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Indoor Air Quality in a Selected Health Resort Facility: Analysis of PM_{10}, $PM_{2.5}$ and ^{222}Rn Concentrations

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ABSTRACT

This paper discusses the importance of indoor air quality and indicates its relation with outdoor air quality in the area of spa treatment. Significant factors and sources causing indoor air pollution by particulate matter and radon are considered. Particular attention is drawn to specific functions of health resorts in spa treatment in the context of the importance of indoor and outdoor air for patients staying in the resorts. It is underlined the specificity of health resorts providing treatment for patients classified as the group most sensitive to air pollution, i.e. people with chronic respiratory diseases. The study comprised measurements of 24-h concentrations of PM_{10}, $PM_{2.5}$ and 10-minute instantaneous concentrations of radon-222 (^{222}Rn) from February 5 to February 25, 2021 in a treatment room of one of the Polish spas located in Lower Silesia. The analysis of interactions between the concentrations of two fractions of particulates PM_{10} and $PM_{2.5}$ was presented. The interaction and dependence of factors affecting the elevated values of concentrations of the studied pollutants were discussed. The presented analyses showed that the concentrations of the studied pollutants were influenced by air exchange and air infiltration from outside to inside. In the case of short-term measurements of ^{222}Rn activity concentration in the air, it was found that the observed changes in hourly concentrations of ^{222}Rn are analogous to those observed in residential buildings. For the specific time intervals, the variation of PM concentrations and ^{222}Rn activity concentration was found to be similar.

Keywords: health resort, indoor air, particulate matter (PM), ^{222}Rn.

INTRODUCTION

Indoor air pollution: particulate matter and radon

Air is a very important component of the environment, including a key role in a spa treatment [Kobus et al., 2020]. Indoor air is affected by pollutants from indoor as well as outdoor sources, such as combustion of solid fuels, fuels derived from oil and gas, emissions from transportation, emissions from decorative materials, and pollutants associated with various activities, such as cooking, cleaning, and biological emissions that emit PM (Sun et al., 2022). Pollutants in indoor air include biological pollutants (e.g. mold, Legionella pneumophila bacteria) from humidifiers and air conditioning systems, as well as tobacco smoke, pesticides and household products (e.g. detergents, air fresheners), gases (e.g. radon and carbon monoxide), particulates, materials used for construction (e.g. asbestos, lead, formaldehyde) [EPA, 2022].

According to WHO, particulate matter is one of the most harmful air pollutants (WHO, 2013). They differ not only in origin but also in their physicochemical properties, effects on human health, environment and materials. The magnitude and chemical composition of particulate matter depend on many factors: geographic location, terrain and land cover, meteorological conditions, and the time of year or day of the week.

Particulate matter, small (PM_{10}) and tiny ($PM_{2.5}$), is generated by both natural and anthropogenic origins. They are particularly prevalent in large human hubs, such as urban agglomerations [Cichowicz et al., 2020]. In addition to the combustion of solid fuels for heating purposes, another important source of PM formation is transportation, where particulate matter is produced by the combustion of fuels in internal combustion engines, as well as by abrasion of vehicle components and roadway surfaces [CIEP, 2021].

The outdoor PM concentration and the air tightness of the building have a large influence on the indoor PM concentration due to the possibility of pollutant transport from the outside to the inside [Poupard et al., 2005]. The correlation between indoor to outdoor PM pollution largely depends on ventilation efficiency, in addition to the persistence and chemical transformations of particulate pollutants [Liang, 2013]. Indoor sources of PM are determined by the type and manner of specialized activities, as in the case of spas – the type of rehabilitation and balneological treatments, and ordinary activities such as cooking, burning (burning candles, using fireplaces, heaters, stoves or smoking cigarettes), cleaning, repair, maintenance and other activities. Elevated concentrations of specific PM fractions are generally influenced by, the presence of tobacco smoke, pesticides, solvents, cleaning agents, mold, fibers, and allergens [Micallef et. al. 1998, Tran et al., 2020]. Thus, indoor PM concentrations are a function of emission rates: primary (combustion, mechanical generation, and resuspension) and secondary (gas-particle phase conversions and reactions) of the source, the air exchange rate determining the infiltration of air pollutants into and out of the indoor environment [Habriel et at., 2014].

Radon-222 (^{222}Rn) should also be considered as an unwanted component of the air (and therefore as a pollutant) when a given health resort does not provide balneological treatments. It is a radioactive isotope of the uranium-radium series with a half-life of 3.82 days. Its parent isotope is rad-226 (^{226}Ra) and its direct decay product is polonium-218 (^{218}Po). Increased activity concentration may have negative effects and then it should be treated as one of the air pollutants. Then it should be eliminated and more specifically its activity concentration should be reduced to the parametric value defined in the law, i.e. below 300 Bq/m^3 [Act, 2000]. One of the sources contributing to the presence of ^{222}Rn in indoor air is its supply with water brought to the building from the water supply system or from own wells. Radon can therefore enter a building in many ways (Fig. 1) such as through cracks and pores in the foundations, structural connections, the water supply and also through finishing materials. Due to the fact that radon is a heavier gas than air, in a building, it will accumulate mainly in the basement – in the case of buildings with basements, or on the first

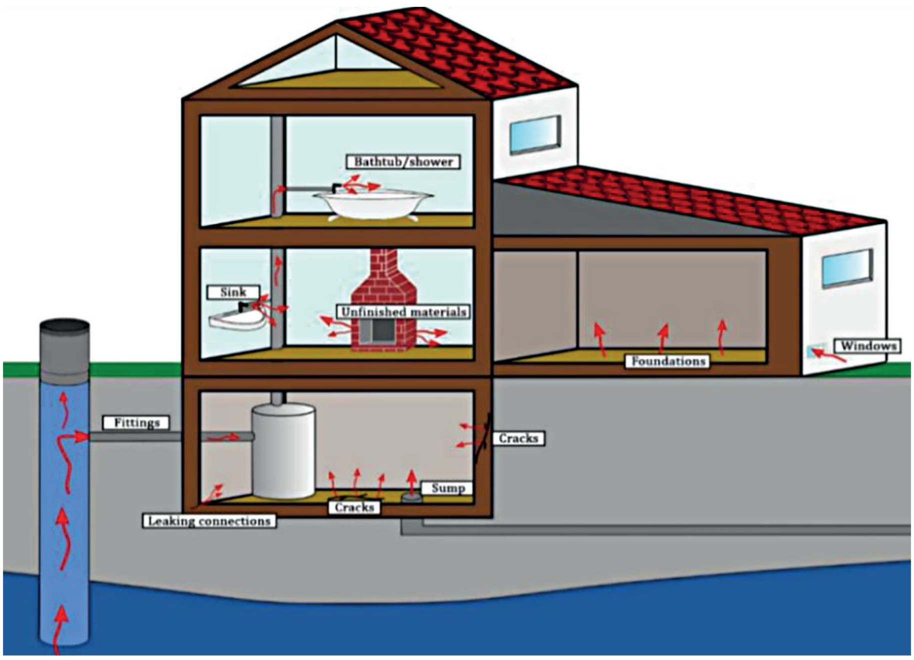

Figure 1. Routes of entry of radon gas into the building [Maciejewski & Kowalska, 2021]

floor – in the case of buildings without basements [Korzeniowska-Rejmer, 2008; Bilska, 2016].

Radon also enters buildings through exhalation from the ground. The concentration of activity of this radioactive isotope in indoor air is therefore mostly influenced by the ground on which the building is situated. Concentrations of uranium, thorium and radium that may be present there are the reason for increased radon content in soil air. Maximum concentrations of the three elements mentioned above occur in magma rocks, mainly granites, phonolites, alkaline syenites, riolites and leucogranites. Their concentrations (of uranium and thorium) are also formed as a result of the postmortem processes – pegmatitic, pneumatolitic and hydrothermal. Radium occurring together with uranium and thorium, as the next link in the chain of nuclear reactions, as a result of the α transformation of radium nuclei, causes the formation of ^{222}Rn. It can then move from the pore space towards the surface of the lithosphere by diffusion due to its concentration difference, convection due to temperature difference or advection due to pressure difference. It can also be efficiently transported through flowing groundwater or gas streams (as long as the fractures and pores are an interconnected system) [Przylibski, 2005; Przylibski 2018].

Formal and legal conditions for the functioning of health resorts, air quality standards

In Poland, the localities that have or apply for the status of a health resort must fulfill the criteria laid down in Polish legislation – in the Act on health resort treatment, health resorts and areas of health resort protection and on health resort communes [Act, 2005]. The requirements concern selected aspects of importance for health resort treatment. The granting or maintaining of the status of a health resort must be preceded by the development of a health resort operative – a report in which the characteristics of the health resort are included, with an indication of the natural medicinal raw materials available there and the health-promoting features of the climate [SAO, 2016]. The municipality in the area of which the health resort or health protection area is located is obliged to prepare and present a health resort operation, at least once every 10 years. The operative must confirm that the locality meets the criteria on the basis of which the minister in charge of health will issue a decision to grant or maintain the status of a health resort [Sówka et al., 2019].

In the current legal state of Poland, there are no requirements that would define parameters for the desired state of indoor air and guarantee its good quality in terms of human health, including in spa facilities. There are also no standards in Poland to which one can refer when analyzing the quality of indoor air. Therefore, we draw conclusions about the quality of indoor air on the basis of measurement data taken in the outdoor environment. Polish and European law impose an obligation to measure concentrations and composition of PM_{10} and $PM_{2.5}$ fractions. Measurements of selected air quality parameters (including PM_{10}, $PM_{2.5}$) are carried out under the State Environmental Monitoring. The atmospheric air monitoring system is mainly based on a network of measurement stations distributed as needed in hot spots of individual provinces, mainly in cities [Sówka et al., 2019].

According to the WHO Global air quality guidelines, the permissible levels of $PM_{2.5}$, considered the most harmful pollutant to health, have been exacerbated twice, the concentration of $PM_{2.5}$ in ambient air should not exceed 5 μg/m³. The permissible daily concentration for $PM_{2.5}$ should not exceed 15 μg/m³ and for PM_{10} 45 μg/m³ [WHO, 2022]. In Poland, the PM_{10} limit level is set for two averaging times: a calendar year and 24 hours. In the first case the standard is 40 μg/m³ and in the second 50 μg/m³, but it is allowed to exceed it 35 times a year. These are values that have not been regulated at a uniform level within the European Union and therefore different standards apply in different Member States in this respect [Kobus et. al., 2020]. For example, US EPA guidelines specify concentration levels in indoor air during a specific duration (i.e., 1 hour, 24 hours, or 1 year). Respectively: 0.15 mg/m³ for a day, 0.05 mg/m³ for a year. These guidelines are usually applied to control indoor air quality, inside households, schools, hospitals, public buildings and offices rather than occupational activity facilities [Tran et al., 2020].

In the case of radon, its permissible activity concentration in Canada was set at 200 Bq/m³ [Paleologos et al., 2021]. The general WHO recommendations (2014) indicate a reference level of 100 Bq/m³. In the event that this level cannot be achieved under the prevailing country-specific conditions, the benchmark chosen should not exceed 300 Bq/m³. In the International Basic Safety

Standards (IAEA, 2014) issued by seven organizations: European Commission (EC / Euratom), FAO, ILO, OECD / NEA, PAHO, UNEP and WHO, the permissible average annual concentration of radon activity is 300 Bq/m^3. In Polish law, the Atomic Law Act (2000) also defines the reference level for the annual average concentration of radon in the air in buildings intended for people and indoor workplaces. This level is 300 Bq/m^3 and it complies with the provisions of the Directive of the Council of the European Union [2013/59/ Euratom].

Indoor air quality as an important health risk factor for spa patients

Spas should exhibit a high level of infrastructure quality as well as environmental quality, especially air quality, which has a great impact on the health of the patients staying there [Expert opinion, 2017]. This is because the spa is frequented by immunocompromised people, including the elderly, children and chronically ill people, classified as so-called vulnerable groups. These individuals are generally at higher risk and thus susceptible to more severe health effects after exposure to air pollution than the general population [Sówka et al., 2019; WHO, 2006].

Air quality in health resorts is an important element embedded in meeting the conditions for obtaining or maintaining health resort status. The state of indoor air is influenced by both internal sources of pollution and external sources of air polluting compounds [Sowa J., 2018]. Air quality is adversely affected by various types of pollutants that arise from transportation, energy production, natural resources used, and therefore, PM and gas emissions pose a serious threat to life in various communities [Kumar et al., 2021]. Air quality affects human well-being, health and life. Breathing polluted air contributes to increased mortality from cardiovascular and respiratory diseases, including lung cancer and chronic obstructive pulmonary disease (COPD), and results in reduced life expectancy. Air is an essential factor in maintaining the proper balance of the body, and includes all forms of existence [Taberham, 2021].

The modern European civilization, living in a temperate climate zone, spends from 80% and even up to 90% of its time indoors [Cincinelli, Martellini, 2017, Saini, 2020], breathing indoor air. During the COVID-19 pandemic, time spent indoors has taken on particular importance. Indeed, isolation and staying indoors were the important elements to counteract the spread of the virus and COVID-19-induced morbidity. In case of so-called hazardous air conditions, it is recommended to stay indoors, in homes, schools, offices, and to avoid excessive contact with outdoor air. However, the question arises whether spas are really safe from outdoor pollution and whether there can be a direct relationship between indoor and outdoor air quality [Su et al., 2013].

It is important to note that the percentage distribution of indoor and outdoor time spent by patients is individual and depends on many factors, including length of stay, number of treatments and their duration, sleep, meals, season, or others such as predispositions or habits. Taking into account the kinesitherapy treatments conducted in the health resort, causing an increased physical effort needed to perform the exercises entails an increased respiratory rate. The increased respiratory rate means that not only fine (generally means less than about 2 μm) but also larger particles are carried to deeper areas of the airways (i.e., tracheobronchial) or parts of the lungs [Lippi et al., 2008; Abelsohn, Stiebm 2011; Sracic, 2016]. Furthermore, most air enters the body directly through the mouth during intensive training, which bypasses the natural filtration mechanisms of the upper respiratory system (nose). The body's oxygen consumption increases by about three times during physical activity. As a result, lung ventilation intensifies. Once training begins, there is a 2- to 3-fold increase in lung minute ventilation (VE) and deepening of breathing [Castro et al., 2015]. If the oxygen demand is even higher, the respiratory rate increases [Bralewska et al., 2019].

AIM, OBJECTIVES OF THE STUDY AND SAMPLING METHODOLOGY

Purpose and scope of the study

It is estimated that exposure to air pollution leads to millions of premature deaths and lost years of healthy life each year. Patients staying at spas have numerous medical conditions and thus are at increased risk of harmful exposure to various agents – including radon and particulate matter. WHO has identified radon as a major factor in lung cancer in non-smokers [WHO, 2022]. According to estimates, radon causes between 3% and 14% of all lung cancers. This level is dependent on national average radon levels and smoking prevalence. The results of the health effects of particulate air pollution are mainly concerned with the causes of respiratory, as well as cardiovascular diseases. Mortality caused by PM pollution is also considered. Particulate air pollution has been classified by the International Agency for Research on Cancer (IARC) as a cause of lung cancer. $PM_{2.5}$ is also the most commonly used indicator to assess the health effects of exposure to ambient air pollution [EEA, 2018] The ACS Cancer Prevention II study found that each 10 μg/m^3 increase in fine particulate matter ($PM_{2.5}$) increases the overall risk of death by 4%, from cardiovascular causes by 6%, and from lung disease by 8%. [WHO, 2022]. Other published data on mortality from PM exposure confirm the risk for long-term $PM_{2.5}$ levels as low as 5 μg/m^3 [WHO, 2022].

Due to a high impact of radon and particulate matter on health, the authors decided to investigate their activity concentrations in indoor air at the spa. In the case of radon, the goal was to determine if the activity concentration level of ^{222}Rn was exceeded – and thus whether the spa guests might be exposed to the carcinogen. In the case of particulate matter, the aim of the study was to investigate the variation in concentrations of particulate matter in indoor air and to see what level of concentrations of pollutants of the PM_{10} and $PM_{2.5}$ fractions are exposed to by patients receiving treatments for therapeutic purposes, in relation to the WHO recommended average daily concentration levels for PM_{10} and $PM_{2.5}$. At the same time, concentrations of particulate pollutants in outdoor and indoor air were compared. The relationship between the measured values of PM_{10} from the monitoring network station with PM_{10} and $PM_{2.5}$ sampled in the spa room was checked. In addition, an analysis of the variability of radon and PM concentrations and atmospheric factors was performed. In the case of the analysis of variability of radon activity concentrations, the effect of the activity of opening windows in the treatment room was also investigated, which, if necessary due to the radon concentration level, made it possible to determine the required frequency of room ventilation to achieve the reference level (below 300 Bq/m^3).

Site description and building site's physical characteristics

The measurements were conducted in the Spa House, built at the beginning of the last century. The building has stone foundations, masonry walls made of solid brick with cement-lime mortar, basement floors are reinforced concrete and the rest is solid ceramic. The sanatorium is located between historic parks and near a provincial road. A treatment room with an area of 114.6 m^2 and capacity of 522 m^3 (the room's height is approximately 4.45 m) was selected for the measurements. This room is connected with another treatment room with a 303 m^3 capacity. Heating and domestic hot water heating is obtained from the local boiler room, where gas-fueled boilers are operated. Additionally, the room was heated by an electric device (local air heater).

The study was carried out during the COVID-19 pandemic, therefore the health resort, due to the suspension of admissions to spas, did not conduct round-the-clock treatment activities, and patients of the health resort accommodation base were not admitted. The therapeutic programme was carried out for external patients, i.e. those arriving or commuting, residents of the health resort or its vicinity, referred under the program of the Social Insurance Institution. The treatments were taken by patients mainly with orthopedic and trauma recommendations, who suffered as a result of traffic accidents or others. And the treatments carried out in the measuring room were especially motor rehabilitation, covering 5 UGUL positions, i.e.: universal therapeutic improvement rooms, 2 positions for cryotherapy treatments and a position for a dry carbonic acid bath. Thus, patients with musculoskeletal diseases together with other accompanying diseases, complications and injuries were rehabilitated in the measurement room. From Monday to Friday, about 100 patients received treatments from 7 a.m. to 3 p.m., on Saturdays from 7 a.m. to 1 p.m., about 60 patients received treatments, and on Sundays, no treatments were performed. During the measurements, there was daily repair and construction work, modernization work in connection with the reconstruction of the operation of the facility, as well as carpentry, painting, assembling, welding work for the needs of the external entity implementing the cultural event.

Experimental setup

A sampling of particulate fractions was carried out in the winter period for 21 consecutive days. In the study of air in the Spa House in one of the Lower Silesian spas – PM fractions were collected simultaneously indoors and outdoors using Harvard impactors from Air Diagnostics and Engineering Inc., Naples, ME, USA. PM concentration measurements were performed according to the European standard for the determination of particulate matter by gravimetric method, PN-EN 12341 [PN-EC 12341, 2014-07].

Two ultra-quiet, oil-free vacuum pumps (Air Diagnostics and Engineering, model SP-280E) and two oil-free, vane-type Becker pumps model VT 4.8 were used for particulate sampling. The airflow rate for particles < 1.0 µm was 23 dm^3/min, whereas for particles < 2.5 µm and < 10 µm – 10 dm^3/min. The airflow rate was determined using a gas meter type ACTARIS G2.5 GALLUS 2000. The control of the airflow rate as a factor important for the proper selection of the PM fraction that passes through the separation head was performed using a rotameter and a Madd-Stream Bronkhorst gas flow controller (Fig. 2).

For particulate matter sampling, QM-A quartz filters of 37 mm diameter made by Whatman were used. Internal sampling was performed closest to the patients' breathing zone, the air intakes were set at a height of about 1.5 m above the treatment room floor. Clean filters were conditioned and weighed in the laboratory, placed in transport containers, then transported to the measuring station and placed in the sampler before insertion into the sampler. The filters, after exposure of 24 h, were transported to the laboratory. In the laboratory, the filters were conditioned and weighed a second time, already as post-exposure filters. From the differences in masses before and after filter exposure, related to the volume of air flow in the sampler, the PM concentrations were calculated. These concentrations are given in micrograms per cubic meter [µg/m^3].

Airborne particulate matter (PM) concentrations were calculated using eq. 1:

$$c = \frac{ml - mu}{\varphi at} \quad (1)$$

where: c – PM concentration [µg/m^3]; ml – mass of the filter with particulates [µg]; mu – mass of clean filter [µg]; φa – air flow under actual conditions [m^3/h]; t – PM extraction time [h].

Measurement uncertainty for the reference sampler was estimated for the limit values based on the standard: PN-EN 12341:2014-07 Ambient air. Standard gravimetric measurement method for determination of mass concentrations of PM_{10} and $PM_{2.5}$ fractions of particulate matter (for $k = 2$ and 95% confidence level). The mass of particulate matter was gravimetrically determined using a Radwag MYA 5.3Y.F1 electronic microbalance, with a reading accuracy of $d = 1$ µg.

Airborne ^{222}Rn activity concentrations were measured using an AlphaGUARD PQ 2000 PRO instrument (Manufacturer: Genitron Instruments) with a measurement range of 2 Bq/m^3 to 2,000,000 Bq/m^3. It is a portable device enabling continuous measurements of radon activity concentration and its derivatives in the air as well as measurements of climatic parameters such as atmospheric pressure (p), air temperature (T) and relative humidity (f). The device is equipped with internal memory, in which the measurement results are stored. Two operating modes are possible: diffusion and flow. The air enters the ionisation chamber, which is the

Figure 2. Particulate measuring devices in outdoor and indoor air used for the study at the spa; a) Harvard impactor for measurement of PM_1, $PM_{2.5}$, PM_{10} and fractions, b) Measurement set: impactor, Becker pump, ACTARIS G2.5 GALLUS 2000 gas meter; c) Vacuum pump – Air Diagnostics and Engineering, Air sampling pump, model SP-280E

ionising radiation detector, through a glass fiber filter so that neither radon derivatives nor contaminants in the form of PM enter the chamber. The cylindrical ionization chamber has an active volume of 0.56 dm^3, and its metal interior has a potential of +750V. A 0V potential electrode is placed along the longitudinal axis, which is connected to a very sensitive preamplifier. The measurement signals are sent to an electronic network for further processing (Manual, 2012). A flow-through mode with a 10-minute time step was used to perform ^{222}Rn activity concentration measurements. Measurements were conducted from 04–26/02/2021 with a technical break from 16–24/06/2021. A total of 1980 results of 10-minute measurements were obtained. Data Expert software was used to read the recorded results from the device's internal memory.

The air, analogous to the PM measurements, was collected by the device from a height corresponding to the location of the initial elements of the patients' respiratory system during the reception of treatments. Both devices (for radon and PM measurements) were located in the same treatment room, located in a basement building on the first floor.

RESULTS AND DISCUSSION

The variability of PM_{10} and $PM_{2.5}$ concentrations

The analysis of measurement results shows that values of 24-hour mean concentrations of PM_{10} and $PM_{2.5}$ in the whole series of measurements ranged from 13.76 to 42.24 µg/m³ for $PM_{2.5}$ and from 15.56 µg/m³ to 49.81 µg/m³ for PM_{10} (Table 1). However, the mean for all obtained measurements was 24.50 µg/m³ for $PM_{2.5}$ and 32.23 µg/m³ for PM_{10}, with a standard deviation of 8.20 µg/m³ and 10.59 µg/m³, respectively.

The distribution of variability of PM_{10} and $PM_{2.5}$ concentrations in indoor air showed an increase in PM_{10} concentration with rising in $PM_{2.5}$ levels (Fig. 3). The obtained value of correlation coefficient $r = 0.96$ indicates a strong relation between 24-hour average concentrations of PM_{10} and $PM_{2.5}$. In the course of variability of PM_{10} and $PM_{2.5}$ concentrations, no relation between measured values of indoor particulate matter concentrations and measured temperature of indoor air was observed.

Conducted measurements of concentrations of two fractions of particulate matter in indoor air indicate a high content of $PM_{2.5}$ in PM_{10}. The percentage of $PM_{2.5}$ in PM_{10} ranged from 69.02% to 91.44%, while the average value was 77.12%. Lower values of $PM_{2.5}$ concentrations in relation to PM_{10} can be observed

Table 1. Average 24-h PM_{10} and $PM_{2.5}$ concentrations

Statistical parameters	Daily mean concentrations of PM fractions [µg/m³]	
	$PM_{2.5}$ inside	PM_{10} inside
Min	13.76	15.56
Median	24.60	30.37
Max	42.14	49.81
Mean	25.25	32.12
Std. deviation	8.54	10.60

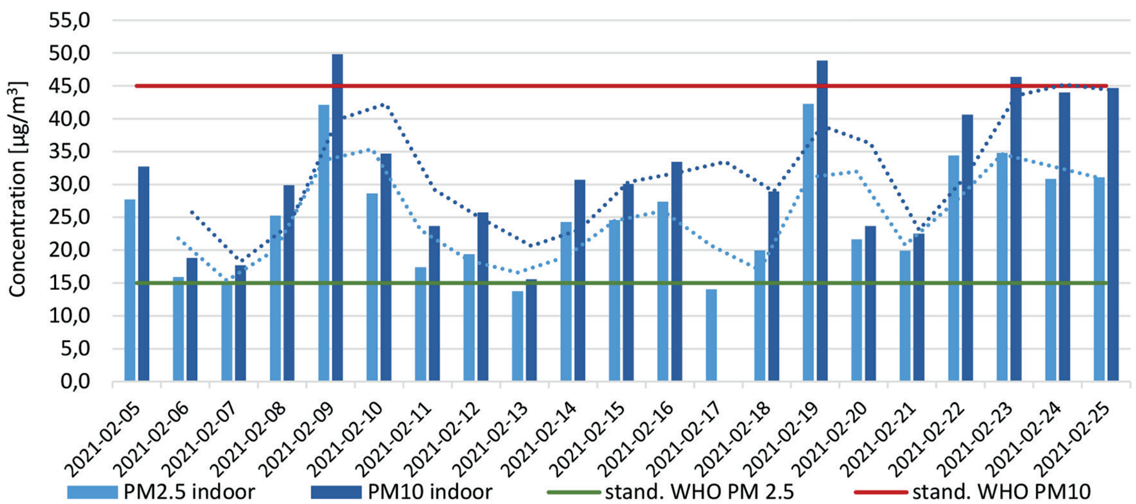

Figure 3. The average 24-h concentrations of PM_{10} and $PM_{2.5}$ measured in the period of 5.02.2021–25.02.2021 in indoor air

mainly in the last days of the measurement cycle (74.98%, 70.15%, 69.56%).

The high content of $PM_{2.5}$ in PM_{10} recorded in outdoor air (at the monitoring network stations) in winter is characteristic for the duration of the heating season and usually results from the combustion of solid fuels for heating purposes. If the source of emission is the combustion of solid fuels in boilers or furnaces, then we are dealing with elevated emissions of very fine particles originating, among others, from soot. The above may also indicate that the source of indoor PM may be the combustion of solid fuels for heating purposes and their subsequent transport inside the treatment room.

At the same time, it can be noted that during the study period the concentrations of PM_{10} and $PM_{2.5}$ fractions measured in indoor air exceed the WHO recommended values for PM_{10} of 45 µg/m_3 and for $PM_{2.5}$ of 15 µg/m^3 specified for outdoor air. The analyses of PM concentrations show that there is a correlation between the variability of concentrations of PM_{10} (O) measured at the state monitoring network station (outdoors), located directly at the Spa House, and PM_{10} (I) and $PM_{2.5}$ (I) measured indoors. In the course of variability of concentrations for PM_{10} (I) and $PM_{2.5}$ (I) as well as PM_{10} (O) (Fig. 4a), it was observed that almost during the entire measurement period there were simultaneously elevated episodes of concentrations of PM measured outside and inside the room (Fig. 4). The exception was the values recorded on 15.02, which could have been related to meteorological conditions prevailing on that day. According to the

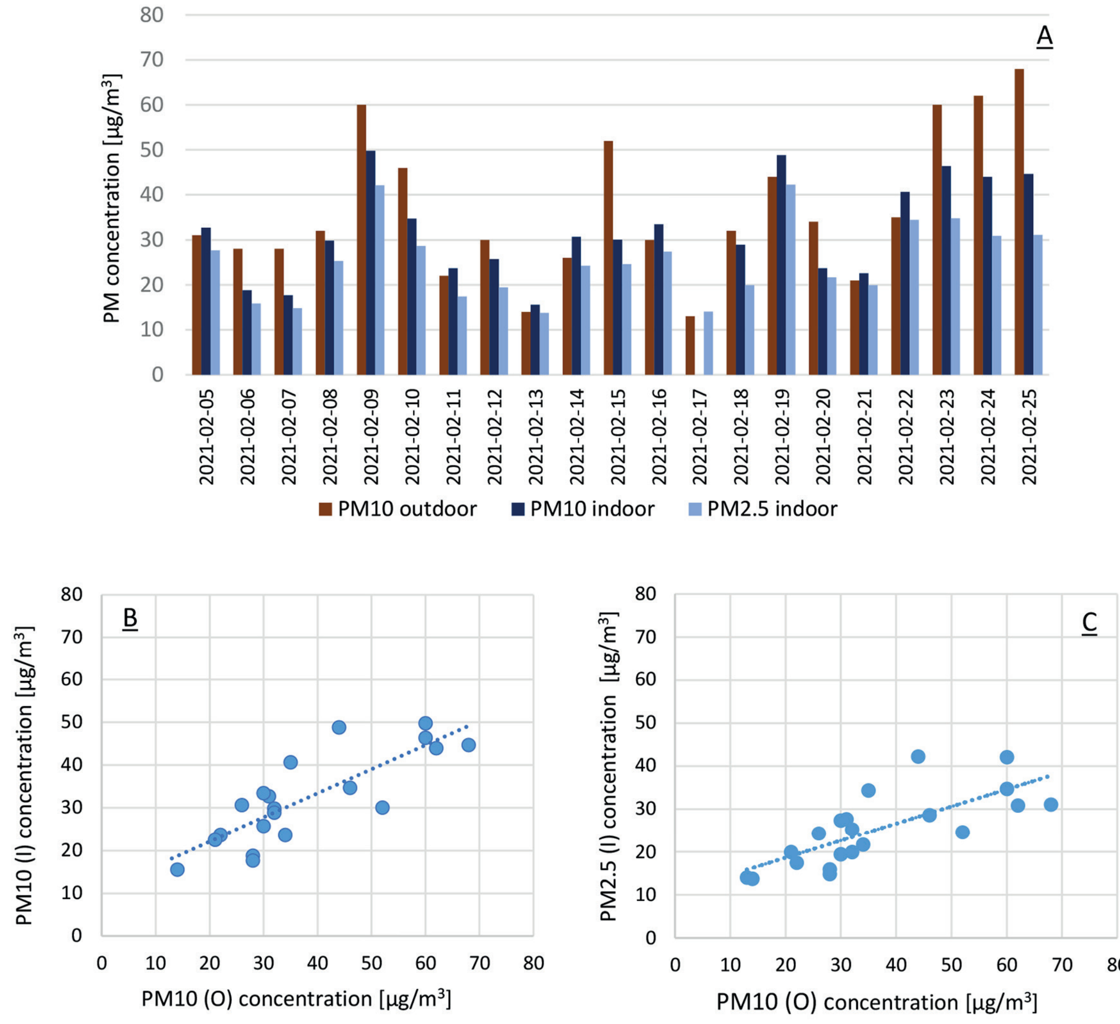

Figure 4. a) Relationship between 24-h average PM_{10} and $PM_{2.5}$ concentrations measured in indoor air and PM_{10} measured in outdoor air for the period 5.02.2021–25.02.2021; b) Correlation between the 24-h average PM_{10} (I) indoor and PM_{10} (O) outdoor concentrations measured at the monitoring network station and c) correlation between the 24-h average PM_{10} (I) indoor and $PM_{2.5}$ (O) concentrations measured at the monitoring network station

data made available by IMGW (Institute of Meteorology and Water Management), the wind speed was 0.9 m/s, qualified for the 2nd degree on the Beaufort scale. The almost windless weather conditions on that day most probably could have had an impact on the limited dispersion of particulate pollutants in the outdoor air and their movement into the indoor environment.

To express the differences in PM fraction concentrations, the average I/O (indoor/outdoor) ratio for daily average concentrations was calculated. The calculated I/O ratio for PM_{10} was 0.8. It is believed that a lower ratio indicates the influence of outdoor particulate sources. During the study, air exchange from outside to inside took place, which is indicated by the value of the ratio below 1. The I/O ratio and the values of PM_{10} (O) PM_{10} (I) and $PM_{2.5}$ (I) concentrations (Fig. 4b and 4c) indicate the migration and infiltration of particulates from the outside environment to the inside, caused by leaks in the housing of the treatment facility, including leaky wooden windows.

It should be added that air exchange may have occurred primarily through the ventilation opening, windows, and doors opened with very high frequency, i.e., with the entrance and exit of each of more than 100 patients per day. Apart from external sources of air pollution with particulate matter, the values of PM_{10} and $PM_{2.5}$ concentrations were also influenced by other internal sources, such as cleaning works carried out every day after the completed treatments and carpentry works carried out during the study period, assembly and processing of wooden and, to a lesser extent, metal elements (more detailed analyses of trace elements in PM will provide knowledge). The above-mentioned works were conducted in connection with the change of functional organization inside the facility and another event of a cultural nature with its organization of props.

Air temperature determines the activity of heating sources in the winter period, which also influences the amount of pollutants emitted from the municipal and household sector. Due to the fact that the measurement period was characterized by very diversified meteorological conditions, it is difficult to unequivocally determine the relation between air temperature and 24-h average values of PM concentrations in outdoor air for the analyzed time interval. From 5.02 to 25.02 the air temperature based on the air measurements taken outside the health resort facility ranged from –10.1 to +11.7°C. On the other hand, according to data from IMGW, wind speed ranged from 0.9 to 3.8 m/s.

In the last days of the study, i.e. 22.02. to 25.02. there were anticyclone weather conditions with dry, very warm air, which favored more lightening up to a cloudless sky. Such conditions in turn contributed to the formation of strong radiation inversion and gradual accumulation of pollutants in the near-ground air layer. Additionally, on 23–24.02 over Poland appeared large plume of Saharan dust moving. This phenomenon, associated with a higher concentration of particulates, was also observed in the form of elevated values of PM_{10} concentration in the atmospheric air. Consequently, the above factors and phenomena resulted in elevated values of particulate matter concentrations being recorded at higher air temperatures.

Indoor radon activity concentration levels

The measurements of radon activity concentration show that during the whole cycle it varied from the lower limit of detection (LLD) – below 2 Bq/m^3, to a maximum of 62 ± 21 Bq/m^3 (Table 2). The mean of the results on individual days ranged from 15 Bq/m^3 to 32 Bq/m^3 while the mean of the whole cycle was 18 Bq/m^3 with a standard deviation of 10 Bq/m^3.

Considering the average hourly fluctuations of ^{222}Rn activity concentration it should be stated that they were very high (Fig. 5). The dependence of the concentration of ^{222}Rn activity on the temperature or, more precisely, on the air exchange was also observed (the change of temperature inside the room was connected with the opening of windows by the Spa House employees). The opening of windows caused a decrease in temperature, but what is important, it was connected with bringing to the treatment room air from outside the building with a lower concentration of ^{222}Rn activity and, consequently, with a decrease in the concentration of ^{222}Rn activity in internal air due to the exchange of internal air (with higher ^{222}Rn concentration activity) with external air (with lower concentration). An apparent episode of temperature decrease on 7.02.2022 causing the recording of its lowest value was connected with leaving the window open by employees for the whole night and, consequently, with continuous air exchange.

Based on the average value of ^{222}Rn activity concentration from all measurement days at

Table 2. Average 24-h ^{222}Rn activity concentrations

Sampling date	Statistical parameters of ^{222}Rn [Bq/m^3]				
	Min	Median	Max	Mean	Std. deviation
04.02.2021	4	16	34	16	7
05.02.2021	< 2	13	43	15	8
06.02.2021	< 2	16	32	15	8
07.02.2021	< 2	13	43	15	9
08.02.2021	1	18	46	18	8
09.02.2021	< 2	18	55	18	9
10.02.2021	< 2	16	48	17	9
11.02.2021	< 2	13	35	14	8
12.02.2021	< 2	16	40	16	8
13.02.2021	< 2	15	45	15	9
14.02.2021	< 2	16	40	15	8
15.02.2021	< 2	18	40	18	9
16.02.2021	< 2	17	36	17	7
24.02.2021	4	24	62	25	12
25.02.2021	< 2	25	50	26	10
26.02.2021	10	32	56	32	11
04–26.02.2021	< 2	16	62	18	10

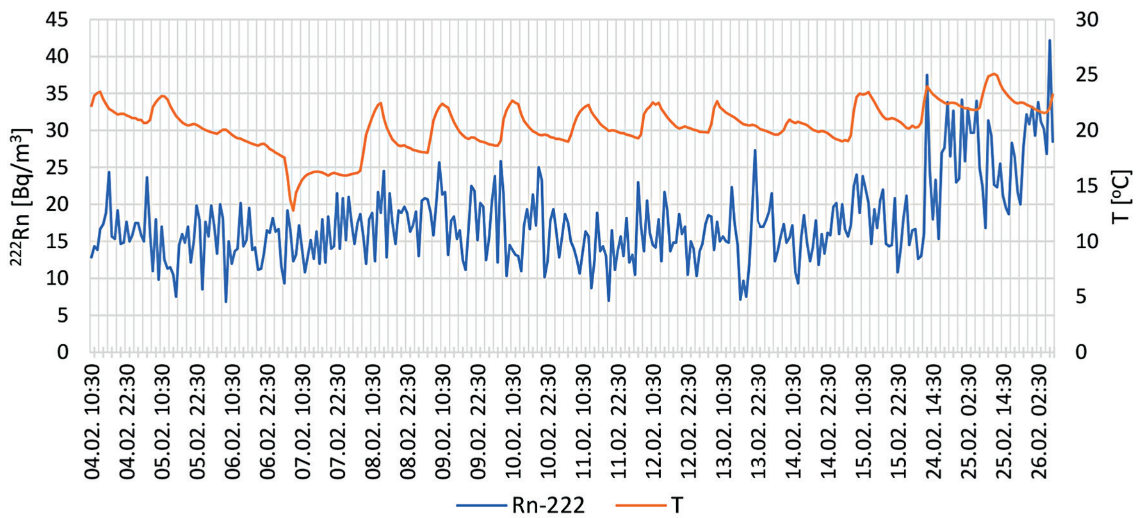

Figure 5. Changes in indoor air temperature and ^{222}Rn activity concentration in the air inside the study room during the period 5.02–25.02

individual hours (Fig. 6), it is evident that an increase in concentration was recorded from the moment the treatments were completed and the treatment room was closed. The decrease in the rate of air exchange (no open windows, no patients opening the door repeatedly and moving around the room) in effect causes an accumulation of ^{222}Rn inside the treatment room. Maximum values occurred during the night hours.

Each day, the room was ventilated in the morning (about 7 a.m.). A significant decrease in ^{222}Rn activity concentration was evident at that time (from 22.5 Bq/m^3 at 7 a.m. to about 15.8 Bq/m^3 at 9 a.m.; a decrease of about 23%). Peaks in ^{222}Rn activity concentrations are evident during the hours when patients were admitted and the room was ventilated at different intervals and intensities. These occur until the hour when the treatments are finished and the room is closed. The measurements show that the average ^{222}Rn activity concentration for particular hours varies from 15.7 Bq/m^3 (at 12 p.m.) to 20.5 Bq/m^3 (at 7 a.m.).

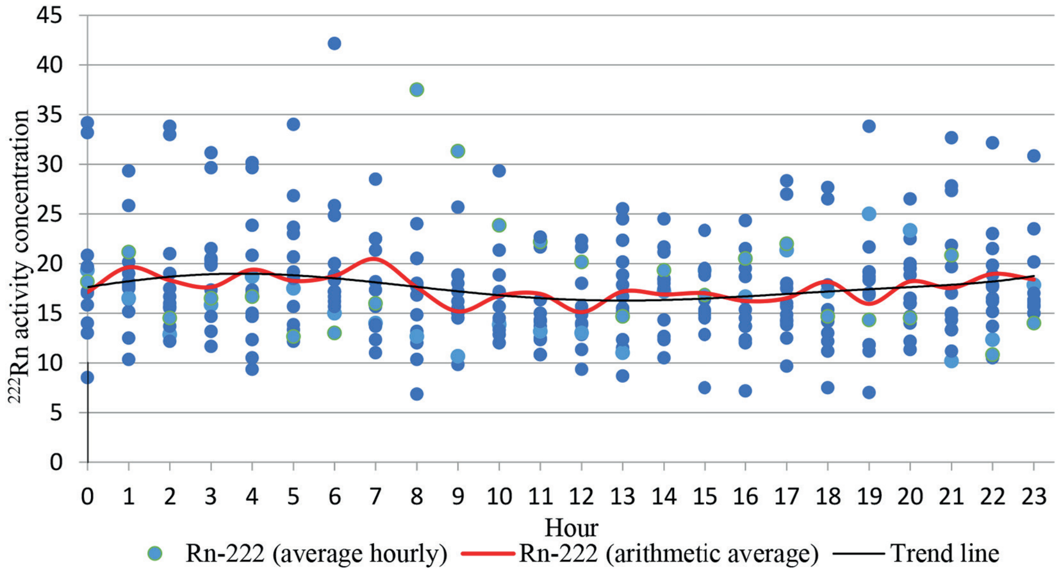

Figure 6. The average activity concentration of ^{222}Rn and the trend line in the air inside the study room during the period 5.02–25.02

In the case of short-term measurements of radon activity concentrations in the air, it was found that they do not exceed reference levels specified by the Atomic Law or WHO. It was also found that the observed changes in the course of daily hourly concentrations of ^{222}Rn activity are analogous to those observed in residential buildings.

The analyses were carried out to compare the course of variability of ^{222}Rn activity concentration and PM levels (Fig. 7 a–d) – especially

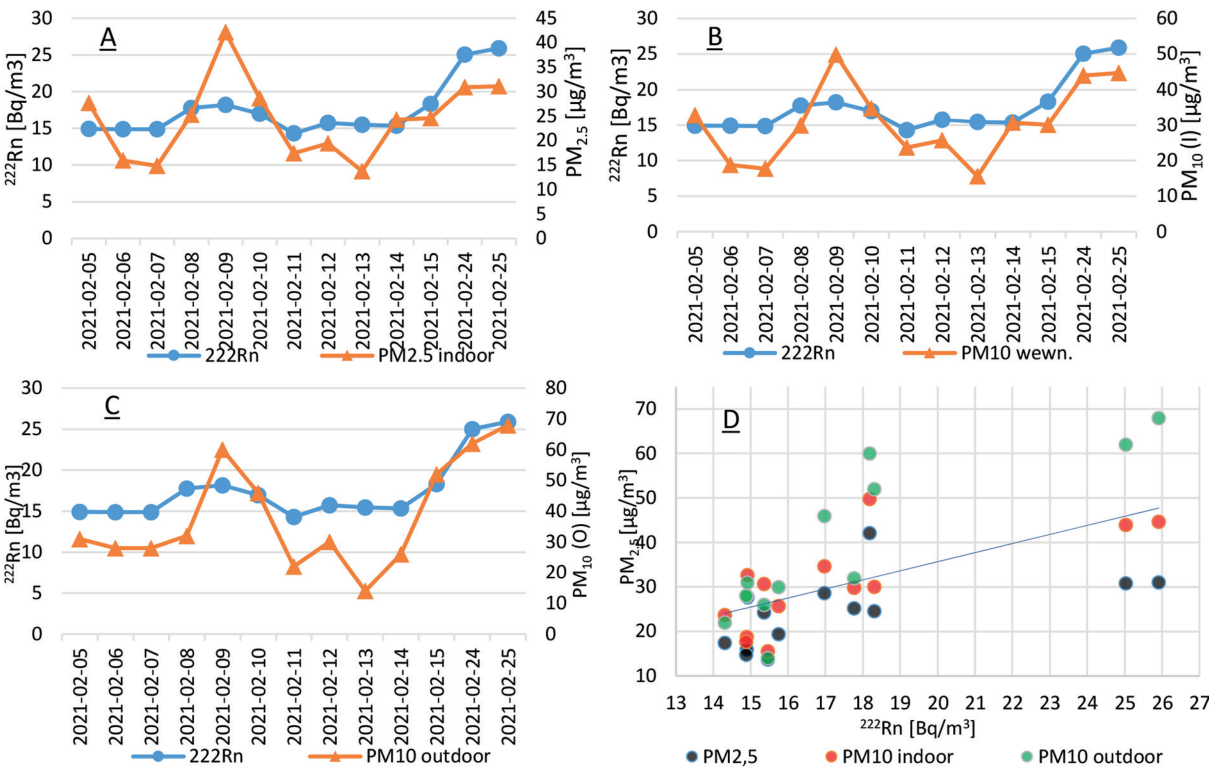

Figure 7. Average 24-h ^{222}Rn concentration activity (dark blue continuous line) and 24-h average concentration of particulate matter (orange continuous line): $PM_{2.5}$ (a), PM_{10} indoor air (b), PM_{10} outdoor air (c) according to measurement days and the dependence of particulate matter concentration: $PM_{2.5}$ (black points), PM_{10} indoor (orange points), PM_{10} outdoor (green points) on the concentration of ^{222}Rn activity (d)

in the period of high concentration episodes on 08–09.02.2021 and 24–25.02.2021. Preliminary analysis of measurement results shows that for selected periods there were simultaneous increases in concentrations of PM_{10} and $PM_{2.5}$ and ^{222}Rn. The simultaneous increase in radon activity and particulate concentrations was related, on the one hand, to the decreased intensity of weathering and, on the other hand, to the occurrence of internal sources of pollution with a simultaneous high concentration of PM in the external air.

CONCLUSIONS

The conducted measurements indicate that the spa patients, during the selected periods, were exposed to PM_{10} (I) and $PM_{2.5}$ (I) concentrations exceeding the WHO recommendation levels for outdoor and indoor air. From the study of particulate matter in the indoor air of the Spa House, a relationship was observed between the variation in indoor $PM_{2.5}$ and PM_{10} concentrations and the variation in PM_{10} levels measured in outdoor air (O). The coefficient calculated from the I/O ratio for PM_{10} of 0.8, indicates that there was an exchange of air from outside to inside during the study. Thus, air migration with particulate matter pollution occurred, mainly due to daily opening of windows and multiple opening of doors during the day. Additionally, infiltration of outside air into the interior occurred, caused, among others, by window leaks. The contribution of external sources to indoor air pollution by particulate matter is confirmed by the obtained relationship between the variation of PM_{10} (O) and PM_{10} (I) and $PM_{2.5}$ (I) concentrations and the value of the I/O coefficient as well as the share of $PM_{2.5}$ (I) in PM_{10} (I). At the same time, the share of $PM_{2.5}$ (I) in PM_{10} (I) in the studies carried out in the winter period indicates that combustion of solid fuels is a source of particulate emission to the outdoor air, transported/infiltrated subsequently indoors.

In the area of the spa town, solid fuel is still used for heating dwellings, this concerns about 300 objects located around the object of the measurements conducted. Additionally, the Spa House is located in a low-lying area, which hinders dispersion of the transported PM pollution and favors its concentration, as evidenced by high values of particulate matter concentrations (exceeding the reference values) measured at the monitoring network station located next to the facility measured by the authors. Particulate matter pollutants get inside mainly through open windows and doors as well as partitions and building leaks of the spa facility. Moreover, it is not excluded that the concentrations of PM inside the building could have been influenced by additional sources of particulate emission, namely: cleaning works (sweeping, vacuuming) and works, carpentry, assembly, painting and others, which were performed during the measurements.

In the case of ^{222}Rn activity concentration, it was found that ventilation of the treatment room, in addition to a decrease in temperature, resulted in air exchange and due to the inflow of external air of lower ^{222}Rn activity concentration, the concentration inside the room decreased. Taking into account ^{222}Rn only, it should be concluded that there was no need to ventilate the room in order to reduce ^{222}Rn activity concentration. It should be concluded that during the measurement period the reference level of ^{222}Rn activity concentration defined by IAEA (2014) as well as the Atomic Law (2000) at 300 Bq/m^3 or more restrictive, the first of two reference levels defined by WHO (2013) i.e. 100 Bq/m^3 was not exceeded. It can also be concluded that the daily distribution of radon activity concentration is analogous to that observed in residential buildings.

Based on the analyses of ^{222}Rn activity concentration and $PM_{2.5}$ and PM_{10}, it was noted that 222Rn activity concentration also increased during periods of elevated values for PM. Analyses indicating correlations between the concentrations of ^{222}Rn activity and $PM_{2.5}$ and PM_{10} require further study.

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