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Temporal Trend of PM₁₀ and Associated Human Health Risk over the Past Decade in Cluj-Napoca City, Romania

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Abstract: The human health risk associated with PM₁₀ exposure was assessed for the residents of Cluj-Napoca city, Romania, for a best case-scenario based on the monthly average PM₁₀ and for a worst-case scenario based on the monthly 90th percentile of PM₁₀ concentration. As no toxicity value for PM₁₀ was available, for the calculation of the hazard quotient, the toxicity value was considered to be equal to the annual limit value (40 µm/m³) set in the European Union (EU), and to air quality guidelines (20 µm/m³) set by the World Health Organization (WHO). The daily PM₁₀ concentrations for the period 2009–2019, at the four monitoring stations existing in Cluj-Napoca, were obtained from the National Air Quality Monitoring Network. The annual PM₁₀ values ranged between 20.3 and 29.5 µg/m³, and were below the annual limit value (40 µg/m³) set by European and national legislation, but above the annual air quality guideline (20 µg/m³) set by WHO. Generally, the monthly PM₁₀ concentrations were higher from October to March than in the rest of the year. The monthly air quality index (AQI) showed the good to moderate quality of the air during the whole decade; however, there were days when the air quality was unhealthy for sensitive population groups. The air quality was more or less constant during the warm months, and improved significantly for the cold months from 2009 to 2019. In the best-case scenario, calculated using the EU annual limit value for PM₁₀, the potential non-carcinogenic chronic health risk was present only in 2009 and 2010, but in the worst-case scenario, in each year there were periods, especially in the cold months, in which health risk was present. When considering the WHO air quality guidelines in the calculation of the health risk, the potential non-carcinogenic chronic health risk was present between October and March in each year in the best-case scenario, and in most of the months in the worst-case scenario.

Keywords: air quality; PM₁₀; human health risk; hazard quotient

1. Introduction

Despite the important measures taken to reduce pollutant emissions in the last few decades, air quality remains a major issue of concern all over the world [1,2]. Exposure to air pollution may

cause acute and chronic health effects, determining increased mortality and morbidity in the exposed population [3]. In Europe, air pollution is considered the most important environmental risk to human health, and one of the biggest environmental concerns [4]. In this sense, the EU air quality policy was revised based on the research data obtained through several international research projects [5]. At European level, the major sources of outdoor air pollution, with particulate matter with a diameter below 10 μm (PM_{10}), are road transport and space heating, followed by shipping, energy production and distribution, and agriculture and industry [5]. Waste burning, a common practice especially in developing countries, was found to have a high impact on the PM_{10} concentration [6]. In Romania, the main pollution source of PM_{10} is space heating (62%), followed by agriculture (11.5%), industrial processes and product use (10.3%), energy use in industry (4.5%), road transport (3.8%), energy production (3.9%) and waste (0.8%) [7]. Previous studies made for Romania concerning the PM_1 non-refractory fraction, done through receptor-oriented models, reveal season-dependent sources. The receptor-oriented models used were based on the assessment of sources' contributions to pollutants concentration, measured at a specific point [8]. In winter, the dominant sources are related to road transport, wood combustion associated with residential heating and coal combustion, while during the summer the main sources are traffic, biomass-burning associated with agricultural activities and long-range transport. Agricultural and industry tracers such as ammonia, and sulphate, are present all over the year [9]. Due to high population density, high energy demand and large number of vehicles, cities are facing more intense air pollution than rural areas [4,10,11].

Particulate matter (PM) is a complex mixture of solid particles and liquid droplets suspended in the atmosphere, that varies in concentration, size, shape, chemical composition, surface area, acidity, solubility, reactivity and origin. Exposure to high PM levels is a potential health risk for humans, as inhaled particulates may reach different depths of the lungs, according to their size, causing severe adverse health effects. Several studies revealed the association of high PM levels with the increase of respiratory and cardiovascular pathology incidences [12–16]. Generally, natural sources emit particles with diameters above 2.5 μm , while anthropogenic sources emit fine and ultrafine particles. As PM_{10} contains the fine and ultrafine fraction, the health risk calculated based on PM_{10} concentration may be slightly overestimated. Other studies revealed that a decrease in $\text{PM}_{2.5}$ of 10% may determine an increase in life expectancy of more than a half year [17–21]. Vaduganathan et al. reported that PM_{10} levels below the daily limit (50 $\mu\text{g}/\text{m}^3$) set by the EU Directive 2008/50/EC are associated with excess risk for acute cardiovascular events [22,23]. To reduce the health impacts of air pollution, the World Health Organization (WHO) developed Air Quality Guidelines (AQG) for several pollutants, among them also for PM_{10} . These AQG are independent from any policy restrictions or local consideration, and are neither standards nor legally binding criteria, but they offer guidance for governmental authorities in developing health-based national air quality management strategies. Moreover, outdoor air pollution and particulate matter were included on the list of carcinogens by the International Agency for Research on Cancer [24]. Although air pollution with PM_{10} affects the whole population, it was found that certain groups of workers (professional drivers, urban traffic police, street vendors, etc.) or age groups such as children, older adults and those with pre-existing health problems are more vulnerable to developing specific pathologies [4,5,24,25]. Furthermore, population groups with low socio-economic status tend to be more exposed to PM_{10} , as they most probably live in highly polluted areas, use low quality fuels for heating and have deficient waste management practices [4]. Besides the health effects on humans and the environmental impact, considerable economic impacts are attributed to air pollution, as it reduces life expectancy, increases medical costs and reduces productivity through working days lost [4].

To estimate the health risk resulting from exposure to a specific pollutant, the United States Environmental Protection Agency (US EPA) risk assessment methodology is frequently used [26]. Another tool that allows assessing the relationship between pollutant concentration and health impacts is the Air Quality Health Impact Assessment software [11]. The importance of conducting human health risk assessments is confirmed by the fact that in 2017, concentrations of PM_{10} above the EU daily

limit value ($50 \mu\text{m}/\text{m}^3$) were measured by 22% of the monitoring stations in 17 out of the 28 EU member states, while levels above *WHO AQG* ($20 \mu\text{m}/\text{m}^3$) were measured by 51% of the monitoring stations in the majority of the countries [4]. Furthermore, around one fifth of the European urban population was exposed to PM_{10} levels above the EU daily limit value, and half of it to PM_{10} above *AQG* [4]. Air quality in Romania represents a European and national concern. In this regard, the EU opened the infringement procedure against Romania in 2018 for not taking measures to reduce atmospheric pollution in the major cities. Bucharest, the capital city of Romania, received penalties for PM_{10} levels exceedance, while Cluj-Napoca city received the same for NO_2 . In 2015, the European Environmental Agency estimated 27,280 premature deaths in Romania associated with PM, ozone and nitrogen dioxide, out of which 25,400 were attributed to PM concentrations [27]. The urban population of Romania exposed to PM_{10} values above the EU daily limit values decreased from 53.1% in 2013 to 21.4% in 2017 [28]. This decrease could be the consequence of the initiation of air monitoring and pollution abatement measures requested by Law 104/2011, which transpose into the national legislation the Directive 2008/50/EC ambient air quality and cleaner air for Europe [29]. Numerous studies reported the level of PM in cities all over the world [1,30–33]. Human health risk assessment studies regarding PM_{10} and toxic elements in PM_{10} were also carried out in several cities [34–39]. These studies revealed that although PM concentrations do not exceed air quality standards, the health impacts associated to air pollution exposure are still important [40]. Therefore, the assessment of the human health risks caused by exposure to airborne PM is an important step in order to control and mitigate urban air pollution. As the chemical composition of PM depends on a high number of factors, such as geography, season, climate and combustion sources, and differs from region to region, the health risk assessment in different cities could give a better insight into the population exposure and health risks [18]. In Romania, Leitte et al. presented the adverse effects of air pollution on the respiratory tract for the inhabitants of Drobeta Turnu Severin [39], while Dunea et al. showed the impact of $\text{PM}_{2.5}$ and associated metals on the health of children in the industrial area of Targoviste [40]. However, to the best of our knowledge, no health risk assessment associated with PM_{10} has been conducted in Romania.

This research aimed to assess the health risks associated with PM_{10} for the residents living in one of the largest urban agglomerations in Romania. The air quality index (*AQI*) was applied in the urban agglomeration of Cluj-Napoca, in order to analyze the temporal variation of the pollution level of PM_{10} from 2009 to 2019, and to identify the seasons when air quality can become an issue for the local population. The health risk assessment, according to EU and *WHO* recommendations, was conducted for the first time in a Romanian city. The health risk assessment will offer information to local governments regarding the efficiency of air quality management. The results of this study can support decision-makers in implementing and developing better strategies and regulations to improve air quality and to mitigate the effects on human health.

2. Materials and Methods

2.1. Study Area and Population

Cluj-Napoca city represents the fourth most populated city in Romania, with 326,687 inhabitants (47.1% men and 52.9% women) in the city and 420,000 in the metropolitan area. According to the 2011 Population and Housing Census reported by the National Institute of Statistics and the Statistical Division of the Cluj County, the population is 4.3% children (age under 5), 12.9% pupils and students (age 5–19), 70.3% adults (age 20–64) and 12.5% elders (age > 65) [41].

Located in the *Somes Mic* river valley, surrounded by forests and grasslands, Cluj-Napoca has a continental climate characterized by warm, dry summers and cold winters. The city is an important trade and tourist center and a hub of the European network roads, connecting the country with Western Europe. It has a large industrial park with modern facilities, and the second main airport in Romania, after Bucharest, is located 9 km east of the center of the city. The local topography, mainly the *Somes Mic* River that crosses the city, influences the dominant NW (~15%), NE (~12%), W (~10%) and SW

(~10%) wind direction [42]. Further, the dominant wind direction can favor the accumulation processes or the upwind emissions.

The Integrated Air Quality Plan for Cluj-Napoca city for 2019–2023 identified five primary PM_{10} pollution sources that account for 89% of the total PM_{10} emission in the city, as follows: household heating (32%), road reconstructions (18%), traffic (16%), industrial activities (12%) and construction/demolition sites (11%) [43].

2.2. Air Quality Data

Cluj-Napoca has 4 stations belonging to the National Air Quality Monitoring Network that monitors the traffic (CJ-1), urban (CJ-2), suburban (CJ-3) and industrial (CJ-4) influences on the air pollution (Figure 1).

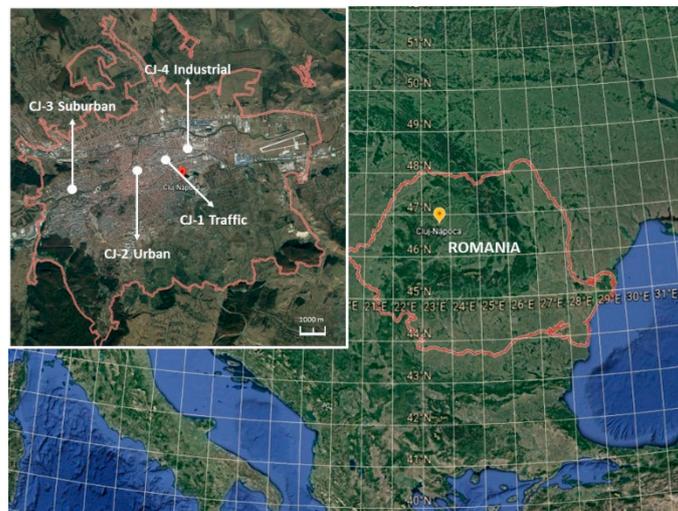


Figure 1. Location of the monitoring stations in Cluj-Napoca city.

At the CJ-1, CJ-2 and CJ-3 stations, the PM_{10} concentration is measured gravimetrically using Skypost Tecora (Cogliate, Italy) low volume samplers, while at the CJ-4 station it is done by optic measurements using an LSPM10 (Unitec, Veggiano, Italy) analyzer. In this study, the assessment of PM_{10} air pollution in Cluj-Napoca was carried out using the daily PM_{10} concentrations provided by the operational monitoring stations. The daily PM_{10} was further aggregated to monthly and annual concentrations for the period 2009–2019. Data are publicly available on the National Air Quality Monitoring Network website [44].

2.3. Air Quality Index

The air quality can be comprehensively presented to citizens using the *AQI*, calculated according to Equation (1). While the *AQI* is normally used to report the daily air quality at each monitoring station, in this study it was used to assess the monthly variability and to underline the multi-annual trends using all four operational stations in Cluj-Napoca city, considering that a single monitoring station is not representative for a city and did not reflect spatial variations of PM_{10} . Therefore, the daily *AQI* was calculated based on the average PM_{10} concentration obtained for the operational stations and the EU daily limit value ($50 \mu\text{g}/\text{m}^3$) [45,46]. The daily *AQI* was further aggregated to obtain the monthly *AQI*.

$$AQI = \frac{PM_{10} \text{ concentration}}{\text{threshold concentration}} \times 100 \quad (1)$$

According to *AQI*, the air quality is classified as good (0–50), moderate (51–100), unhealthy for sensitive groups (101–150), unhealthy (151–200), very unhealthy (201–300) and hazardous (301–500).

High AQI values indicate a high level of air pollution with PM₁₀. The AQI values below 100 are considered satisfactory, while values above 100 are considered to pose health risks [45].

2.4. Human Health Risk Assessment

To estimate the human health risk related to PM₁₀ exposure by inhalation, the hazard quotient (HQ) was calculated according to Equation (2) [26]:

$$HQ = EC/TV \tag{2}$$

where EC is the exposure concentration of PM₁₀ (µg/m³) calculated according to Equation (3), and TV is the toxicity value [26]. As no toxicity value for PM₁₀ was found in the literature, the calculations were made assuming that the TV was equal to the EU annual limit value of 40 µm/m³ (HQ₁), and to the WHO AQG of 20 µm/m³ (HQ₂).

$$EC = (CA \times ET \times EF \times ED) / AT \tag{3}$$

where CA is the monthly average PM₁₀ concentration (µg/m³) for the best-case scenario and the monthly 90th quartile for the worst-case scenario, ET is the exposure time (hours/day), EF is the exposure frequency (days/year), ED is the exposure duration (year) and AT is the averaging time calculated as ED × 365 days/year × 24 h/day. For the exposure of residents, ET was considered 24 h/day, EF was 350 days/year and ED was 30 years for adults [26].

Non-carcinogenic risk includes all the adverse health effects in the organism, excluding cancer caused by exposure factors. The used safety benchmark level for HQ is 1. Thus, the exposure to PM₁₀ could induce non-carcinogenic chronic effects if HQ > 1, while no non-carcinogenic health risk is expected if HQ < 1 [47].

3. Results and Discussion

3.1. Air Quality

The basic statistics for PM₁₀ for the period 2009–2019, calculated based on the daily values of the operational monitoring stations, are presented in Table 1. The annual average, calculated based on daily values, ranged between 20.3 and 29.5 µg/m³, this being below the annual limit value set by the EU legislation (40 µg/m³), but above the AQG set by the WHO (20 µg/m³) [4,29].

Table 1. Basic statistics of the annual PM₁₀ concentrations (µg/m³) in Cluj-Napoca for 2009–2019.

PM ₁₀	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
Minimum	2.0	6.35	2.7	5.8	6.7	7.0	7.5	2.7	3.2	7.9	3.6
Maximum	86.8	111.3	59.7	54.8	61.6	65.7	61.6	76.6	56.5	59.5	62.2
Average	27.4	29.5	23.2	24.0	24.4	24.9	25.7	22.0	22.2	24.4	20.3
Standard deviation	15.9	14.5	11.1	9.8	10.7	11.2	10.1	10.6	10.1	9.4	10.7
10th percentile	9.8	14.2	11.4	12.2	12.8	11.8	14.4	9.7	10.4	13.8	8.7
50th percentile	24.6	26.8	19.9	22.5	21.3	22.6	23.7	20.4	21.04	22.7	18.0
90th percentile	51.6	47.6	40.9	38.1	39.2	40.1	39.9	36.5	38.1	37.6	35.3
Skewness	1.1	1.5	0.8	0.5	0.9	0.6	0.7	1.0	0.7	0.8	1.1
Kurtosis	0.6	4.0	−0.2	−0.6	0.4	−0.2	0.1	2.3	0.17	0.5	1.4
Days with PM ₁₀ > 50 µg/m ³	29	26	4	1	6	6	5	3	3	5	9

The highest annual maximum was measured in 2010 (111 µg/m³), while the lowest annual maximum in 2012 (54.8 µg/m³). During the monitoring period, the annual average concentration was comparable, ranging between 20.3 and 29.5 µg/m³, but the annual maximum almost halved from 111 µg/m³ in 2010 to 62.6 µg/m³ in 2019. The difference between the average and median annual PM₁₀ value decreased from 2009 to 2019, indicating a reduction of the days with high PM₁₀ values in favor

of days with low PM₁₀ values. The positive skewness of the data also confirms this decrease. Highly skewed data were obtained for 2009, 2010 and 2019, the years that also have the highest numbers of days with PM₁₀ exceeding the daily limit value. The daily maximum PM₁₀ threshold value (50 µg/m³) was not exceeded more than 35 times in the studied years. The highest number (29) of breaches occurred in 2009, of which 20 breaches occurred during October–December. With few exceptions, in each year, the majority of breaches occurred between October and March.

The observed decreasing trend could be explained by the existence of some pollution reduction measures related to traffic restriction in the city center, and to the decrease of emissions from domestic heating following the increase in ambient temperatures and reduction of the number of frost days in Cluj-Napoca during the winters. Temperature fluctuation is the main factor that favors the increase in PM₁₀ concentrations, while the wind speed influences the dispersion of pollutants (Figure 2). Both temperature ($r = -0.68$) and wind speed ($r = -0.44$) are negatively correlated with the PM₁₀ concentration. The highest monthly average wind speed was recorded in spring (4.51 m/s), while the lowest monthly average wind speed was recorded in December (3.13 m/s). Generally, the highest PM₁₀ concentrations were recorded when the average monthly temperature was below 0 °C and the intensity of wind was low.

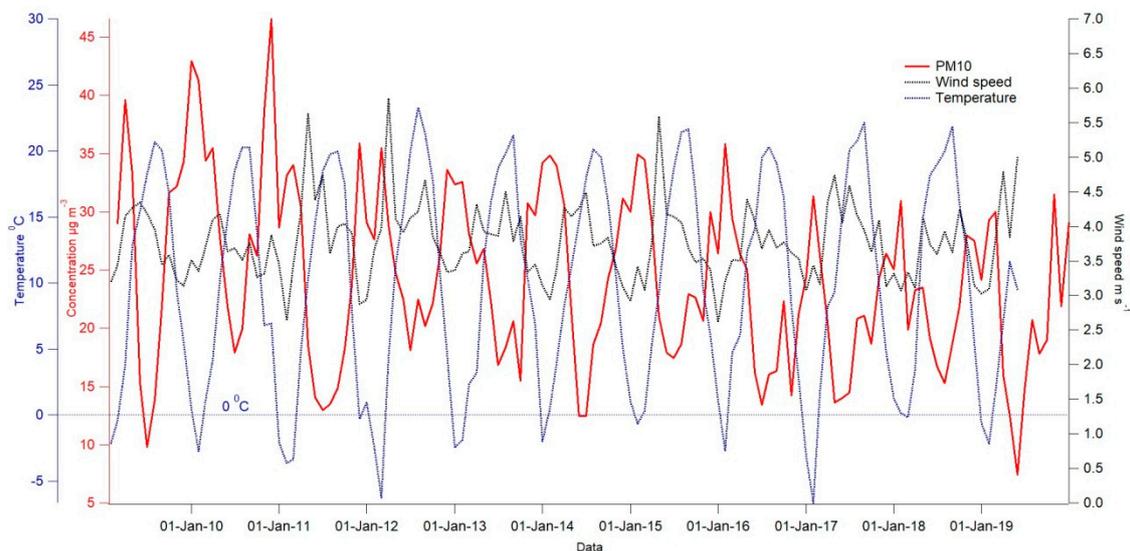


Figure 2. Temporal evolution of PM₁₀, temperature and wind speed in Cluj-Napoca between 2009 and 2019.

The heavy traffic from the city center was redirected to the city outskirts, starting in 2011, when the ring road became functional. The decrease in traffic was noticed, especially at the CJ-1 traffic station, where a constant reduction in PM₁₀ concentrations was recorded. In CJ-1, the PM₁₀ levels decreased from 38.6 µg/m³ in 2009 to 32.1 µg/m³ in 2019. A constant decrease was also remarked at the CJ-4 industrial station, where the annual average concentrations reduced from 28.4 µg/m³ in 2009 to 15.7 µg/m³ in 2019; a possible explanation could be the decrease in economic activities after the 2008 recession.

The monthly PM₁₀ (Figure 3a) was the highest in December 2009 (44.4 µg/m³) and the lowest in May 2019 (7.4 µg/m³). Generally, the monthly PM₁₀ concentrations were higher in the cold seasons than in the warm seasons. A possible explanation for high PM₁₀ levels in winter could be the accumulation of PM in the low atmosphere due to the reduction of mixing layer thickness, determined by meteorological conditions. Household heating and the use of solid road deicing products during winter could also have increased the PM₁₀ emissions [1]. Both the maximum and the 90th percentile of monthly PM₁₀ (Figure 3b) showed decreasing trends, confirming the lowering of the value and frequency of high concentrations. In the case of the 90th percentile, the higher values were measured between October and March, and a decreasing trend in monthly PM₁₀ from 2009 to 2019 was observed.

To assess the spatial variability of PM_{10} concentration in Cluj-Napoca, the average PM_{10} for each monitoring station was calculated for the periods in which all four stations functioned. The average PM_{10} concentrations were comparable for the CJ-2 (urban) and CJ-3 (suburban) stations, with values of $26.8 \mu\text{g}/\text{m}^3$ and $26.7 \mu\text{g}/\text{m}^3$, respectively. These values were almost two times higher than the average PM_{10} concentrations found for the CJ-1 (traffic, $13.6 \mu\text{g}/\text{m}^3$) and CJ-4 (industrial, $15.1 \mu\text{g}/\text{m}^3$) stations. The low PM_{10} values were measured by the industrial and traffic background stations, which confirms that industry and traffic are responsible for a lower share of PM_{10} pollution than the other urban pollution sources, such as heating and constructions/demolitions.

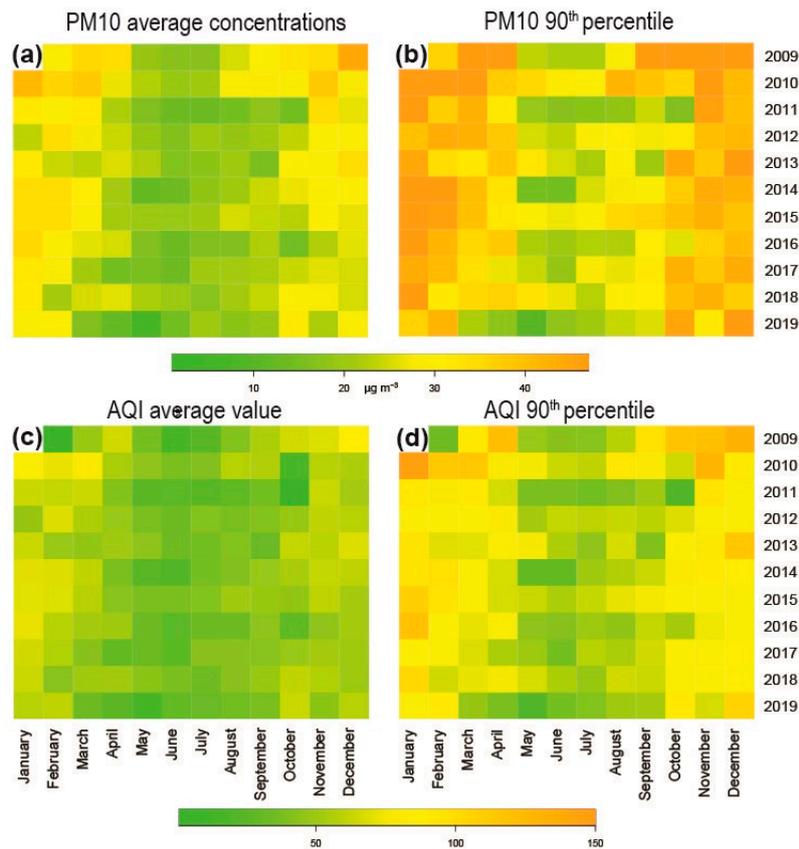


Figure 3. Temporal variation of (a) average of PM_{10} concentration, (b) 90th percentile of PM_{10} concentration ($\mu\text{g}/\text{m}^3$), (c) average of air quality index (AQI) and (d) 90th percentile of air quality index (AQI) in Cluj-Napoca city for 2009–2019.

The obtained annual PM_{10} values were comparable to those reported by Kassomenos et al. (2014) in London ($26\text{--}44 \mu\text{g}/\text{m}^3$), Athens ($23\text{--}51 \mu\text{g}/\text{m}^3$) and Madrid ($29\text{--}39 \mu\text{g}/\text{m}^3$) [31]. Slightly lower PM_{10} multiannual (2000–2006) average values were obtained by Pascal et al. (2014) in nine French cities [35]. The PM_{10} values measured in Cluj-Napoca were lower than those from Bucharest [48,49].

The average monthly AQI (Figure 3c) ranged between 15 (May 2019) and 87 (December 2009), indicating good to moderate air quality. However, the 90th percentile (Figure 3d) ranged between 22 (May 2019) and 147 (January 2010), indicating that there were months when the air quality was unhealthy for sensitive population groups. Similar to the PM_{10} values, good air quality was found in the warm months, the air quality decreasing in the cold months. Furthermore, an air quality improvement from 2009 to 2019 was observed, especially for the cold months. The number of months with average AQI above 50 decreased from eight months in 2010 to four months in 2019, while instances of 90th percentile of AQI above 100 were identified in four months in 2010, but in only one month between 2011 and the present.

The high PM₁₀ levels reported all over Europe indicate that despite the progress made toward meeting the air quality standards, air pollution is still a critical issue. Even though Cluj-Napoca is not included in the top most polluted cities across Europe, there are still concerns about the air quality, especially during the cold season. In order to reduce the health risk associated with air pollution, effective air quality policies and tailored pollution abatement measures are needed, especially in cases of high urban agglomerations. In urban areas, most of the measures address the transport sector by encouraging the use of public transport or low pollution vehicles, such as electric cars or bicycles, and by limiting heavy vehicles' access to the city center and enhancing road infrastructures. The reduction of waste burning together with the shift towards low emission fuel-use for residential heating, along with emission-control and -law enforcing, could further reduce PM₁₀ emissions.

3.2. Human Health Risk Assessment

The exposure concentrations (EC) for the study periods, calculated based on monthly average PM₁₀ concentration, ranged from 7.1 µg/m³ (May 2019) to 44.7 µg/m³ (December 2009), while the EC calculated based on monthly 90th percentile of PM₁₀ ranged from 9.3 µg/m³ (May 2019) to 64.2 µg/m³ (January 2010). This data shows that the EC gradually decreased from 2010 to 2019.

The non-carcinogenic chronic HQ₁ calculated based on the EU limit value obtained for the residential best-case scenario ranged between 0.2 and 1.1 in all studied years, indicating potential non-carcinogenic chronic health risk in the first years of the study (2009 and 2010), and no health risks for the other study years (2011–2019). However, for the worst-case scenario, the HQ₁ values were higher (0.2–1.6), with monthly peaks with values above the safety benchmark being found in each year. The monthly variation of HQ₁ in the best-case (Figure 4a) and worst-case scenarios (Figure 4c) was similar to that in the PM₁₀ concentration, being higher in the cold months than in the warm months. In the best-case scenario, non-carcinogenic health risk was identified only in December 2009, while in the worst-case scenario, non-carcinogenic health risk was identified in each year, mainly in the winter seasons. In both exposure scenarios, over the years, a clear decreasing trend from 2009 to 2019 was observed.

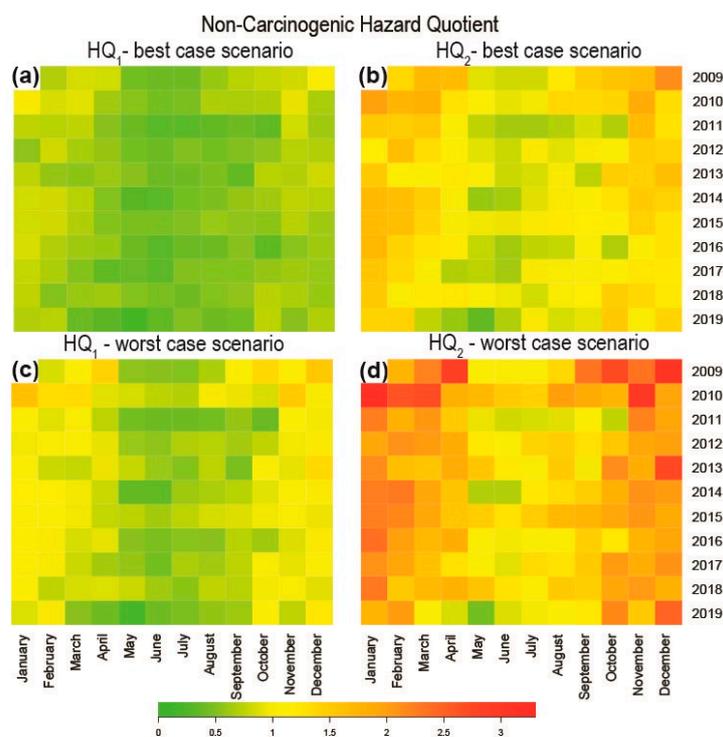


Figure 4. Temporal variation of hazard quotient (HQ) calculated based on EU limit values for best-case (a) and worst-case (b) scenarios, and calculated based on World Health Organization air quality guideline for best-case (c) and worst-case (d) scenarios in Cluj-Napoca city for 2009–2019.

The non-carcinogenic chronic HQ_2 calculated based on the WHO AQG obtained for the best-case scenario ranged between 0.4 and 2.1 in all studied years, indicating a potential non-carcinogenic chronic health risk between October and March in each year, while for the worst-case scenario, the HQ_2 values were higher (0.5–3.2), exceeding the security benchmark in most of the months. The monthly HQ_2 values, both in the best-case (Figure 4b) and worst-case scenarios (Figure 4d), were higher in the cold months than in the warm months, but slightly decreased from 2009 up to the present, without frequently falling below the security benchmark, indicating a non-carcinogenic risk for almost the whole period. The high HQ values suggest that environmental PM_{10} levels, although below the legislative thresholds, are likely to induce various chronic pathologies.

The differences between the human health risk measurements from the air quality monitoring stations for the period 2009–2019, in Cluj-Napoca city, were observed both for the best-case and worst-case scenarios (Figure 5). Thus, HQ_1 values, calculated based on the EU limit values, did not exceeded the unity threshold at any of the monitoring sites, suggesting no potential health risk, but they were higher at the urban (CJ-1) and suburban (CJ-3) background monitoring stations than at the traffic (CJ-1) and industrial (CJ-4) stations. Similarly, in the case of HQ_2 , the health risk was higher at the urban and suburban stations than at those monitoring the traffic and industrial background, but in this case potential non-carcinogenic risk might possibly appear. A similar pattern was observed for the worst-case scenario, both for HQ_1 and HQ_2 when the unity threshold was above 1, indicating the presence of potential non-carcinogenic risk.

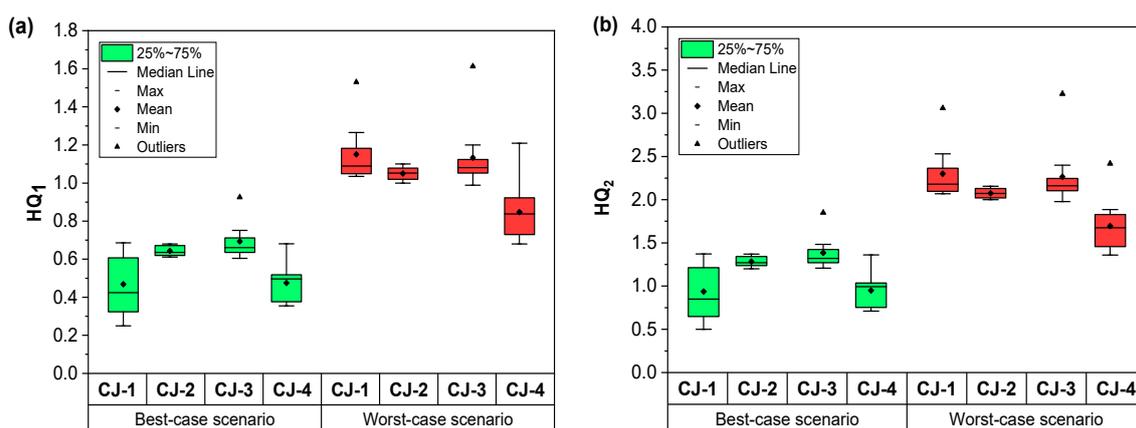


Figure 5. Variation of hazard quotient (HQ) calculated based on EU limit values (a) and World Health Organization air quality guidelines (b) between the air quality monitoring stations in Cluj-Napoca city.

Similar to Cluj-Napoca, a non-carcinogenic risk associated with PM_{10} exposure was reported in Krakow, Poland and Shanghai, China [10,50]. Opposite to Cluj-Napoca, no human health risk associated with exposure to PM_{10} for the general population, and a low risk for the sensitive population groups, were found for Pretoria West, South Africa [51].

Generally, during the warm months, the average HQ_1 and HQ_2 values were more or less constant, while during the cold months they varied widely. The obtained data suggest that, under the best-case scenario, presently, the inhabitants of Cluj-Napoca are unlikely to develop adverse health effects after PM_{10} exposure. However, in the worst-case scenario, the occurrence of adverse chronic health effects is still possible. The cumulative effects of PM_{10} with other air pollutants could increase the probability of manifesting adverse chronic non-carcinogenic health effects, although the additivity or synergism of the air pollutants is not clearly elucidated, and is influenced by the meteorological conditions [52–54]. A possible mitigation measure could be the reduction in the hours spent in urban environments with high levels of PM_{10} or other pollutants, and an increase in time spent in clean outdoor environments such as parks and green areas.

The obtained results highlighted the contribution of local meteorology and winter sources to the PM_{10} concentration. Although the PM_{10} concentrations for Cluj-Napoca city generally did not

exceed air quality standards, this study reveals that the human health risks resulting from exposure to air pollution are still possible. The health risk assessment method used in this study has a number of limitations, some of which are due to the fact that this approach focuses only on PM₁₀ without considering simultaneous exposure to other air pollutants, and thus underestimates the risk. The interactions between different pollutants were also neglected. The non-carcinogenic health risk was calculated based on several assumptions regarding the exposure time, exposure frequency and exposure duration that are not specific for the studied population, and neglects the different behaviors of individuals inside the studied population. Furthermore, we assumed that the average daily PM₁₀ concentrations measured by the for air quality monitoring stations are representative for the whole city. Despite these limitations, the findings may contribute to a better understanding of the potential health risks associated with human exposure to PM₁₀ in urban areas, providing valuable information for air quality managers and specialists. The next steps should include sources apportionment and the assessment of their contribution to PM₁₀ concentration, as well as assessing the carcinogenic human health risk of toxic compounds associated with PM₁₀.

4. Conclusions

The average annual PM₁₀ level in Cluj-Napoca city, ranging between 20.3 and 29.5 µg/m³, was below the legislative threshold, and had a decreasing trend from 2009 to 2019, while the annual maximum almost halved from 111 µg/m³ (2010) to 62.6 µg/m³ (2019). Generally, the monthly PM₁₀ concentrations were higher in the cold seasons compared with the warm seasons. The average monthly AQI ranged between 15 (May 2019) and 87 (December 2009), indicating good to moderate air quality. However, the 90th percentile indicated several months when the air quality was unhealthy for sensitive population groups. The non-carcinogenic chronic risk, calculated based on the EU limit value, obtained for the best-case scenario ranged between 0.2 and 1.1, indicating potential non-carcinogenic chronic health risk for 2009 and 2010, and no health risks for 2011–2019, while for the worst-case scenario, monthly values above the safety benchmark were found in each year. The non-carcinogenic chronic risk calculated based on the WHO AQG for the best-case scenario was higher, indicating a potential non-carcinogenic chronic health risk between October and March in each year, while for the worst-case scenario this exceeded the security benchmark in most of the months. The decreasing trend of the HQ values in the last decade could indicate the existence of air quality improvement measures; however, the implementation of more mitigation measures could further reduce the health risk, especially in the case of sensitive population groups. The measures must mainly target heating systems, taking into account the high concentrations of PM₁₀ during the winter, and in particular the type of fuel and its quality.

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