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Air Quality Integrated Assessment: Environmental Impacts, Risks and Human Health Hazards

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Abstract: The monitoring and evaluation of air quality is a topic of great global interest as, with the decline of air quality, there are negative effects on human health and ecosystems. Thus, the purpose of this paper was to evaluate the air quality over 11 years, in the period 2011–2021, in four cities in Romania, reported as most polluted, namely, Brasov, Cluj-Napoca, Iasi, and Timisoara. Pollutants of interest included arsenic, carbon monoxide, and $PM_{2.5}$. The measured concentrations of the selected pollutants were collected from the National Environmental Protection Agency public reports. The database considered the daily measurements for the selected pollutants, from three monitoring stations in each city so that the air quality and trends for the last 11 years and impact assessment could be developed. Therefore, the input data were statistically analyzed to identify the trends of air quality, and then, on this basis, the environmental impacts and risks and health hazards were quantified. High concentrations of $PM_{2.5}$ were recorded for Iasi city, while for Timisoara city, significant concentrations of arsenic were reported. The results regarding the air quality aggregate index, air pollution index, and health hazard index were in the regular range, but in the case of sensitive, vulnerable targets such as children, they were triple compared to adults. The results show that the alert threshold value for $PM_{2.5}$ was exceeded every year in all four cities, while in the case of Timisoara city, the arsenic air pollution proved to be at a significant level with a major risk for human health.

Keywords: air quality assessment; integrated approach; impact; risk; health hazards

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1. State of the Art

Air is an essential environmental factor, and by changing its quality, negative effects can occur in ecosystems, at the same time generating an impact on human health. Thus, air pollution has become a pressing problem globally, and monitoring, maintaining, and even improving the air quality are topics of great interest to specialists because air can quickly transport certain amounts of pollutants, which lead to environmental damage and negative effects on human health [1]. Therefore, air quality standards aim to protect health and the environment through various regulations, including threshold limits for air pollutants [2]. At the European level, there are several laws and directives regarding the quality of the environment, targeting the monitoring presence of the polycyclic aromatic hydrocarbons, mercury, arsenic, nickel, and cadmium in the air (Directive 2004/107/EC; 2008/50/EC, Decision 2011/850/EU), and establishing a common international framework for air quality protection [3]. Additionally, there are various strategies to protect air quality, such as the "Clean Air Policy Package", published in 2013, which aims to significantly reduce air pollution in Europe by 2030 and includes a set of much stricter emission standards [4]. The Green Deal plan, presented in 2019, comprises a set of initiatives at the European Union (EU) level. The aim of this plan is to make Europe a neutral continent in terms of pollution and climate change by 2050 [5].

In Romania, the basis of the air quality legislative framework is strictly in accordance with the EU regulations, and the main legislation is Law no. 104/2011 [6]. At the international level, one example is the Clean Air Act, a specific US law that sets several standards for environmental quality and air pollutant emissions [7]. Comparing the EU and US air quality legislative framework, it can be observed that the EU's strategy is oriented to limit methane emissions, while the US is pursuing a green economy, but both are focused on climate change limitation. The European legislative framework, unlike that of the US, is based on the precautionary principle and pollution prevention, so that the ecological balance and human health can be assured [8]. To properly implement the pollution prevention principle, the air quality assessment process must include accurate information on air pollutants type, dispersion processes, and physical and chemical reactions, and the monitoring data used must be in accordance with the environmental standards. Thus, accurate data on air quality and air pollution levels can be delivered. There are several definitions of air quality monitoring activity, but the simplest indicates that this activity can be characterized by air sampling and analysis using analyzers or sensors [9]. Air quality monitoring methods are diverse and may include the Holo SpeckleNet tool [10], the improvement of some sensors [11], or the use of low-cost sensors and Internet of Things systems [12,13].

Another topic of great interest in the world of research is the development of tools for modeling the dispersion of air pollutants. In this field, specialists try to combine various approaches to reach better results, such as combining the SINDy algorithm with Lasso regression [14] or using the Caliope system together with principal component analysis (PCA) [15]. Air quality modeling has evolved into a new discipline that includes various technologies and sciences from disciplines such as mathematics, environmental statistics, meteorology, physics, and air quality, which makes this system increasingly complex and advanced. Over time, air quality models have been classified into global, regional, and urban models, and knowing continuous advance, at the international level, small- and medium-scale regulated models such as AERMOD, ADMS, ISC3, etc., are currently used. Among the regional models are CMAQ, WRF-CHEM, CAMx, etc., and GEOS-CHEM is the best-known global model. Among those mentioned, according to the US Environmental Protection Agency, the most-used models belong to the following categories: photochemical, dispersion and receptor models. Although air quality modeling has evolved rapidly, there are still problems in its application, caused by limiting factors such as unusual emissions or meteorological factors [16,17]. In this regard, a series of patents have been published in Romania referring to devices for air purification and salinization [18], methods for reducing pollutants in industrial flue gases by combining ionizing radiation with non-ionizing radiation [19] or with fine water droplets [20], methods for determining the tritium concentration in the air [21], or wet desulfurization of waste gases [22]. Internationally, there are several patents in the field of air quality, and some include methods for the real-time monitoring of suspended particle concentrations [23], removal of air pollutants [24], or fusing of fire sensors [25], while others refer to monitoring systems based on improved chemical spectrophotometric sensors [26] or automatic air sample analysis [27].

Based on all aspects previously mentioned, it is generally agreed that air quality monitoring is extremely important to prevent damage to environmental quality and human health and to improve air quality as well [28,29]. The purpose of this work was to prove that even though the air quality indicator measurements are under the maximum allowed concentrations, according to the environmental standards, there are remaining high risks in the case of children's exposure to certain pollutants. To our best knowledge, this is the first study in Romania that considers 11 years of air quality measurements, based on which an integrated approach to impact, risk, and health hazard assessments are developed. Having all these aspects in mind, the objectives of this paper were (1) to evaluate, over the last 11 years (2011–2021), the air quality of four cities in Romania, considering the daily measurements of three monitoring stations in each city for three pollutants: $PM_{2.5}$, As, and CO, (2) to assess the air quality using the air pollution index, and (3) to assess the impacts, risks, and health hazards. Depending on the toxicological effects and the major impacts on human health, three different pollutants were selected, namely, one in the form of gas (CO), one in the form of dust ($PM_{2.5}$), and one heavy metal (As), all of them with high toxicity. Suspended particles are very dangerous (especially $PM_{2.5}$), managing to penetrate deep into the respiratory system, thus causing major health problems, such as cancer and cardiovascular or neurodegenerative diseases, as well as playing a role in viral infections. Current data claim that there are approximately four million deaths caused by cardiopulmonary diseases, with the inhalation of $PM_{2.5}$ as the triggering factor. One of the most recent studies published in 2022 describes the mechanisms and pathways of $PM_{2.5}$, as well as the toxic effects of this pollutant, along with the causal associations between health and suspended particles [30]. Arsenic, an inorganic priority pollutant with high toxicity, can be found among the finest particles of $PM_{2.5}$ and has carcinogenic effects. At the same time, heavy metals can bioaccumulate in the human body, and they have a low degradation potential and a high degree of toxicity. One of the latest studies draws attention to the high potential of this pollutant to cause cancer and highlights the negative effects on the digestive and cardiovascular systems, as well as DNA [31]. In the case of carbon monoxide, among other negative aspects, it is well-known for its destructive effects on the atmosphere, contributing to global warming. The toxicity of this pollutant consists in its reaction with hemoglobin, which results in carboxyhemoglobin, causing serious health problems, including poisoning and even death from hypoxia [32].

2. Materials and Methods

2.1. Site Boundaries and Brief Description

The National Air Quality Monitoring Network performs continuous measurements of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), ozone (O_3), particulate matter (PM_{10} and $PM_{2.5}$), benzene (C_6H_6), lead (Pb), arsenic (As), cadmium (Cd), nickel (Ni), and benzo(a)pyrene, and it includes 148 automatic air quality monitoring stations and 11 mobile stations distributed as follows: 30 traffic-type stations, 58 industrial stations, 37 urban background-type stations, 13 suburban-type stations, 7 stations of the regional background type, and 3 EMEP-type stations. By region, the monitoring stations cover 20 automatic stations in the North-East Region, 21 in the South-East Region, 27 in the South-Muntenia Region, 12 in the South-West Oltenia Region, 21 in the West Region, 18 in the North-West Region, 21 in the Central Region, and 8 in the Bucharest-Ilfov region [6]. In the last two years, several charts have been produced for the most polluted cities in Romania, and each time, among other cities, Iasi, Brasov, Cluj-Napoca, and Timisoara were found to be the most polluted. Table 1 presents the similarities of the four selected cities for this study.

Table 1. Similarities between the evaluated cities [33,34].

City	Population	Number of Water Treatment Plants	Number of Wastewater Treatment Plants	Number of Air Quality Monitoring Stations
Brasov	275,514	2	5	7
Cluj-Napoca	303,047	1	8	5
Iasi	318,871	8	4	6
Timisoara	306,462	3	1	7

The monitoring stations considered for this study are presented in Figure 1.

Figure 1. Location of air quality monitoring stations.

Brasov is a tourist city in the center of the country and has 7 air quality monitoring stations, of which we chose to take the data recorded by 3 since they have the most complete set of measurements. BV-2 is an urban-type station and BV-4 is a suburban-type station, meaning that they aim to monitor the influence generated by pollutants within a radius of 1–5 km. The third station is EM-1, an EMEP-type station that deals with monitoring long-distance air quality in a cross-border context, and it is located at 1231 m elevation [6,34]. Cluj-Napoca, in the northwest of the country, is also called the "heart of Transylvania" and is also a tourist city. Air quality monitoring is carried out through 5 stations. For this area, we chose the CJ-1 station (traffic type), which, as its name suggests, registers the influences generated by traffic on air quality, and CJ-2 and CJ-3, which are urban and suburban types, respectively [6,34]. *The city of the 7 hills*, or the city of Iasi, is in the northeast of the country, and within its territory, there are 6 air quality monitoring stations. IS-2 and IS-5 are urban and suburban types, respectively, and IS-6 is a rural-type station [6,34]. To the west of the country is Timisoara, a city known for its commercial, industrial, cultural, medical, and university activities. There are 7 stations here, of which TM-1 is a traffic-type station, TM-2 is an urban-type station, and TM-3 is a suburban-type station [6,34].

All monitoring stations are equipped with high-performance analyzers that continuously monitor selected air pollutants, including $PM_{2.5}$, As, and CO. The measurement methods used to determine pollutant concentrations are the reference methods, according to the EU environmental standards, as follows: for CO, the determination method is non-dispersive infrared spectrometry, and for $PM_{2.5}$, it is gravimetric and nephelometric determination. The reference method for measuring arsenic is in accordance with the SR EN 14902 standard, specifying absorption spectrometry with atomic or coupled plasma [34].

2.2. Experimental Data

The air quality assessment was performed based on the data from the last 11 years, considering 3 pollutants: arsenic, $PM_{2.5}$, and carbon monoxide. Arsenic, which is part of the heavy metal category, can be found in $PM_{2.5}$ fractions that are known to be very dangerous and can enter the respiratory tract. Carbon monoxide, a toxic gas, can be lethal in high concentrations, reducing the ability to carry oxygen in the blood [6]. Their measured concentrations for the last 11 years (2011–2021) in the 4 cities were obtained from the EPA public reports. For each of these cities, we considered the daily data recorded by 3 monitoring stations. To better observe the evolution of the air quality and its trend during the 11 years, statistical analyses were run, considering the minimum and maximum concentrations measured within each year. Where there are no data presented in the figures for a specific pollutant, year, or monitoring station, there were no measurements included in the report.

After a deep analysis of the information reported, the data were organized into tables, for each monitoring station, year, and pollutant, for each city, resulting in 4 complex tables. For each pollutant, the annual averages measurements, and the maximum and minimum concentrations were considered as well. Based on these tables, we also produced the first set of graphs that show the average concentration for each year, station, and pollutant. The errors could not be calculated for this dataset because the values were taken from the reports validated by the Environmental Protection Agency (EPA).

Figure 2 shows the evolution of air quality in the four cities during the 11 years (2011–2021), depending on the average annual concentration of $PM_{2.5}$. Through this representation, the evolution and variability of the values can be clearly visualized. For each city, there were concentrations recorded by only one monitoring station, as can be seen in Figure 2. Thus, in the case of Brasov, a downward trend of the concentrations recorded in recent years can be observed, reaching a maximum of 25.89 [$\mu g/m^3$] in 2017, exceeding the legal limit for this year. This value was registered by the BV-2 station. The evolution of air quality in terms of particulate matter pollution for Cluj-Napoca was monitored by the CJ-2 station. Thus, a maximum annual average was reached in 2011, of 28.32 [$\mu g/m^3$], also exceeding the legal limit calculated for one year. Regarding the city of Iasi, the trend of decreasing concentrations in recent years has been absent for $PM_{2.5}$, with variation in the values between 20 and 25 [$\mu g/m^3$], and with the highest concentration recorded for this pollutant among the four cities (28.70 $\mu g/m^3$) in the 11 years. The lowest values among the 4 cities were registered in Timisoara, being between 9.94 and 21.50 [$\mu g/m^3$], in 2016. All four monitoring stations that recorded $PM_{2.5}$ concentrations are urban types and are in the vicinity of residential areas.

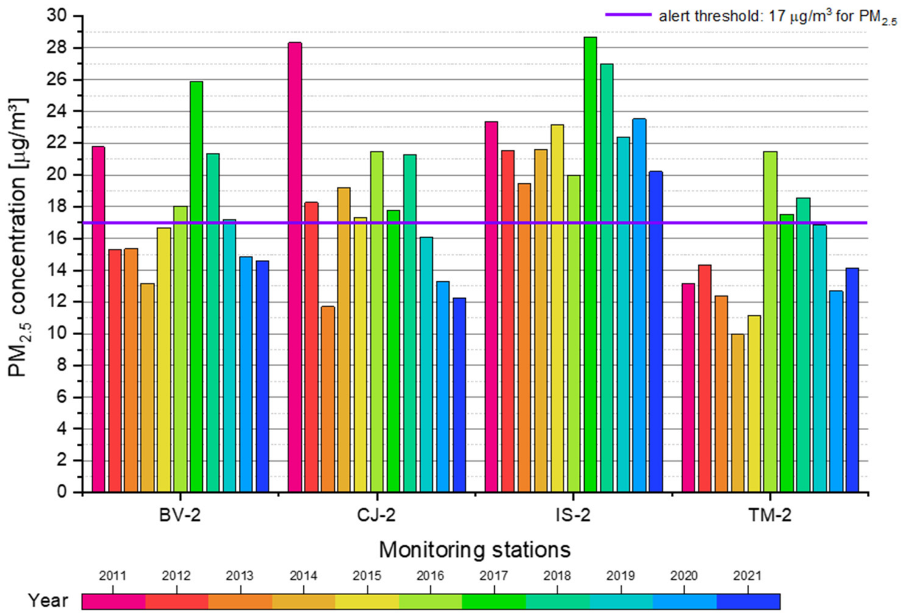


Figure 2. Average annual concentration of $PM_{2.5}$ in the four cities (Brasov, Cluj-Napoca, Iasi, and Timisoara).

Regarding the average annual concentrations of carbon monoxide in the case of Brasov, Timisoara, and Cluj-Napoca (Figure 3), the values did not remain in a constant range, varying depending on the location of the stations, but within normal limits. For Iasi, the data from the IS-6 monitoring station were highlighted, which were higher than those from the other two stations. Even if there were some variations, the values were within normal limits for this city as well, with a maximum of 1.32 [mg/m^3] in 2014.

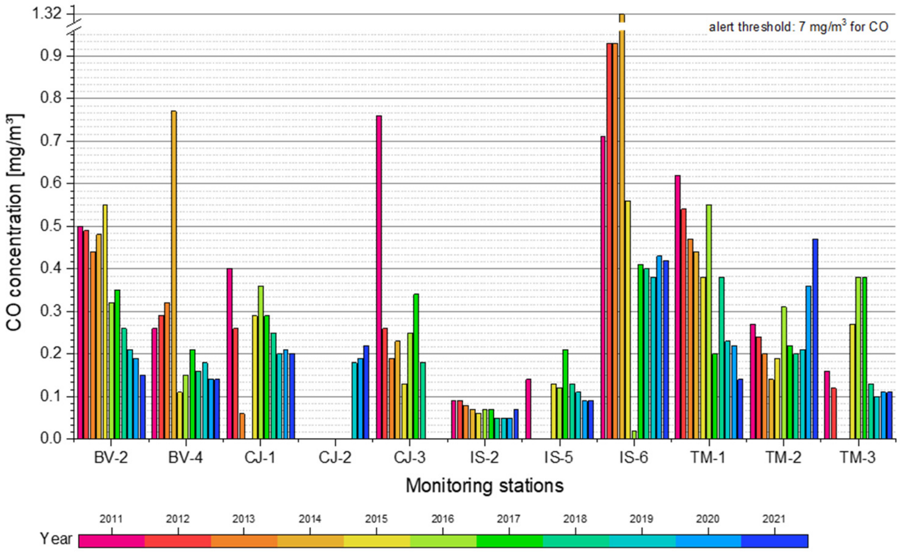


Figure 3. Average annual concentration of CO in the four cities (Brasov, Cluj-Napoca, Iasi, and Timisoara).

The representation of the average annual concentration for arsenic (Figure 4) shows that in Brasov, the stations recorded concentrations within normal limits starting in 2020; until then, there were no reported values. The same situation applies to Iasi, but here, the first year concentrations were reported was 2017, increasing slightly every year, and in 2021, there were no significant data. Moving on to Cluj-Napoca, as can be seen in Figure 4, no significant values were highlighted until 2016. After this interval, variations in concentrations were recorded, suddenly rising to 2.59 [ng/m^3] in 2020, and in 2021, the values became insignificant again. A more special situation is presented by the report on arsenic pollution in Timisoara because, unlike the other 3 cities, arsenic concentrations were recorded here every year, between 0.34 and 1.45 [ng/m^3].

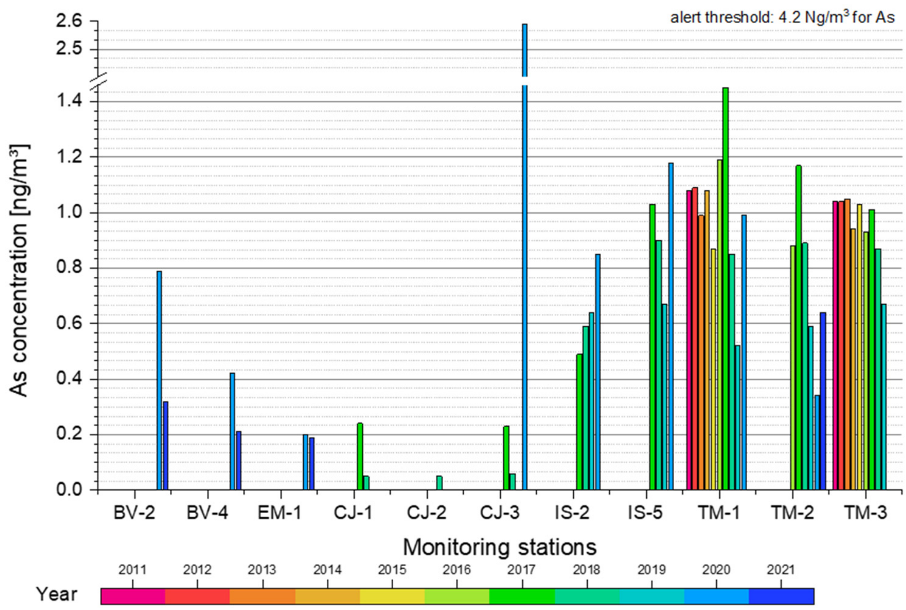
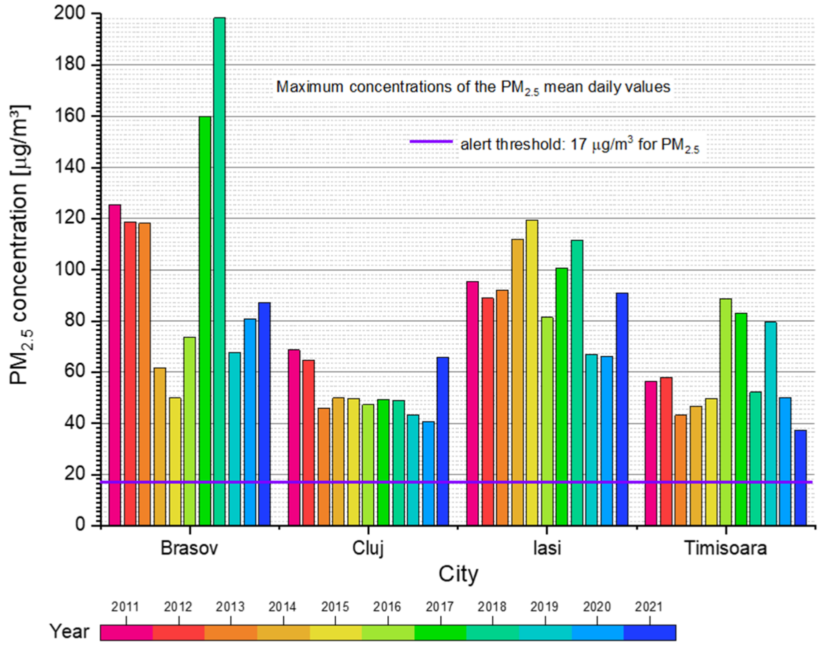


Figure 4. Average annual concentration of As in the four cities (Brasov, Cluj-Napoca, Iasi, and Timisoara).

Furthermore, the maximum and minimum concentrations of each year, for each city and pollutant, depending on the alert threshold, are represented. Figure 5 shows the values for $PM_{2.5}$, the threshold value being 17 [µg/m^3]. As can be seen, in the case of all 4 cities, the maximum values for each year exceeded the alert threshold. A constant trend can be noticed for Cluj-Napoca, Iasi, and Timisoara, but for Brasov, there are variations from 49.87 [µg/m^3] to 198.31 [µg/m^3]. For Cluj-Napoca and Timisoara, the concentrations were lower, unlike in Iasi, where $PM_{2.5}$ pollution ranged between 80 and 120 [µg/m^3]. The minimum annual measured concentrations were below the alert threshold value for each case.



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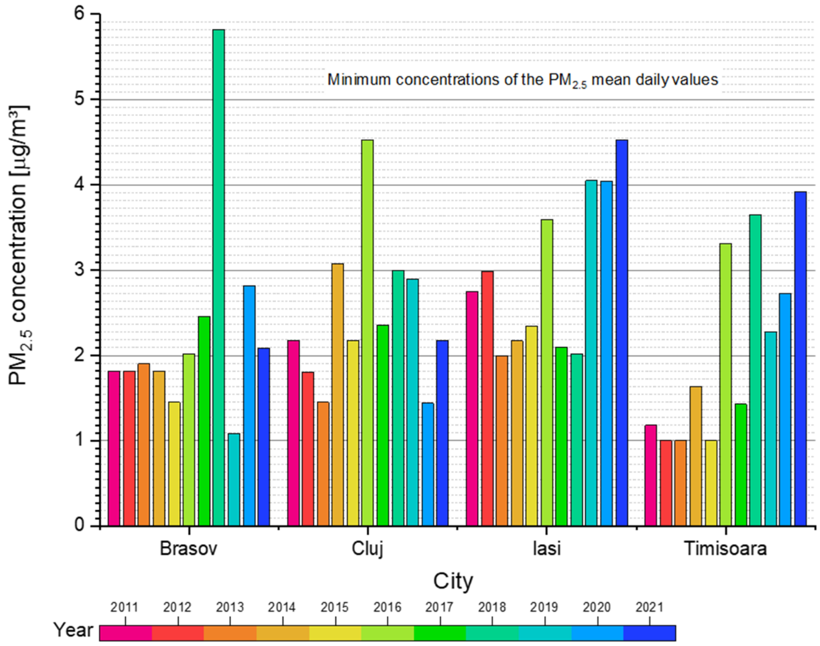
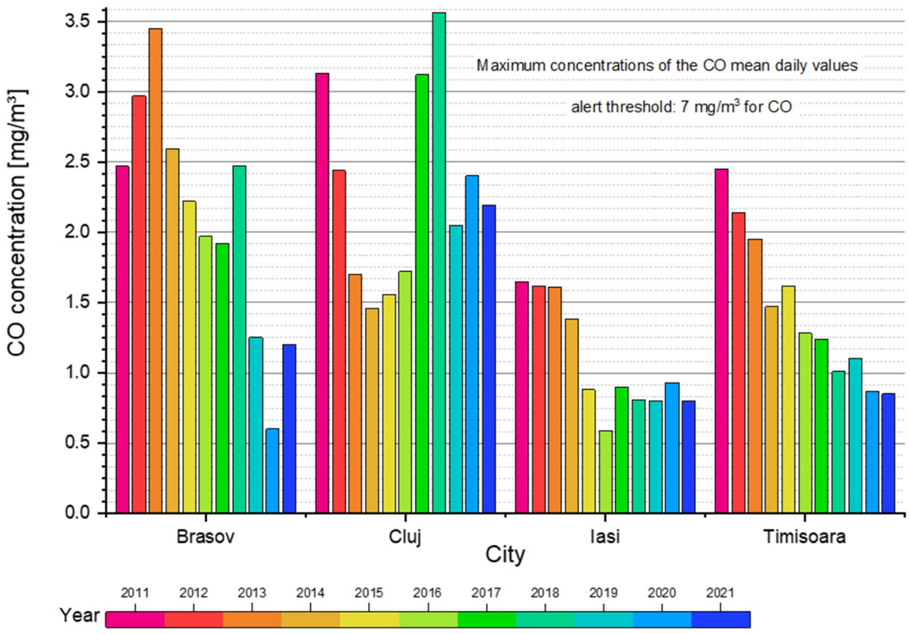


(**b**)

Figure 5. (**a**) Maximum concentrations of $PM_{2.5}$ mean daily values depending on the alert threshold; (**b**) minimum concentrations of $PM_{2.5}$ mean daily values depending on the alert threshold.

The carbon monoxide alert threshold sets a value of 7 [mg/m^3], which was not exceeded in any of the four cities during the last 11 years (2011–2021). The concentrations remained in a constant line, with there being several variations in Brasov and Cluj-Napoca, the maximum concentrations being 3.45 [mg/m^3] and 3.56 [mg/m^3], respectively. Additionally, in the case of the cities of Iasi and Timisoara, the trend of decreasing values in recent years can be best observed (Figure 6).



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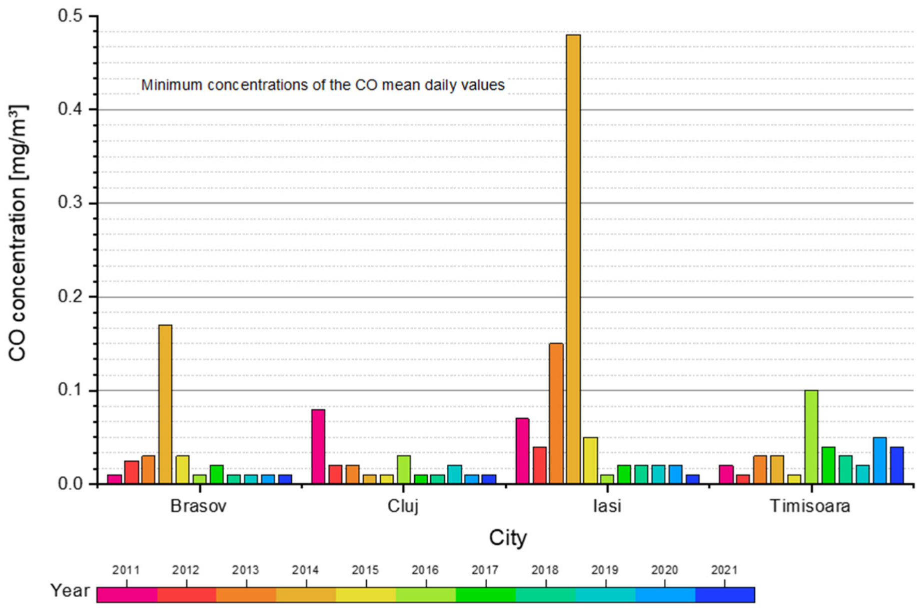
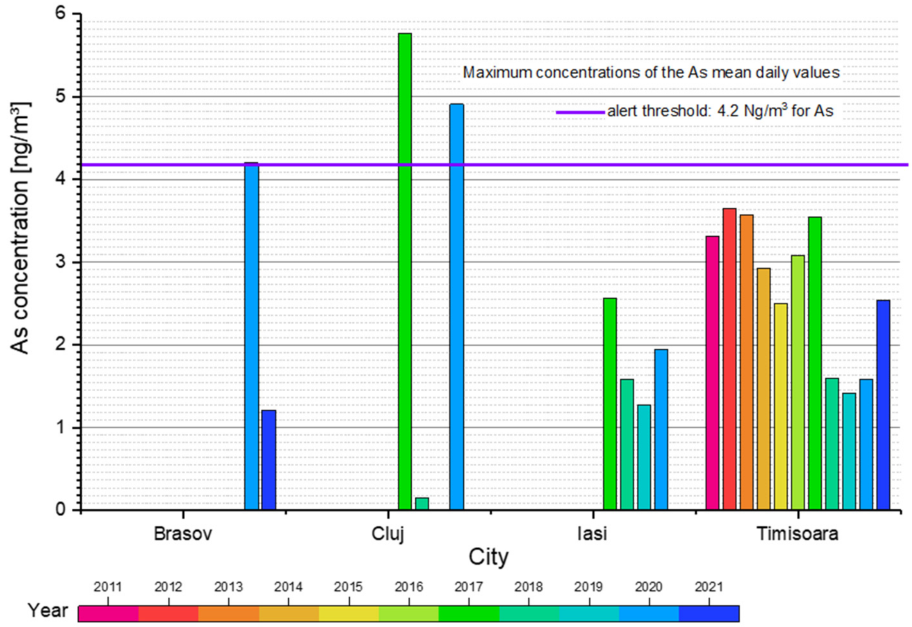


(**b**)

Figure 6. (**a**) Maximum concentrations of CO mean daily values depending on the alert threshold; (**b**) minimum concentrations of CO mean daily values depending on the alert threshold.

Regarding arsenic, a pollutant whose alert threshold is 4.2 [ng/m^3], there were only two situations in which this number was exceeded, namely, in Cluj-Napoca in 2017 and 2020, with concentrations of 5.76 [ng/m^3] and 4.91 [ng/m^3], respectively. In the other 3 cities, the alert threshold limit was not exceeded. As can be seen in Figure 7, a special situation was highlighted in Timisoara, where arsenic was present every year and the concentrations recorded varied, increasing to 2.54 [ng/m^3] in 2021, compared to 1.58 [ng/m^3] in 2020.



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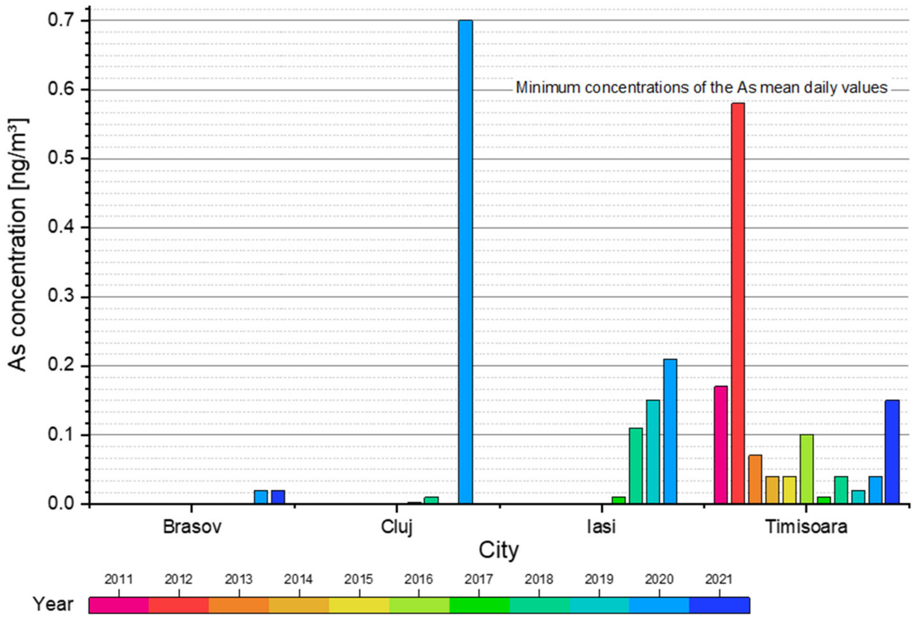
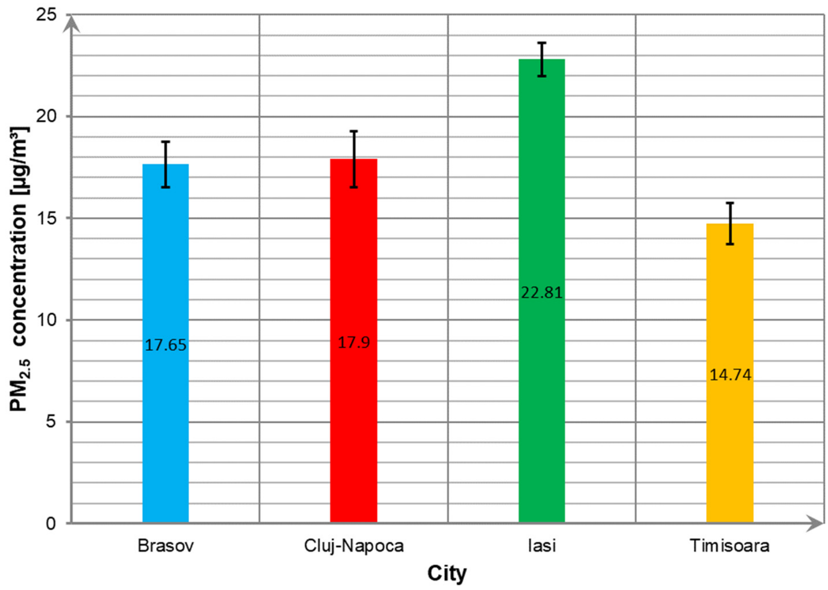


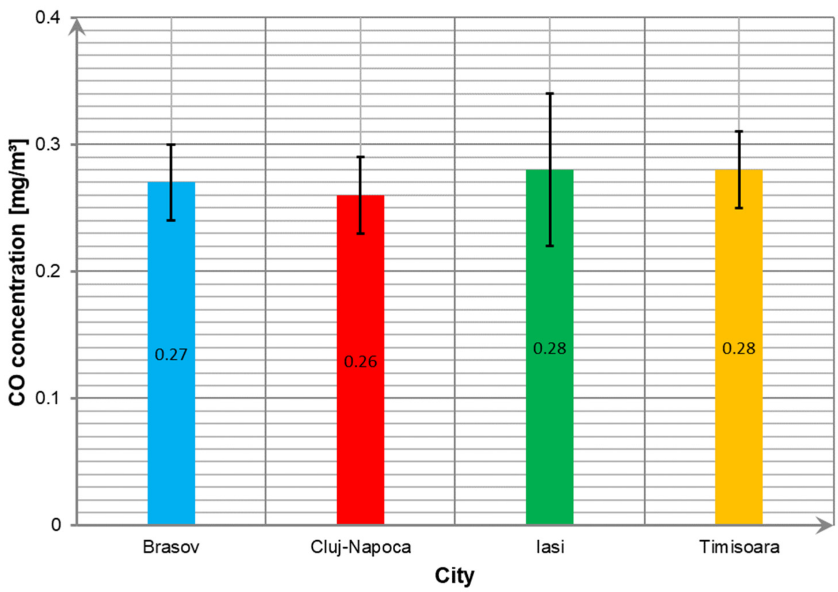
(**b**)

Figure 7. (**a**) Maximum concentrations of As mean daily values depending on the alert threshold; (**b**) minimum concentrations of As mean daily values depending on the alert threshold.

The last type of graph was aimed at highlighting the predominant pollutant for each city by calculating the aggregate index, which represents the average of the concentrations for all 11 years (2011–2021), considering all the values from the previously mentioned monitoring stations [29,35]. According to Figure 8, for Iasi, the most significant values of $PM_{2.5}$ were reported, the index level being 22.81 ± 0.83 [µg/m^3]. This value is higher than that obtained in the other 3 cities because higher concentrations of the annual values were highlighted in 2017 (28.7 µg/m^3) and 2018 (27 µg/m^3); at the same time, there were also exceedances of the alert threshold value for $PM_{2.5}$, as observed in Figure 5. In the case of carbon monoxide, the values were close, with a maximum of 0.28 ± 0.06 [mg/m^3] in Iasi and Timisoara and a minimum of 0.26 ± 0.03 [mg/m^3] in Cluj-Napoca. For this pollutant, the values were normal, without sudden variations or exceedances of the alert threshold value. As in the graphs presented above, in the evolution of air quality in terms of arsenic pollution, Timisoara stands out, with 0.93 ± 0.05 [ng/m^3] for the aggregate index. In this city, unlike the other cities, significant concentrations were recorded every year, with a maximum value of the average annual concentration of 1.2 [ng/m^3] in 2016, all values being close to the alert threshold value, but without exceeding this level. We cannot say the same about Cluj-Napoca, where the largest error for arsenic can be observed, with an aggregate index of 0.54 ± 0.38 [ng/m^3], a fact caused by the variation in the maximum concentrations obtained on certain days during 2017 (5.76 ng/m^3) and 2020 (4.91 ng/m^3), exceeding the alert threshold value for this pollutant, as can be seen in Figure 7a. No significant concentrations were recorded for the rest of the analyzed period.

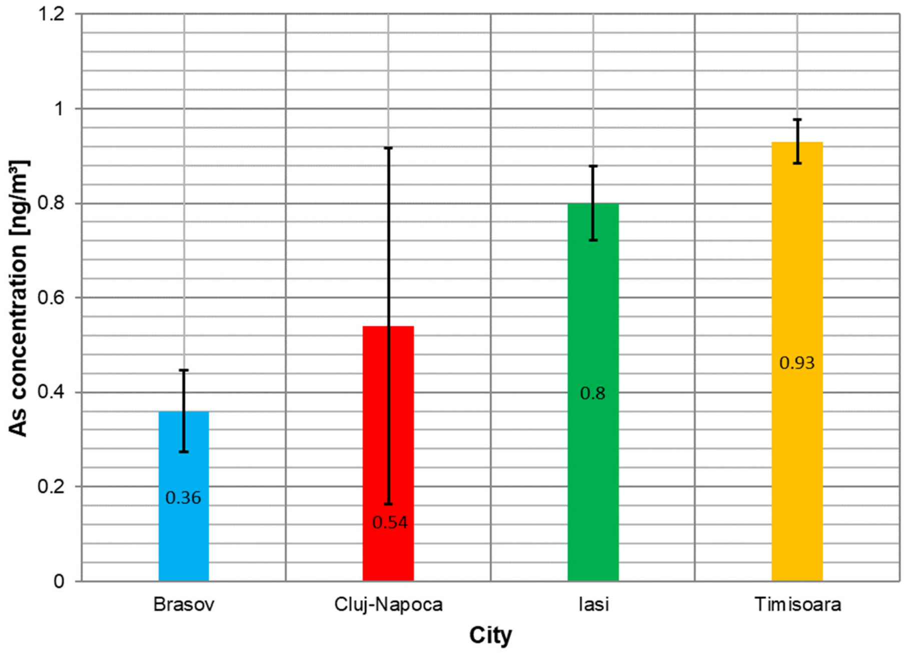


(a)



(b)

Figure 8. *Cont.*



(**c**)

Figure 8. (**a**) Air quality aggregate index for $PM_{2.5}$ with standard error (2011–2021); (**b**) air quality aggregate index for CO with standard error (2011–2021); (**c**) air quality aggregate index for As with standard error (2011–2021).

3. Air Quality Assessment Methodology

In order to evaluate the impact generated by the 3 pollutants ($PM_{2.5}$, CO, and As) on air quality and human health, the air pollution index (PI) and the health risk index for the period 2011–2021 (11 years) were calculated.

3.1. Air Pollution Index

The calculation formula for the air pollution index (PI) is represented by the ratio between the concentration of a pollutant in the air and the maximum value allowed by legislation for that pollutant. Further, Equation (1) represents the calculation formula for PI [36]:

$$\mathrm{PI} = \frac{\mathrm{Ci}}{\mathrm{MACi}} \tag{1}$$

Equation (1). Air pollution index formula.
where:

- Ci: the determined concentration of a pollutant in the air, in this case, the annual average concentration (mg/Nm^3);
- MACi: the maximum allowed concentration of a pollutant (mg/Nm^3).

The next step is to calculate the pollution load index (PLI) in order to estimate the air quality in a certain area. The calculation method is presented in Equation (2) [36]:

$$\mathrm{PLI} = (\mathrm{PI1} \times \mathrm{PI2} \times \mathrm{PI3} \times \ldots \times \mathrm{PIn})^{1/n} \tag{2}$$

Equation (2). Air pollution load index formula.
where:

- PI1, PI2, PI3: air pollution indices for each pollutant, with the calculations conducted according to the formula presented above;
- N: number of pollutants analyzed.

Furthermore, the framing of the values in one of the possible scenarios can be carried out according to Table 2 [36].

Table 2. Classification of the pollution load index.

PLI Scale	Classification
PLI < 1	Unpolluted air
$1 \leq$ PLI < 2	Unpolluted to moderately polluted
$2 \leq$ PLI < 3	Moderately polluted
$3 \leq$ PLI < 4	Moderately to highly polluted
$4 \leq$ PLI < 5	Highly polluted
PLI $\geq$ 5	Very highly polluted (excessive)

3.2. Human Health Risk Index

The health risk generated by the effects of pollutants is realized by assessing the carcinogenic and non-carcinogenic risks, following several stages presented below. The applicability of the formulas and the scenario used can be found in Section 4. There was another formula used for this index, which also included the PEF parameter (pollution emission rate) but according to the EPA the use of that equation is no longer recommended. Thus, the new formula does not consider this factor anymore. The first step is to determine the daily inhaled dose according to the updated Equation (3) [37,38]:

$$\mathrm{ADD_{inh}} = \frac{\mathrm{C_i} \times \mathrm{InhR} \times \mathrm{ET} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{3}$$

Equation (3). Average daily inhaled dose formula.
where:

- $\mathrm{ADD_{inh}}\left(\frac{\mathrm{mg}}{\mathrm{kg}} \cdot \frac{1}{\mathrm{day}}\right)$: average daily inhaled dose;
- $\mathrm{C_i}\left(\frac{\mathrm{mg}}{\mathrm{m}^3}\right)$: air concentration of a pollutant—in our case, we used the value of the aggregate index for each individual pollutant, after converting all units of measure into mg/m^3, in order to have the same measurement unit, according to the formula;
- $\mathrm{InhR}\left(\frac{\mathrm{m}^3}{\mathrm{hour}}\right)$: volume of inhaled air;
- $\mathrm{ET}\left(\frac{\mathrm{hours}}{\mathrm{day}}\right)$: exposure time;
- $\mathrm{EF}\left(\frac{\mathrm{days}}{\mathrm{year}}\right)$: frequency of exposure;
- ED (years): duration of exposure;
- BW (kg): body weight;
- AT (days): ED $\times$ 365 (days)—average exposure time.

Furthermore, in order to characterize the non-carcinogenic risks, the hazard coefficient (HQ) was determined, the calculation formula of which is represented by Equation (4) [39]:

$$\mathrm{HQ} = \frac{\mathrm{ADDinh}}{\mathrm{RfD}} \tag{4}$$

Equation (4). Hazard coefficient formula.
where:

- HQ: hazard coefficient;
- $\mathrm{ADD_{inh}}$: daily dose that can be inhaled;
- RfD: daily reference dose (as shown in Table 3)—the values were taken from the IRIS and RAIS databases [40,41].

Table 3. Calculation of the average daily dose (ADD_{inh}) and the hazard coefficient (HQ).

City	Indicator	AT_{Adults} (Days)	$AT_{Children}$ (Days)	$ADD_{Inh\ Adults}$ (mg/kg/Day)	$ADD_{Inh\ Children}$ (mg/kg/Day)	RfD	HQ_{Adults}	$HQ_{Children}$
Brasov	$PM_{2.5}$	8760	2190	0.00112	0.00415	0.035 (RAIS)	0.03200	0.11857
	CO			0.01720	0.06351	9 (RAIS)	0.00191	0.00705
	As			0.22942×10^{-7}	0.84690×10^{-7}	0.0003 (IRIS)	0.00007	0.00028
Cluj-Napoca	$PM_{2.5}$	8760	2190	0.00114	0.00421	0.035 (RAIS)	0.03257	0.12028
	CO			0.01656	0.06116	9 (RAIS)	0.00184	0.00679
	As			0.34414×10^{-7}	1.27035×10^{-7}	0.0003 (IRIS)	0.00011	0.00042
Iasi	$PM_{2.5}$	8760	2190	0.00145	0.00536	0.035 (RAIS)	0.04142	0.15314
	CO			0.01784	0.06587	9 (RAIS)	0.00198	0.00731
	As			0.50984×10^{-7}	1.88200×10^{-7}	0.0003 (IRIS)	0.00016	0.00062
Timisoara	$PM_{2.5}$	8760	2190	0.00093	0.00346	0.035 (RAIS)	0.02657	0.09885
	CO			0.01784	0.06587	9 (RAIS)	0.00198	0.00731
	As			0.59269×10^{-7}	2.18783×10^{-7}	0.0003 (IRIS)	0.00019	0.00072

Equation (5) presents the calculation formula for the hazard index (HI), with the help of which the impact generated by the non-carcinogenic effects can be estimated [39]:

$$HI = \sum_{n=1}^{i} HQn \quad (5)$$

Equation (5). Hazard index formula.

Thus, the hazard index is characterized as follows:

- If HI < 1, there are no health risks;
- If HI > 1, there are possible health risks, depending on the value of the index—the higher it is, the higher the risk [39].

4. Results and Discussion

4.1. Air Pollution Index

The results of the air pollution index calculated for the last 11 years, for four cities in Romania, are presented in Table 4.

According to Table 2 and the results on the air pollution load index in Table 3, the air quality in the four cities falls into the "unpolluted" scenario. This is possible because all values for this index are less than 1, even if the alert threshold was exceeded in some situations. At the same time, it is important to mention that in this paper, only three pollutants ($PM_{2.5}$, As, CO) were considered, but in one city, they are much more diversified. Therefore, the highest value calculated for the air pollution load index was found in Iasi, while Brasov, has the lowest value. The value of the pollution load index depends a lot on the specifics of the area under analysis. This is also supported by a 2022 study on the risk assessment of potentially toxic elements in the Indonesia region. Thus, in this case, values higher than 1 were obtained for PLI, this value increasing more and more with the proximity to industrial areas, being also influenced by winds, which transport the dust to populated areas [42].

Table 4. Calculation of the air pollution load index in Brasov, Cluj-Napoca, Iasi, and Timisoara.

City	Indicator	MAC	C_i	PI	PLI
Brasov	$PM_{2.5}$ CO As	20 10 6	17.65 0.27 0.36	0.88 0.03 0.06	0.117 (<1)
Cluj-Napoca	$PM_{2.5}$ CO As	20 10 6	17.90 0.26 0.54	0.90 0.03 0.09	0.134 (<1)
Iasi	$PM_{2.5}$ CO As	20 10 6	22.81 0.28 0.80	1.14 0.03 0.13	0.164 (<1)
Timisoara	$PM_{2.5}$ CO As	20 10 6	14.74 0.28 0.93	0.74 0.03 0.16	0.153 (<1)

The pollution sources in the case of $PM_{2.5}$, can be mentioned as the incomplete burning of fuels by car engines, combustion processes (such as waste burning or residential heating), some industrial processes, and also road wear or construction sites. The incomplete combustion of fossil fuels, as well as traffic or some industrial processes, is also a source of carbon monoxide and arsenic emissions [34].

4.2. Human Health Risk Index

In this paper, the determination of the health risk index was carried out following the method presented by USEPA [38,43]. The specific parameters (InhR and ED) used to quantify the health hazards are in accordance with the literature. According to the previous studies, the EF was considered 350 days/year and ET 8 h/day [44–46]. For this purpose, two scenarios were considered: the first corresponded to adults weighing 65 kg (BW_{Adults}), and the second to children weighing 15 kg ($BW_{Children}$). The results are presented in Tables 3 and 5.

Table 5. Specific values for the calculation of the health risk index.

City	Indicator	C_i (mg/m^3)	$InhR_{Adults}$ (m^3/h)	$InhR_{Children}$ (m^3/h)	EF (Days/Year)	ED_{Adults} (Years)	$ED_{Children}$ (Years)	ET (h/Day)
Brasov	$PM_{2.5}$ CO As	0.01765 0.27 3.6×10^{-7}	0.54	0.46	350	24	6	8
Cluj-Napoca	$PM_{2.5}$ CO As	0.01790 0.26 5.4×10^{-7}	0.54	0.46	350	24	6	8
Iasi	$PM_{2.5}$ CO As	0.02281 0.28 8×10^{-7}	0.54	0.46	350	24	6	8
Timisoara	$PM_{2.5}$ CO As	0.01474 0.28 9.3×10^{-7}	0.54	0.46	350	24	6	8

After calculating the indices presented in the previous tables, the determination of the hazard index followed, the results of which can be found in Table 6.

Table 6. Health risk index calculated for each city.

City	HI_{Adults}	$HI_{Children}$
Brasov	0.03398	0.12590
Cluj-Napoca	0.03452	0.12749
Iasi	0.04356	0.16107
Timisoara	0.02874	0.10688

The graphical representation for the health risk index can be found in Figure 9 for all four cities, both for adults and children. We noticed that all values were below 1, which means that there are no health risks [47]. One difference is that in the case of children, the index is higher than that obtained for adults, which means that they are more exposed to the negative effects generated by air pollution [47]. This is also supported by the results of other studies with the same theme, with varying results, but always obtaining higher values of the index for children compared to adults, indicating children's vulnerability to the negative effects of pollution [48,49]. Previous studies reported similar results in the case of HI, also highlighting the cumulative effect of heavy metals (arsenic, lead) and the low degree of tolerance to pollutants of children, as well as the variation of the index depending on the activities carried out in the area or the difference between the urban and rural environment [50]. Evaluations carried out in 2021 show higher values of the index in the urban environment compared to the rural area [51]. According to the graph, the index had the highest value in Iasi, both for adults and children, of 0.04356 and 0.16107, respectively. For Timisoara, the lowest values were determined, with 0.02874 for adults and 0.10688 for children. The results of this work are also supported by other international publications. Even though the air quality indicators measurements are under the maximum allowed concentrations, according to the environmental standards, there are no completely safe limits, especially for children, who are much more vulnerable than adults [52–54].

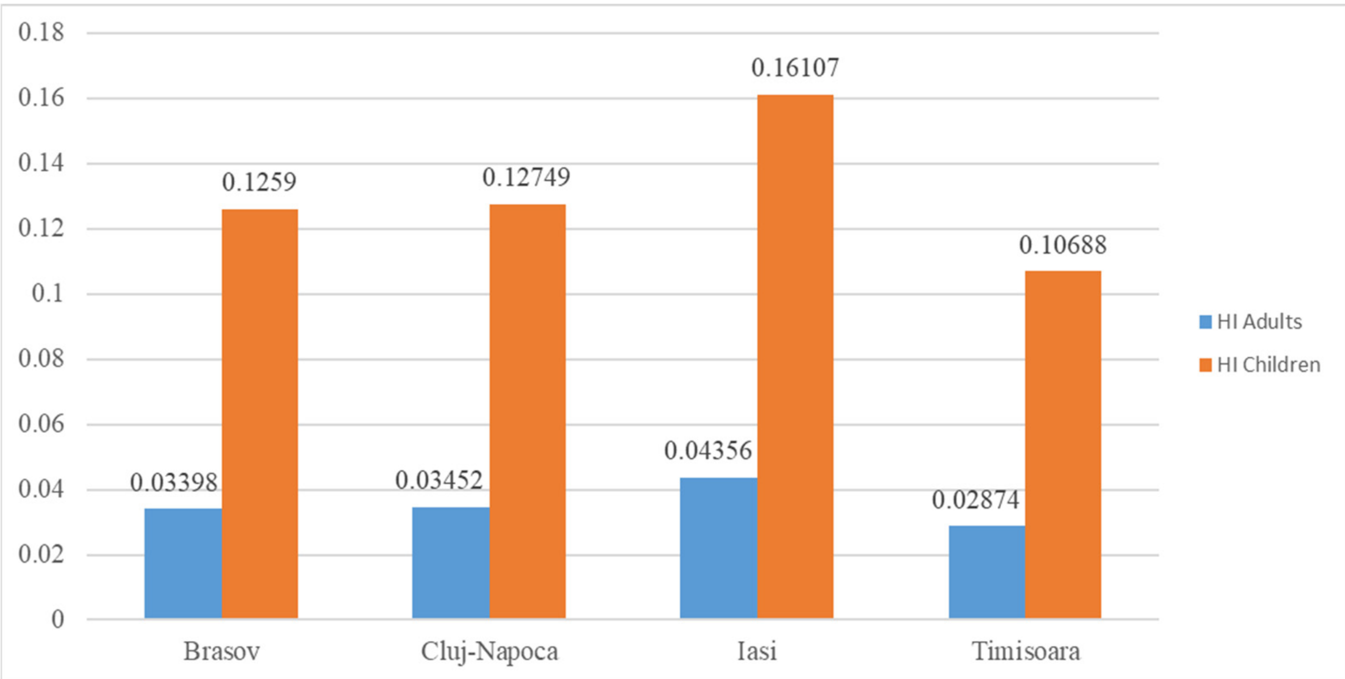


Figure 9. Health risk index for adults and children for the main four cities of Romania.

As future work, from an immediate perspective, the results of this study will be the basis of a new calculation and the correlation of health indices considering various meteorological parameters as well. For example, the planetary boundary layer (PBL) majorly influences the cumulation and dispersion of pollutants at ground level and the interpretation of the PBL behavior is obtained through its characterization as a mathematical object of multifractal type [55]. Therefore, the theoretical parameters of the PBL characterization validated by the experimental data will contribute to the realization of the correlations mentioned above, using the machine learning concept. Also, recent studies regarding the air quality in Romania have shown that due to the circulation of air masses that can transport, for example, dust over long distances, a pattern of optical characterization of aerosols has been created, over time, through modern monitoring remote sensing techniques such as Aerosol, Clouds and Trace gases Research InfraStructure—Romania (ACTRIS—RO) [56,57]. Accordingly, considering the specific aerosol type at different scales (local, regional), the health indices presented in this paper will thus better emphasize the specific situations and vulnerability as similar studies applied in the case of water health hazards assessments [58].

5. Conclusions

The air quality index and health hazards were determined based on data provided by the EPA reports for the last 11 years (period of 2011–2021), for three pollutants ($PM_{2.5}$, CO, and As). For each city, three monitoring stations for daily measurements were considered. Statistical analyses were performed using the minimum and maximum concentrations measured within each year for each city, while the air quality index and health hazards considered the annual average measured concentrations. The results prove that Iasi is the most polluted city in the case of fine particulate matter, with the highest measured value in 2017, 44% higher compared to 2016. Carbon monoxide concentrations remained below the alert threshold, registering variations with a maximum increase in 2017, increasing by 75% compared to 2016. The arsenic concentrations were recorded close to the alert threshold value in Timisoara, for each year. In this case, the highest increase was observed in 2020, increasing by 90% compared to 2019. The air quality situation in Brasov indicates a decreasing trend of the concentrations of the three pollutants starting in 2018. Regarding the city of Cluj-Napoca, for $PM_{2.5}$ and carbon monoxide, a decreasing trend of pollutant concentrations was recorded. A more unusual situation was reported for arsenic because no concentrations, or insignificant ones, were measured until 2020, the COVID-19 pandemic year, when a value of 2.59 [ng/m^3] was recorded, but in 2021, it decreased to 0 again. Regarding the variation in the maximum concentrations recorded on certain days of each year, compared to the value of the alert threshold, it was found that the threshold was exceeded for $PM_{2.5}$ in the case of all four cities.

Since the values of the air pollution index and health hazard index are within the regular range, the immediate perspective of this study includes a more complex statistical analysis. This also includes data from low-cost sensors for intercomparison and to create a degree of confidence that could be considered, for instance, in smart city applications. The results of the aggregate index in the case of the four cities considering the three pollutants for the period 2011–2021 show that the most polluted city is Iasi, followed by Cluj-Napoca, Brasov, and Timisoara. The air pollution index and the health hazard index showed that, even though their values are within normal limits, there is still a risk for human health since the values are almost triple for children compared to adults. This work offers certain perspectives, being a starting point for a much more detailed future analysis of the data and the correlation of possible increases or exceedances of the normal limits depending on the season, or even other parameters, because it is known that one of the factors that influence pollutant concentration variations is the clime of each season.

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