [μGy/h]			
	Background	A Pipeline Valve	A Filter Station	A Transmitting/ Receiving Lock
SP-B	0.044–0.061	0.044–0.061	0.044–0.061	0.044–0.061
SP-C	0.044–0.061	0.044–0.061	0.044–0.061	0.044–0.061
WG-J	0.044–0.061	0.052–0.070	0.052–0.070	0.052–0.070
WG-L	0.052–0.070	0.052–0.070	0.052–0.070	0.052–0.070
WG-O	0.044–0.061	0.044–0.061	0.044–0.061	0.044–0.061
WG-Lw	0.044–0.070	0.061–0.087	0.061–0.087	0.061–0.087
WG-G	0.044–0.061	0.044–0.061	0.044–0.061	0.044–0.061
WG-H	0.052–0.061	0.052–0.070	0.052–0.070	0.070–0.096

The results clearly indicate no risk associated with external exposure from gamma radiation emitted by selected elements of the natural gas pipeline.

For the service procedure related to the pigging process, in order to identify the radiation hazard, measurements were carried out around the transmitting and receiving locks, around the pig after removing it from the receiving lock and around the plastic cuvette with dust (black powder) removed from the pipeline as a result of the pigging process (measurements were carried out in two locations, LS-K and LS-DN). The measured dose around the transmitting locks is equal to background radiation, while the dose around the receiving locks upon arrival of the pig slightly exceeds the level of background radiation. It amounts to 0.087–0.20 μGy/h (with background radiation of 0.052–0.061 μGy/h) for the receiving lock in LS-K and is at the level of background radiation for LS-DN. The measured gamma radiation dose rate near the pig removed from the lock in the case of the brush cleaning pig was (LS-K) up to 0.21 μGy/h, and for the magnetic flux leakage (MFL) testing pig (LS-DN) was up to 0.1 μGy/h. The measured dose over the dust placed in the plastic cuvette ranged from 0.13 μGy/h to 0.87 μGy/h (see Table 4).

Table 4. Gamma radiation dose rate during the maintenance process of gas pipeline (the pigging).

Location	Description of the Measurement Point	Gamma Radiation Dose Rate [μGy/h]
LS-K (brush-cleaning pig)	Background (transmitting lock)	0.087–0.096
	The pig (before pigging process)	0.070–0.078
	Background (receiving lock)	0.052–0.061
	Receiving lock after the pig's arrival	0.087–0.21
	The pig (after pigging process)	0.087–0.20
	Black powder in cuvette	Up to 0.87
LS-K (MFL pig)	Background (transmitting lock)	0.087–0.096
	The pig (before pigging process)	0.061–0.070
	Background (receiving lock)	0.052–0.061
	Receiving lock after the pig's arrival	0.052–0.061
	The pig (after pigging process)	0.052–0.070
	Black powder in cuvette	0.13–0.15
LS-DN (MLF pig)	Background (transmitting lock)	0.052–0.070
	The pig (before pigging process)	0.052–0.070
	Background (receiving lock)	0.052–0.061
	Receiving lock after the pig's arrival	0.052–0.070
	The pig (after pigging process)	0.072–0.10
	Black powder in cuvette	Up to 0.35

4. Discussion

4.1. Gamma Radiation Dose Rate—External Exposure

The radiological risk associated with external exposure is mainly related to short-lived radon decay products and, as measurements show, is minor. Normal operation of the gas pipeline does not pose a health risk to workers from a radiation protection point of view. In the case of the pigging process, the measured gamma radiation dose rate exceeds the level of background radiation, but the exceedances do not cause significant exposure of workers to ionizing radiation. Additionally, in case of external exposure around the plastic cuvette with black powder, the study of gamma radiation dose rate changes over time was carried out. The results of the measurements are shown in Figure 2.

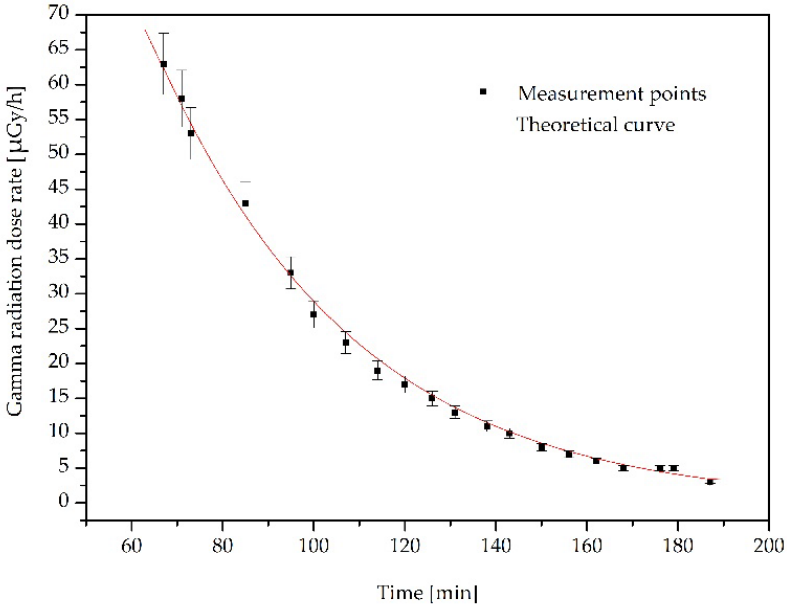

Figure 2. Gamma radiation dose rate as a function of time.

Assuming radioactive equilibrium between short-lived radon decay products at the time of the pig arrival, a theoretical time-dependence curve of the gamma dose rate obtained on the basis of the Bateman equation was also plotted. The obtained half-life time of the gamma radiation dose rate is about 27 min, which means that after about 2 h from the arrival of the pig to the receiving lock, the dose rate decreases about 20 times.

4.2. Gamma Radiation Dose Rate—Internal Exposure

^{210}Pb content in black powder can reach significant amounts, contributing to the increased internal exposure of employees during the service procedure, i.e., the pigging process and the filters replacement at the filter station. The activity concentrations of other radionuclides are very low, much lower than, for example, in soil. The concentration of ^{40}K, ^{226}Ra and ^{232}Th is, respectively about 10 Bq/kg, below 5 Bq/kg and below 5 Bq/kg. The average activity concentration of these radionuclides in the 0–10 cm surface layer of soil in Poland amounts to 415, 25 and 24 Bq/kg, respectively [22]. Therefore, only ^{210}Pb and its decay products (^{210}Bi and ^{210}Po) were considered to estimate the committed effective dose resulting from the intake of black powder.

The committed effective dose (E(τ)) resulting from the intake of radionuclides occurring in black powder can be estimated by the following equation:

$$\mathrm{E}(\tau) = \mathrm{mA_i e(g)_i} \quad (1)$$

where: m—is a mass of black powder, A_i is the activity concentration of the given radionuclide in black powder expressed in Bq/kg and $e(g)_i$ stands for the dose in Sv resulting from the ingestion of 1 Bq of the given radionuclide [24,25]. Assuming radioactive equilibrium between ^{210}Pb, ^{210}Bi and ^{210}Po, the committed effective dose resulting from intake of 1 g of black powder with the highest ^{210}Pb activity concentration of 16,000 Bq/kg is around 14.7 μSv. The estimated dose is around 1.5% of the annual dose limit to non-occupational workers and members of the public (1 mSv).

4.3. Transport Model of ^{210}Pb in the Gas Pipeline

In atmospheric air, short-lived radon decay products behave like ions with high mobility, and easily adsorb on various types of aerosols [26,27]. In the case of gas transport by gas pipeline, similar behavior can be noted, which consequently leads to the accumulation of ^{210}Pb on aerosols and, finally, black powder. The analyses carried out for two gas pipeline lines from WG-J and WG-L show the relationship between the average ^{222}Rn activity concentration (C_{Rn}) in the transported natural gas, the average content of particulate matter in the gas (k), the gas volumetric flow rate (Q) and the activity concentration of ^{210}Pb in the dust (black powder) accumulated on the filters in the filter station. This suggests the following model of transport of ^{210}Pb in the gas pipeline between two filter stations: 1. Radioactive decay of radon and formation of short-lived decay products; 2. adsorption of short-lived radon decay products on particulate matter resulting from pipeline pipe corrosion and carried with gas stream, or adsorption on the walls of the pipeline pipe; 3. accumulation of solid fraction along with ^{210}Pb on filters. Using the radioactive decay law, the number of nuclei of a radioactive isotope (N) can be determined according to the following formula:

$$N = N_0 e^{(-\lambda \cdot t)} \tag{2}$$

where: N_0—value of N at time t = 0, λ—decay constant [1/s], t—is time [s].

Therefore, the number of decayed nuclei (N_D) can be determined by the following equation:

$$N_D = N_0 - N = N(e^{(\lambda t)} - 1) \tag{3}$$

Based on the average radon activity concentration and the radioactive decay law (Equation 2), the average amount of radon decay products (and thus ^{210}Pb) per cubic meter of gas formed during gas transport from one filter station to another one can be determined using the equation:

$$N_{prod} = \frac{C_{Rn}}{\lambda_{Rn}} (e^{(\lambda_{Rn} \cdot \frac{LA}{Q})} - 1) \tag{4}$$

where: N_{prod}—number of radon decay products formed in 1 m^3 of gas, C_{Rn}—average ^{222}Rn activity concentration [Bq/m^3], λ_{Rn}—radon decay constant [1/s], L—distance between two filter stations [m], A—pipeline cross-sectional area [m^2], Q—average gas volumetric flow rate [m^3/s].

Taking into account the content of particulate matter in 1 m^3 of gas (k) the activity concentration of ^{210}Pb in black powder (A_{Pb}) can be determined by the following formula:

$$A_{Pb} = N_{prod} \frac{\lambda_{Pb}}{k} \tag{5}$$

where: λ_{Pb}—^{210}Pb decay constant [1/s].

Based on the proposed model and parameters of gas pipeline the activity concentration of ^{210}Pb in black powder was calculated. The calculated and measured activity concentration of ^{210}Pb are the same within range of measurement uncertainties. Table 5 presents the results of calculations for the two analyzed gas pipeline lines, along with their parameters.

Table 5. The calculated and measured activity concentration of ^{210}Pb.

Data Description	**Location**	
	WG-J(1)	**WG-L**
Diameter of the gas pipeline [mm]	700	700
Length of the gas pipeline [km]	145	150
Average content of particulate matter in gas [mg/m^3]	0.28–0.42	0.20–0.30
Average gas volumetric flow rate [m^3/s]	4.2–5.6	5.6–8.7
Average ^{222}Rn activity concentration [Bq/m^3]	361	55
Calculated ^{210}Pb activity concentration [kBq/kg]	11.5–12.9	1.9–2.4
Measured ^{210}Pb Activity concentration [kBq/kg]	12.8(1.3) *	2.10(0.20)

* The standard uncertainty was given in the brackets.

5. Conclusions

The primary source of radiological hazard related to natural gas transportation by gas pipeline is radon presented in the transported gas. Radon itself does not pose a threat, but its decay products, primarily Pb-210 and temporarily Pb-214 and Bi-214. These products are easily adsorbed on various types of aerosols or on pipeline elements' surfaces, causing significant radioactivity of black powder. The presented research shows that the real radiological hazard is related to internal exposure to radiation during the service procedure as pigging process or replacement of filter cartridges in filter station. Additionally, black powder with the highest activity concentration of ^{210}Pb concentration should be classified as low-radioactive waste. The gamma radiation dose rate around natural gas pipeline infrastructure objects, such as valves, filter stations, and transmitting/receiving locks is at the level of background radiation. These objects do not pose a radiological hazard. A novel methodology of determination of ^{210}Pb activity concentration in black powder, linking the average ^{222}Rn content in the transported natural gas, the average content of particulate matter in the gas and gas volumetric flow rate with the content of radiolead in black powder, was developed. This in situ method allows determination of the ^{210}Pb content in the residues from pipelines based on the measurement of the average radon concentration in the natural gas in the pipeline.

6. Patents

The result of the research is the patent: PL 237,746 B1—"Method of determination of radioactive lead Pb−210 activity concentration in filter cartridges, in particular in gas filter-separator stations and the system of devices for determining activity concentration of radioactive lead Pb−210 in filter cartridges, in particular in gas filter-separator stations".

Author Contributions: Conceptualization, J.N. and P.J.; methodology, J.N. and P.J.; formal analysis, J.N., P.J., C.N.D. and J.M.; investigation, J.N. and P.J.; resources, J.N., P.J., C.N.D. and J.M.; writing—original draft preparation, J.N., P.J., C.N.D. and J.M.; writing—review and editing, J.N., P.J., C.N.D. and J.M.; visualization, J.N. and P.J.; supervision, J.N. and P.J. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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