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Spatiotemporal Variability of Urban Air Pollution in Bucharest City

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Abstract: Urban air pollution is one of the major challenges that cities around the world face. Particulate matter (PM), nitrogen dioxide (NO_2), volatile organic compounds (VOCs), and other pollutants are many times over the recommended airborne exposure, generating a strong impact on human health and city well-being. Considering Bucharest as a case study, this study aimed to investigate the patterns of particulate matter and nitrogen dioxide concentrations. Multiyear data from the Romanian National Air Quality Monitoring Network were used to investigate spatial and temporal variability. All air pollutants presented a typical bimodal trend during the day, with specific double peaks corresponding to the morning rush hours and nighttime. Spatial variability in NO_2 concentrations was observed, with almost double the concentration values in the city center during midday compared with those for the background and industrial areas. A weekly pattern of PM was noticed, with lower concentrations during the weekends in comparison with those during weekdays, more pronounced in the case of PM_{10} compared with the case of $PM_{2.5}$. The fine particle fraction presented monthly and seasonal variability, with higher levels during the cold months compared with the warm months, mainly corresponding to the increased household heating. The estimated proportion of mortality attributable to annual exposure to an air $PM_{2.5}$ above 5 μg/m^3 in Bucharest ranged between 7.55% and 8.26%, with the maximum from 2021. By contrast, the estimated proportion of mortality attributable to PM_{10} and NO_2 above 10 μg/m^3 was significantly lower, with values around 4%. The results are useful in supporting environmental planning measures to decrease urban air pollution.

Keywords: air pollution; atmospheric pollutants; source apportionment

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Citation: Ilie, A.; Vasilescu, J.; Talianu, C.; Iojă, C.; Nemuc, A.; Spatiotemporal Variability of Urban Air Pollution in Bucharest City. *Atmosphere* **2023**, *14*, 1759. https://doi.org/10.3390/atmos14121759

Academic Editors: Thiago Nogueira, Taciana Toledo De Almeida Albuquerque, Rodrigo J. Seguel, Manousos Ioannis Manousakas and Néstor Y. Rojas

Received: 13 October 2023
Revised: 17 November 2023
Accepted: 20 November 2023
Published: 29 November 2023

1. Introduction

Cities face many environmental challenges that influence their attractiveness and competitiveness. These challenges include climate change, air pollution, and urban expansion. Particulate matter, nitrogen oxides, sulfur oxides, volatile organic compounds, and ozone are among the air pollutants most likely to exceed the recommended levels of population exposure.

Particulate matter (PM) and nitrogen dioxide (NO_2) have an important impact on air quality, climate, and consequently human health [1,2]. Particulate matter is frequently classified depending on its size, with PM_{10} being particles that have diameters less than 10 μm, while $PM_{2.5}$ represents particulate matter with a diameter less than 2.5 μm. Atmospheric aerosols are small particles suspended in the atmosphere, including soot, dust, smoke, pollen, and liquid droplets. They can be a combination of organic and inorganic substances of natural or anthropogenic origin [3]. The main sources of PM are fuel burning, traffic, industrial emissions, soil erosion, and biomass burning [4–6]. Another group of pollutants that contribute to air quality is NO_2, produced mostly by traffic emissions but also due to the production of electricity and due to commercial, institutional, and household activities [7,8]; SO_2 is primarily released into the atmosphere from the combustion of sulfur-containing fossil fuels, such as coal and oil [9,10]; ground-level O_3 is a secondary pollutant formed by the reaction of volatile organic compounds (VOCs) and nitrogen oxides (NOx) in the presence of sunlight [11]; and CO is a colorless, odorless gas produced by the incomplete combustion of carbon-containing fuels, such as gasoline and wood. These can be emitted from various sources, including vehicles, industrial processes, and residential heating [12].

The European Environment Agency (EEA) plays a crucial role in performing air quality monitoring following the guidelines from EU air quality directives, in coordination with the National Environmental Agency network, providing the necessary tools to achieve informed political decisions related to the environment. These guidelines aim to protect human health and the environment from the detrimental effects of air pollution. The EU's directives focus on various pollutants, such as particulate matter (PM) and nitrogen dioxide (NO_2), among others. The EU air quality directives in place, 2008/50/EC and 2004/107/EC, establish a 24 h limit of 50 $\mu g/m^3$ for PM_{10}; no limit for $PM_{2.5}$ yet; an hourly outer limit of 200 $\mu g/m^3$ NO_2; and annual limits of 40 $\mu g/m^3$ for PM_{10}, 25 $\mu g/m^3$ for $PM_{2.5}$, and 40 $\mu g/m^3$ for NO_2 [1,13]. Recently, the European Commission proposed a revision of the Ambient Air Quality Directive to impose stricter limits for air pollution than the current ones, towards alignment with those set by the World Health Organization.

The World Health Organization (WHO) updated its guidelines related to air quality health risk values in 2021, recommending even lower average concentrations of air pollutants than the air quality directives. The WHO recommends an annual limit of 15 $\mu g/m^3$ for PM_{10}, indicating that the average concentration of PM_{10} over a year should not exceed this threshold. Additionally, there is a 24 h limit of 45 $\mu g/m^3$. The recommended annual limit for $PM_{2.5}$ is significantly lower, set at 5 $\mu g/m^3$. This indicates a stricter standard for $PM_{2.5}$ concentrations, emphasizing the need to minimize long-term exposure to this pollutant. Therefore, the 24 h limit for $PM_{2.5}$ is set at 15 $\mu g/m^3$. Another significant air pollutant considered by the WHO guidelines is NO_2. The recommended annual limit for NO_2 is 10 $\mu g/m^3$; the average concentration over a year should not exceed this level to protect human health. Additionally, there is a 24 h limit of 25 $\mu g/m^3$ for NO_2 [14].

Air pollution is responsible for the majority of premature deaths, with heart disease and stroke being the leading contributors, followed closely by respiratory diseases and lung cancer [15–18]. In the European Union, in 2020, there were 238,000 premature deaths due to exposure to $PM_{2.5}$ concentrations above 5 $\mu g/m^3$ and 49,000 premature deaths due to exposure to nitrogen dioxide levels above 10 $\mu g/m^3$. Moreover, according to the European Environment Agency (EEA), from 2022, Romania saw 21,600 premature deaths due to particulate matter, and almost 1,211 years of life were lost per 100,000 inhabitants [19]. A health risk assessment is a crucial process used to systematically evaluate and understand the potential health risks that individuals or entire populations may face due to exposure to various environmental agents or stressors. These agents could include pollutants, chemicals, biological agents, or other hazardous substances found in air, water, soil, or food [20].

Urban air pollution is a critical environmental concern that affects cities worldwide, including Bucharest, the capital city of Romania [21]. As urban areas continue to grow and industrialize, the emission of pollutants into the atmosphere poses significant health risks and ecological consequences. In urban environments, air pollution has become a major issue due to factors such as vehicular emissions, industrial activities, and energy production [22,23]. The assessment of air quality in several European cities is thus emphasized: Serra San Bruno, Italy [24]; Istanbul, Türkiye [25]; Thessaloniki, Greece [26]; Copenhagen, Denmark [27]; and several cities across France [28,29]. Furthermore, the knowledge of air quality variability in Bucharest is very limited [30–32]; only a few studies have focused on identifying the possible particle sources, as well as their properties, and only for short time periods and for one PM size (e.g., [33–36]).

In Romania, air quality monitoring and management are performed by the Ministry of Environment under the Romanian National Air Quality Monitoring Network (RNMCA), which provides standard air pollution monitoring data, with a total of 30 air quality measurement stations distributed throughout Bucharest and its peripheral areas [37]. In recent decades, the monitoring of air quality in Romania has been also motivated by a mandate imposed by the European Union at the proposal of Greenpeace and ClientEarth [30,38–40].

This study focuses on the assessment of air quality, specifically particulate matter and NO_2 concentration variability over three years in Bucharest, emphasizing the seasonal, diurnal, and spatial variability. The article is structured as follows. Section 2 describes the measurement location and data, as well as the data treatment and tools used for data analysis. PMs and gaseous variability are discussed in Section 3. The Section 4 contains the concluding remarks on air pollutant variability in Bucharest city.

2. Materials and Methods

2.1. Study Area and Measurements

Bucharest is located in the south of the Romanian Plain in the eastern part of Europe, characterized by a temperate continental climate [41]. The pollutants are transported from the vicinity, and the urban area suffers from increased pollution due to the large number of inversions influenced by the topography of the area [42,43].

Bucharest, the capital of Romania, is an urban hub facing significant challenges in regard to air pollution. This large city, home to over 2 million people, has experienced a steady increase in air pollution levels in recent years [44]. Furthermore, industrial zones in and around Bucharest, coupled with the use of outdated technologies, release a variety of harmful pollutants into the atmosphere. Finally, during the cold winter months, the city's reliance on solid fuel for heating further exacerbates the air quality issue, as emissions from wood and coal combustion become prevalent [45,46].

This study uses air quality data from the National Air Quality Monitoring Network (RNMCA) stations distributed throughout Bucharest. Measurements performed at 8 stations have been selected based on data availability during the study time frame (2020–2022). All types of stations are selected for the study: urban-type stations (2) in the east and northeast of the city, a suburban station (1) located 4 km south of the capital, industrial-type stations (3) located in the southern half, and traffic-type stations (2) located in the center of the city (see Table 1).

The study focuses only on particulate matter and nitrogen dioxide, which are available in all stations (CO (5 stations), O_3 (3 stations), and SO_2 variability are included in Supplementary Figure S1, due to the low data availability). It should be also mentioned that the 8 stations in the case of PM_{10} and $PM_{2.5}$ measurements are equipped with different sensor types and consequently with different uncertainties, but follow the same standards of measurement.

Table 1. Air quality measurement stations [37].

Station	Type	Available Measurements	Location	Coordinates
B1	Urban	PM_{10}, $PM_{2.5}$, SO_2, O_3, NO_2, CO	Militari	44.45, 26.04
B2	Industrial	PM_{10}, SO_2, NO_2, CO	Titan	44.42, 26.16
B3	Traffic	PM_{10}, NO_2, CO	Obor	44.44, 26.13
B4	Industrial	PM_{10}, SO_2, NO_2	Berceni	44.38, 26.13
B5	Industrial	PM_{10}, $PM_{2.5}$, SO_2, NO_2, CO	Drumul Taberei	44.42, 26.03
B6	Traffic	PM_{10}, $PM_{2.5}$, NO_2, CO	Universitate	44.44, 26.10
B7	Suburban	PM_{10}, $PM_{2.5}$, SO_2, O_3, NO_2	Magurele	44.35, 26.03
B9	Urban	PM_{10}, $PM_{2.5}$, O_3, NO_2	Bucurestii Noi	44.49, 26.03

2.2. Data Treatment

The data set used to assess the pollutants' variability is represented by three years (from 1 January 2020 to 31 December 2022) of measurements. The hourly average concentrations of PM_{10}, $PM_{2.5}$, and NO_2 from the selected RNMCA stations were further used in the study to characterize the air pollutants in the Bucharest area.

The data filtering was performed using a custom-made Python code, through several steps as follows. During the first filter, the consistency of the data for all pollutants was checked using a moving average for 3 points (3 h). All concentrations with values higher and lower than the double mean were removed. These represented only 0.08% of the data. The values of the ratio of $PM_{2.5}$ and PM_{10} were used in the second step to identify any irregularities that might exist within the data set. Any measurements that displayed a ratio exceeding 1 were considered anomalous and subsequently eliminated from the analyzed data set. This filtering approach ensured the reliability and accuracy of the remaining data for the assessment of factors influencing air quality and potential environmental concerns. Moreover, this ratio was further used to gain insights related to the particulate matter granularity, possible composition, and potential sources of pollution.

The Quantum Geographic Information System (QGIS) software (https://qgis.org/en/site/), made by QGIS Development Team, Gossau, Switzerland, was used to visualize and analyze the filtered data on a map, enabling a comprehensive understanding of the spatial characteristics of the data set [47]. Using this tool, we were able to identify patterns, trends, and spatial relationships, providing valuable information on the distribution and concentrations of the $PM_{2.5}$ and PM_{10} pollutants.

During the three years of measurement, the maximum number of possible data points was 26,301. The temporal data coverage was calculated as a percentage to show the data availability for the entire time frame of 3 years. Table 2 presents only the stations with more than 95% data availability, those that measured two thirds of the period, and the B9 station with data available only during 2022. The filtered data set (last column of Table 2) is representative, since the lowest filtered coverage (from the total temporal coverage of the data) is 92.54% for the B5 station.

Table 2. Data coverage for every station.

Station	Measurements	Temporal Coverage	Filtered Coverage from the Total
B1	25,085	95.37%	98.40%
B2	17,593	66.89%	100%
B3	26,116	99.29%	100%
B4	17,804	67.69%	100%
B5	17,527	66.64%	92.54%
B6	25,380	96.49%	95.29%
B7	17,370	66.04%	99.61%
B9	5664	21.53%	100%

2.3. Air Pollution Human Health Risk Assessment

The $AirQ^+$ model has been used to assess the human health risk due to air pollution in Bucharest. $AirQ^+$ is a tool provided by the WHO, built to assist health officials and environmental experts in evaluating the impact of air pollution on health. In our study, we analyzed the effects of exposure to PM_{10}, $PM_{2.5}$, and NO_2 during the three years selected. The software calculated the estimated attributable proportions, estimated number of attributable cases, and estimated number of attributable cases per 100,000 population at risk, taking into account the annual average concentrations for each pollutant and using cut-off values based on emission standards documentation provided by the WHO: 5 $\mu g/m^3$ for $PM_{2.5}$, 10 $\mu g/m^3$ for PM_{10} and NO_2 [48].

Our study focused on mortality analysis for all natural causes for adults over the age of 30. The relative risk (RR) values used were 1.08 for $PM_{2.5}$, 1.04 for PM_{10}, and 1.02 for NO_2 concentrations.

The values related to the total population and the population aged 30 and above in the city of Bucharest, as well as the total number of deaths for each year, were provided by the Romanian National Institute of Statistics [49]. The death incidence was calculated as the ratio between the number of deaths and the total population multiplied by 100,000. (Table 3).

Table 3. Input data for AirQ$^+$ software (https://www.who.int/tools/airq, WHO-EURO: Copenhagen, Denmark).

Year	Population	Over 30	Deaths	Incidence	$PM_{2.5}$ [μg/m^3]	PM_{10} [μg/m^3]	NO_2 [μg/m^3]
2020	2,153,492	1,561,281	26,045	1209	15.2	27.72	29.98
2021	2,161,621	1,554,821	29,972	1386	16.2	26.09	32.79
2022	2,164,506	1,585,489	23,552	1088	15.2	25.83	28.9

3. Results and Discussion

This study presents the assessment of air quality variability in Bucharest, Romania, covering all seasons during three years of measurements, 2020–2022. A particular focus was given to the comparison of the investigated time period including the COVID-19 lockdown period and not, similarly to the work by [50], The trends in the spatiotemporal variability of urban air pollution in Bucharest showed a minimal impact of the lockdown period (as depicted in Supplementary Figures S2–S4); therefore, the full database was considered for further analysis.

The values of the concentrations presented in Table 4 show variability for all three pollutants, PM_{10}, $PM_{2.5}$, and NO_2, over the Bucharest area, but within the same limits for all three years. Significant peaks are observed sporadically in the city center, as measured by the B6 station. The average concentrations are within the same range for all stations and for all three atmospheric compounds. Mean concentrations vary over all stations, with a maximum of 20% for PM_{10}, for example. The maximum loadings for three of the stations (B1, B3, and B6) are considerably high, reaching hourly concentrations of up to 570 μg/m^3 and 644 μg/m^3 for PM_{10}.

Moreover, the other compounds present higher values at these stations, most likely due to their location in the central area of the city and their type (traffic and urban stations, respectively).

By comparing the mean PM_{10} concentrations at background and traffic stations in Bucharest, an increased concentration of PM_{10} in the city center and traffic area can be observed. These differences are significant in all years. An overall decrease in concentrations over the years is seen at traffic stations, while, at industrial ones, there is an increase. Significant differences in mean PM_{10} concentrations are highlighted at the industrial station, B4, possibly due to the proximity to the industrial area in the Berceni district, in the southeast of Bucharest, and a possible effect of the COVID-19 lockdown in 2020.

The target value of 40 μg/m^3 for the annual mean concentration of PM_{10} mentioned in the EU legislation has not been exceeded in Bucharest for the analyzed period. However, the daily mean value concentrations for all stations from the Bucharest area have been exceeded for 22, 17, and 21 days in 2020, 2021, and 2022, respectively. The majority of days exceeding the PM_{10} daily limit concentration are during spring and autumn, seasons characterized by intense anthropogenic activity. Although the air quality EU limits are not exceeded for the PM_{10} annual concentration in Bucharest, the human health EU limit is exceeded during the three years for all sites. Moreover, the $PM_{2.5}$ annual concentration is more than two times the target value of 5 μg/m^3 for human health protection.

The highest concentrations and maximum loadings of $PM_{2.5}$ are noticed in the urban areas, by comparison with industrial and traffic areas, while the mean NO_2 concentrations in Bucharest show increased values at traffic sites, as expected, followed by the industrial ones. At traffic sites, the mean concentrations are almost double those at the background sites. The target value of 40 $\mu g/m^3$ for the annual mean concentration of NO_2 mentioned in the EU legislation has been exceeded in Bucharest only for the traffic sites during the analyzed period. The mean NO_2 concentrations at each station are nearly constant over these three years, with the highest variability being noticed in the city center.

Table 4. Pollutants mean concentration variability at each monitoring station.

Station			B1	B2	B3	B4	B5	B6	B7	B9
PM_{10} [$\mu g/m^3$]	Total	Min	0.3	0.55	0.03	0.23	0.6	0.63	0.3	0.41
		Max	627.36	250.83	570.25	367.55	336.47	644.21	369.61	369.61
		Mean & Std Dev	26.605 ± 20.1	23.96 ± 15.8	24.83 ± 17.4	24.1 ± 15.6	28.13 ± 17.4	29.36 ± 20	25.62 ± 16.9	25.62 ± 24.1
	2020	Mean & Std Dev	27.23 ± 25.7	22.16 ± 15.7	25.91 ± 22.1	15.19 ± 9.2	23.94 ± 17.6	30.94 ± 26.2	24.92 ± 14.5	-
	2021	Mean & Std Dev	26.84 ± 16	24.69 ± 15.9	23.59 ± 13.6	24.03 ± 15.6	26.94 ± 17.2	28.14 ± 16.7	26.05 ± 17	-
	2022	Mean & Std Dev	25.54 ± 17	23.38 ± 15.7	24.98 ± 15.1	24.86 ± 15.7	27 ± 17.6	26.43 ± 14.5	25.18 ± 17	25.62 ± 24.1
$PM_{2.5}$ [$\mu g/m^3$]	Total	Min	0.11	-	-	-	1.72	1.4	0.21	0.11
		Max	234.56	-	-	-	49.79	73.38	187.79	162.37
		Mean & Std Dev	16.63 ± 12.9	-	-	-	13.12 ± 6.9	13.92 ± 7.8	17.83 ± 13.4	15.37 ± 14.5
	2020	Mean & Std Dev	17.7 ± 14.5	-	-	-	15.53 ± 8.8	14.12 ± 9.3	21.94 ± 13.1	-
	2021	Mean & Std Dev	17.1 ± 12.7	-	-	-	13.59 ± 7.4	14.53 ± 7.5	18.95 ± 14.7	-
	2022	Mean & Std Dev	15.47 ±11.2	-	-	-	12.68 ± 6	13.43 ± 5.7	16.54 ± 11.8	15.37 ± 14.5
NO_2 [$\mu g/m^3$]	Total	Min	0.17	5.15	0.71	3.64	4.47	0.33	4.62	0.1
		Max	178.5	161.28	171.68	143.05	177.66	185.04	100.32	108.05
		Mean & Std Dev	25.92 ± 18	29.2 ± 19.6	41.43 ± 21.4	25.47 ± 17.3	31.75 ± 19.4	43.52 ± 23.4	19.1 ± 11.9	25.14 ± 17.9
	2020	Mean & Std Dev	26.97 ± 18.7	27.6 ± 17.3	40.18 ± 20.8	27.29 ± 10	31.15 ±18.7	41.64 ±23.3	19.82 ± 11	-
	2021	Mean & Std Dev	29.38 ±19.8	29.16 ± 19.4	44.79 ± 24.4	25.41 ± 15.9	32.05 ± 19.8	49.16 ± 25.8	18.8 ± 12.5	-
	2022	Mean & Std Dev	22.03 ± 17	29.38 ±19.9	39.31 ± 18.3	25.4 ± 18.8	29.65 ± 19	38.04 ± 19.3	19.34 ± 11.4	25.14 ± 17.9

Figure 1 shows that the mean annual concentration of particulate matter (PM_{10} and $PM_{2.5}$) for the entirety of Bucharest city is almost constant across the years, with smaller variability in 2022. The highest annual concentration and variability of NO_2 is observed during 2021 but within the EC limits.

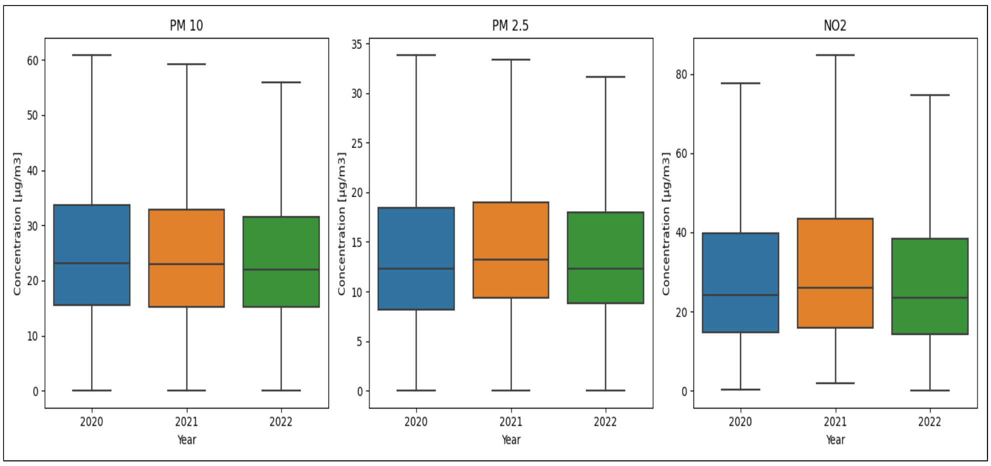

Figure 1. Pollutants' variability for each year between 2020 and 2022.

Figure 2 presents the percentage contribution of the average concentration over the 3-year period of $PM_{2.5}$ and PM_{10}, where the diameter of the circle is given by the value of the concentration and the color of the edge of the circle corresponds to the station type. The highest value of PM_{10} is measured at the traffic-type station, B6, located in the center of the capital, while the lowest concentration of PM_{10} is measured at the industrial-type station, B2. For $PM_{2.5}$, the highest value is measured at the suburban station outside Bucharest, and the lowest value is measured at an industrial-type station, B5.

According to the data of the eight stations, the concentrations of PM_{10} are higher in the west of Bucharest than in the east of the city. Northern Bucharest presents the highest concentrations of large particles. Although the $PM_{2.5}$ data are missing for the stations in the east of Bucharest, B2, B3, and B4, it can be assessed that the stations outside the city or on its outskirts have the highest ratio of fine particles. The Măgurele station (B7) has the highest $PM_{2.5}$ concentration, most probably due to the nearby agricultural areas and specific crops, as already identified by [39,51].

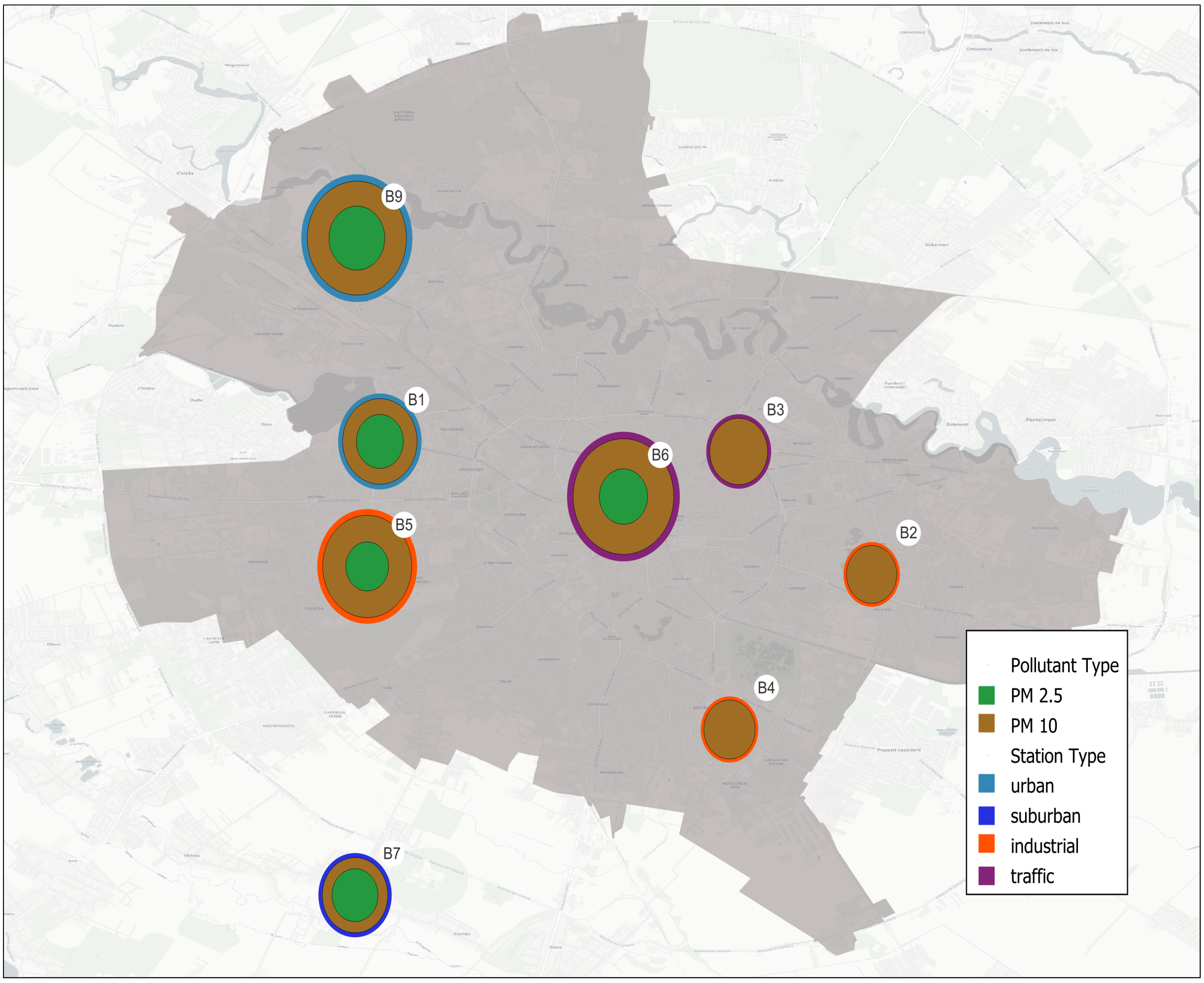

Figure 2. Spatial variability of PM concentration in Bucharest area.

Figure 3 presents the variability of the average NO_2 concentrations in the Bucharest area, with the size of the circle indicating the percentage contribution of the concentration. The highest concentrations of NO_2 are recorded by two monitoring stations, B6 and B3, which are located in the city center, in areas with intense traffic. These stations are placed near major roads or busy intersections, where vehicle exhaust emissions can be significant during the day. Thus, the city center is characterized by high concentrations of NO_2 and consequently increased air pollution.

On the other hand, high average values of NO_2 concentrations are observed also in industrial and urban-type monitoring stations. Emissions from industrial facilities and road traffic may contribute to the higher NO_2 levels in these areas. As expected, low values of NO_2 concentrations are observed in the suburban monitoring station, due to the location in a less dense area in terms of traffic and industrial activity.

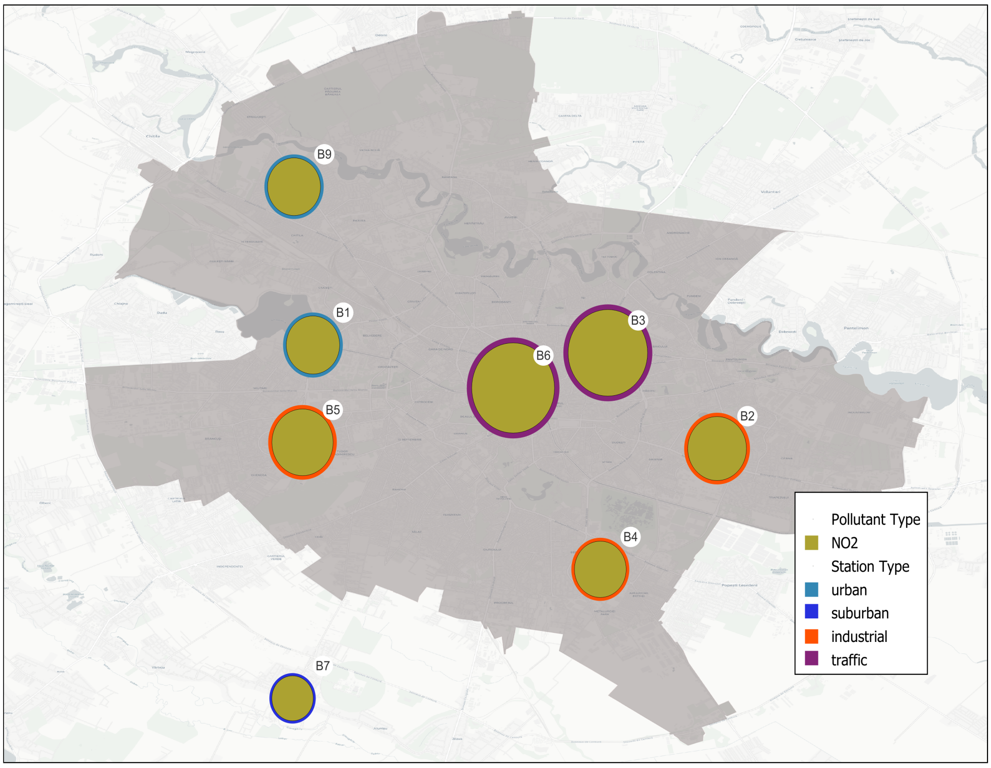

Figure 3. NO_2 concentration variability in Bucharest area during 2020–2022.

The seasonal variation in the fine particle fraction, represented by the ratio between $PM_{2.5}$ and PM_{10} concentrations, is illustrated in Figure 4 for each year, averaged for all stations. An obvious monthly and seasonal trend is noticed, showing higher levels during the cold months than the warm months, mainly corresponding to the increased household heating. Seasonal fluctuations are intricately linked to changes in the height of the planetary boundary layer (PBL), which is directly influenced by variations in air temperature [52–54]. The highest fine particle fraction values occur in winter, with fine particles representing up to 90%, and the lowest values occur in March and September, with fine particle proportions around 20%. The lowest fine particle fraction values may be related to the high values of coarse particles due to frequent long-range dust transport in spring [36,55]. The high values of PM_{10} in March 2020 are most probably due to specific meteorological conditions. High temperatures, a lack of precipitation, and air mass circulation favor the transport of aerosols from anthropogenic sources from Central and Northeastern Europe [56]. Moreover, during spring and autumn, there is an increase in agricultural activity in the rural areas around Bucharest. This may include tillage, harvesting, and the burning of stubble or crop residues. These activities can contribute to increased concentrations of PM_{10} as larger particles such as dust and soil are released into the air. Moreover, the $PM_{2.5}$ concentration increase may be determined by the agricultural activities, since the fine particle fraction is still significant, reaching up to 50%.

The fine particle proportion is smaller during summertime than during cold periods; the warm season is characterized by lower anthropogenic activity, high temperatures, and decreased pollutant emissions, with similar behavior being seen by Fan et al., 2021 [57]. The variation in the fine particle fraction is small from April to mid-September, ranging between 0.4 and 0.6, with the larger particles being predominant. The fraction has similar trends during these three years, with a slightly larger percentage of fine particles during winter–spring 2021 and December 2022. A greater percentage of larger particles was encountered during the spring of 2020.

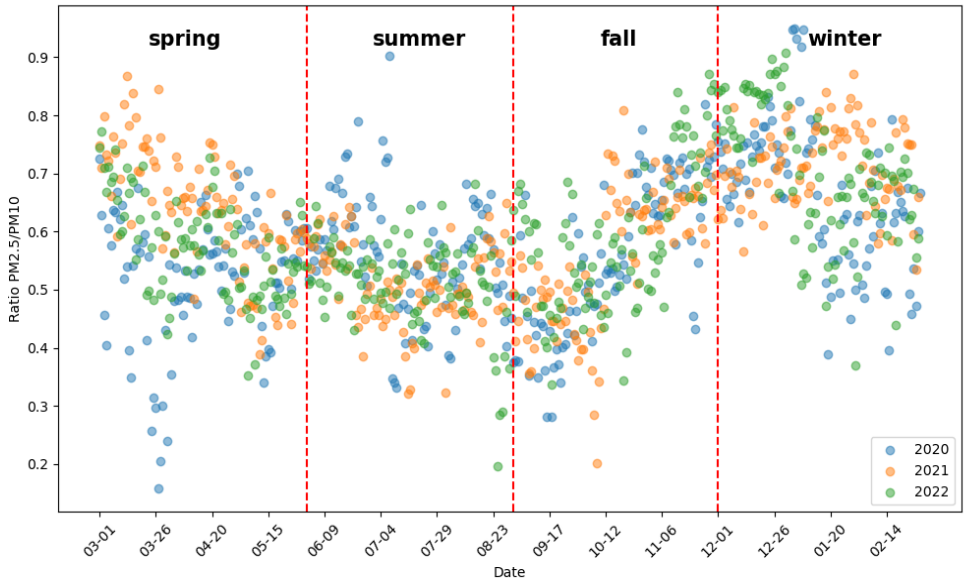

Figure 4. Seasonal variation in the fine particle fraction ratio between $PM_{2.5}$ and PM_{10}.

Figure 5 illustrates the diurnal variation in the three atmospheric compounds along with the temperature during the 3-year period. The air pollutant concentrations at ground level and their dispersion in the troposphere are highly influenced by the planetary boundary layer height. In Bucharest, all air pollutants present a typical bimodal trend, with specific double peaks corresponding to the morning rush hours and nighttime, inversely correlated with the planetary boundary layer height and temperature variability. The diurnal bimodal trend of air pollutants is also characteristic of other cities [28,58].

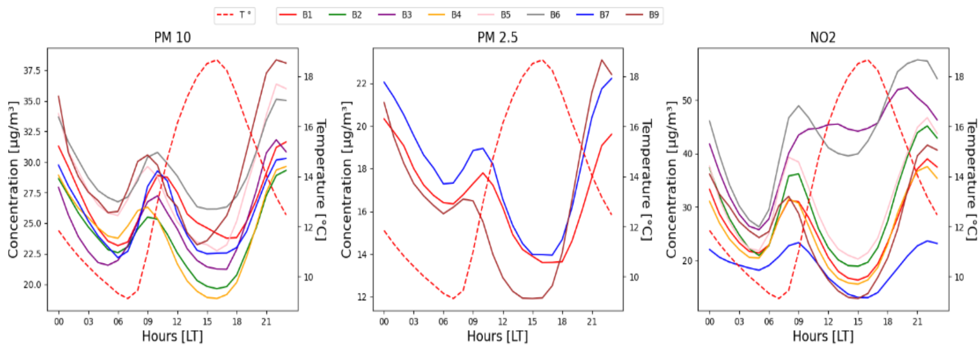

Figure 5. Average diurnal variation in PM_{10}, $PM_{2.5}$, NO_2, and temperature, measured during 2020–2022.

Moreover, the diurnal variation in PM_{10} is strongly influenced by human activities, such as emissions from moving vehicles, construction, and industry, but also other anthropogenic sources. Activities such as heavy road traffic [59], construction work [60], and fossil fuel burning [61,62] lead to the release of large numbers of particles into the air, leading to an increase in the concentration of PM_{10}. During rush hour, when traffic is heavy, the concentrations of PM_{10} can reach higher levels, due to the particle emissions from tire, brake, and road wear [63–65], as well as particles from fuel combustion.

Two distinct morning peaks are noticed, depending on the measurement area. The industrial sites are characterized by a morning peak at around 8 am, while traffic and urban sites are characterized by morning peaks at around 9 am. Nighttime peaks are similar for all measurement sites around Bucharest. The variation in the PM_{10} concentration during the day at the B6 site is smaller relative to the other stations, with the less pronounced peaks highlighting relatively constant traffic and sources during the daytime. A similar diurnal variation is noticed for the $PM_{2.5}$ concentration in Bucharest.

The diurnal variation in NO_2 presents also the bimodal trend, with peaks at 9 am due to the traffic hours and a second peak, more pronounced, at around 7 pm for the traffic stations and 8 pm for the urban and industrial ones. A more pronounced peak in the morning in comparison with the particle diurnal variation points out primary emissions due to traffic exhaust. The surroundings of traffic stations, B3 and B6, contribute to higher exposure to exhaust emissions, including NO_2, which is a byproduct of combustion in internal combustion engines. A less pronounced diurnal pattern is specific to these areas, where the values of NO_2 concentrations are higher during the entire daytime, with small peaks during the traffic hours when the number of moving cars is at its maximum. High variability in NO_2 concentrations in the city center during midday can be observed, with concentration values almost double those for the background and industrial areas.

The diurnal seasonal variation analysis of atmospheric compounds across seasons is shown in Figure 6, highlighting a similar trend across all seasons. PM_{10} concentrations have two peaks around 9 am and around 9 pm, with the highest averages occurring in fall and winter. As for $PM_{2.5}$, the difference between seasons is much more pronounced, with values in winter almost twice as high as in the summer period, indicating a strong influence of household activities and home heating. A similar trend across seasons is evidenced in the case of NO_2 for the morning peak, while, starting in the afternoon, the winter and fall present a more pronounced peak, also identified in another city in Romania [66].

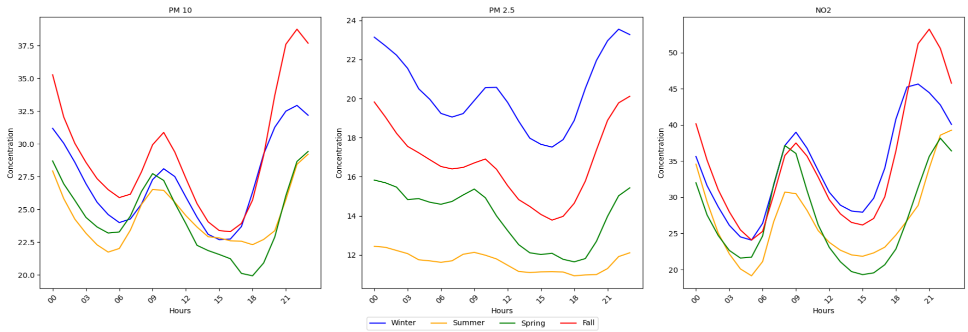

Figure 6. Average diurnal seasonal variation in PM_{10}, $PM_{2.5}$, and NO_2, measured during 2020–2022.

In general, the levels of PM exhibited a weekly pattern, with lower concentrations during the weekends in comparison to weekdays (Figure 7). This weekend effect was more pronounced for PM_{10} compared to $PM_{2.5}$. During weekdays, the daily average concentrations of PM_{10} and $PM_{2.5}$ ranged from 23.3 to 33.06 μg/m^3 and 13.64 to 17.98 μg/m^3, respectively. On the other hand, during weekends, the concentrations were lower, in the ranges of 20.33 to 30.16 μg/m^3 for PM_{10} and 13.22 to 17.76 μg/m^3 for $PM_{2.5}$. A similar pattern of a weekly effect in the case of particulate matter has been observed in Bucharest and, as depicted by Jiao et al. (2023), in France [28]. The weekday distribution indicates two distinct peaks in the data, corresponding to the morning rush hours and nighttime anthropogenic activities. On the other hand, the weekend distribution showed a less pronounced two-peak diurnal pattern, with one during nighttime, when household heating was likely the dominant factor influencing the PM_{10} and $PM_{2.5}$ concentrations. The second peak was less pronounced due to a smaller effect during the morning. An overall lower average concentration on weekends than on weekdays is noticed, as depicted also in several cities [28,67].

Compared to weekdays, the significant decrease in PM_{10} levels observed during weekends indicates a reduction in traffic, sources from fossil fuel combustion, and road dust. The levels of fine particles ($PM_{2.5}$) did not show significant reductions on weekends compared to weekdays, particularly during nighttime. The findings suggest that anthropogenic activities play a significant role in influencing the local air quality in the area, as also noticed in other cities [28]. The ratios of $PM_{2.5}/PM_{10}$ are increased from weekdays to weekends, indicating a potential contribution from anthropogenic sources to the elevated fine particle concentrations during weekend periods.

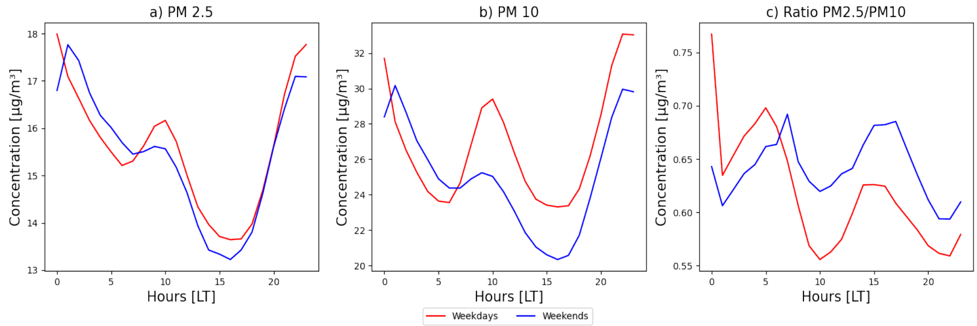

Figure 7. Weekday versus weekend average daily variation for PM_{10}, $PM_{2.5}$, and ratio of $PM_{2.5}/PM_{10}$.

The analysis of the estimated proportion of mortality attributed to annual exposure to air pollutants above the WHO threshold provides valuable insights into the public health impact of air pollution in Bucharest. The findings, illustrated in Figure 8, explain the varying degrees of health risks associated with different air pollutants.

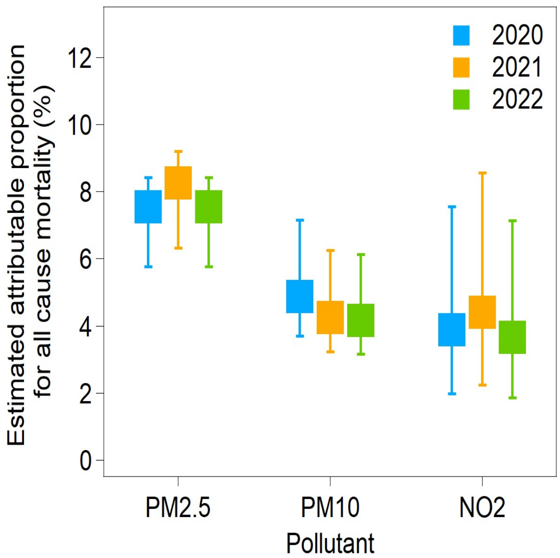

Figure 8. Estimated attributable proportion of all-cause mortality for PM_{10}, $PM_{2.5}$, and NO_2 each year.

Looking at $PM_{2.5}$, the results reveal an alarming proportion of mortality ranging from 7.55% to 8.26%. This means that a significant percentage of deaths in Bucharest are caused by long-term exposure to $PM_{2.5}$ concentrations above 5 $\mu g/m^3$. The estimated number of attributable cases, ranging from 1302 to 1425 within the target population, highlights the severity of the issue, with the highest proportion observed in 2021. Nevertheless, the mortality rates attributed to $PM_{2.5}$ concentrations are much lower than those indicated by recent studies performed in highly polluted areas [68,69] and other European cities [70,71].

In contrast, the proportion of mortality attributed to long-term exposure to PM_{10} above 10 $\mu g/m^3$ is notably lower, ranging from 4.16% to 4.87%. The corresponding estimated number of attributable cases, between 717 and 917 within the target population, indicates a less severe health impact compared to $PM_{2.5}$. This clear difference between the $PM_{2.5}$ and PM_{10} impacts highlights the importance of considering the particle size when assessing the health risks associated with air pollution.

It is worth noting that the lowest proportion of mortality is associated with NO_2 concentrations above 10 $\mu g/m^3$, ranging from 3.67% to 4.41%. This suggests that, among the pollutants considered, NO_2 poses a comparatively lower risk of mortality. However, the estimated number of attributable cases, ranging from 633 to 950 within the target population, still emphasizes the need for measures to mitigate the health impact of NO_2 exposure.

4. Conclusions

This variability study provides a detailed and comprehensive analysis of air quality fluctuations in Bucharest, Romania, over a three-year period from 2020 to 2022, encompassing all seasons. For this study, measurements of PM_{10}, $PM_{2.5}$, and NO_2 performed at eight stations of the Romanian National Air Quality Monitoring Network located in Bucharest were used.

Regarding the levels of particulate matter, it was observed that the western part of the city experiences a greater impact. Conversely, concerning NO_2 pollution, the central area of the city is more significantly influenced.

The investigation revealed notable variations in the particulate matter and nitrogen dioxide concentrations across the Bucharest metropolitan region. Although the concentrations were within a similar range, specific increases were identified, predominantly in the city center. The results showed that air pollutants exhibit a characteristic bimodal pattern throughout the day, characterized by distinctive dual peaks corresponding to the morning rush hour and nighttime hours.

The investigation revealed the significant spatial variability in NO_2 concentrations. The concentrations in the city center during midday were almost double the ones observed in the background and industrial areas. Additionally, a discernible weekly pattern was noticed for particulate matter concentrations, with reduced levels during weekends as opposed to weekdays. This trend was more pronounced in the case of PM_{10} than $PM_{2.5}$ concentrations.

The seasonal variation in the ratio of $PM_{2.5}$ and PM_{10} highlighted the predominance of fine particles in the atmosphere during wintertime, due to the more intense activity of electro-thermal plants and apartment heating.

The analysis of the mortality rates associated with annual exposure to air pollutants in Bucharest above the thresholds set by WHO highlighted the larger proportion of mortality due to $PM_{2.5}$, which is alarming and indicates a significant impact on public health. On the other hand, PM_{10} and NO_2 have a lower impact on mortality, around 4%, indicating lower health risks.

In a broader context, this study significantly contributes to our understanding of air quality dynamics. It provides spatial insights into the distinct contributions of NO_2 and particulate matter across different areas within the city, while also emphasizing temporal variations during the day and changes over the seasons. These findings underline the need for targeted interventions aimed at mitigating air pollution, particularly in urban centers, and emphasize the importance of adopting seasonal adjustments to effectively manage air quality and protect public health.

Supplementary Materials: The following supporting information can be downloaded at https://www.mdpi.com/article/10.3390/atmos14121759/s1. Figure S1: Average diurnal variation of CO, SO_2 and O_3, measured during 2020–2022; Figure S2: Weekend effect of average daily variation without the COVID-19 lockdown for PM_{10}, $PM_{2.5}$ and Ratio $PM_{2.5}/PM_{10}$; Figure S3: Average diurnal seasonal variation of PM_{10}, $PM_{2.5}$ and NO_2, measured during 2020–2022 without the COVID-19 lockdown period; Figure S4: Average diurnal variation of PM_{10}, $PM_{2.5}$, NO_2 and temperature, measured during 2020-2022 without the COVID-19 lockdown period.

Author Contributions: Conceptualization, A.I. and J.V.; methodology, A.I. and J.V.; software, A.I.; validation, A.I., J.V., C.T. and C.I.; formal analysis, A.I.; investigation, A.I. and J.V.; writing—original draft preparation, A.I. and J.V.; writing—review and editing, A.I., J.V., C.T., C.I. and A.N.; visualization, A.I. All authors have read and agreed to the published version of the manuscript.

Funding: This work was carried out through the Core Program within the National Research Development and Innovation Plan 2022–2027, carried out with the support of MCID, project no. PN 23 05; the Romanian Ministry of Research, Innovation and Digitalization, through Program 1—Development of the national research-development system, Subprogram 1.2—Institutional performance—Projects to finance the excellent RDI, Contract no. 18PFE/30.12.2021; the European Regional Development Fund through the Competitiveness Operational Programme 2014–2020; by the project Strengthen the participation of the ACTRIS-RO consortium in the pan-European research infrastructure ACTRIS, ACTRIS-ROC, MYSMIS code 107596 (ctr. no.337/2021); and by the RI-URBANS project (Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban & Industrial Areas, European Union's Horizon 2020 research and innovation program under grant agreement, contract 101036245).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The air quality data are available at https://www.calitateaer.ro/public/home-page (accessed on 9 October 2023).

Acknowledgments: We gratefully acknowledge the National Air Quality Monitoring Network (RNMCA), part of the Ministry of Environment, for providing essential air quality data, which significantly enriched our research. Their commitment to monitoring and sharing air quality information is invaluable, and we appreciate their contribution to our efforts in the characterization of urban air pollution, as well as the anonymous reviewers and editors for their time and effort to improve the paper. We wish to express our sincere appreciation to AirQ+, developed by the World Health Organization, for its invaluable contribution to the methodologies and data used in this work. The robust tools and insights provided by AirQ+ significantly enriched the depth and quality of the research presented. The authors also gratefully acknowledge the Romanian Institute of Statistics for providing invaluable data essential to this research.

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

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