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The human health risk assessment of particulate air pollution ($PM_{2.5}$ and PM_{10}) in Romania

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ABSTRACT

Air pollution, especially the concentration of particulate matter ($PM_{2.5}$, PM_{10}) is a major issue and is the biggest environmental risk for early death. In the present study, we aimed to estimate the human health risk and to describe the spatial and temporal variation of particulate matter in Romania between 2009 and 2018. The average concentration of $PM_{2.5}$ and PM_{10} particulate matter in the eight studied regions varied between 17.01 and 22.91 μg m^{-3} and 23.02–33.29 μg m^{-3}, while the $PM_{2.5}/PM_{10}$ ratio varied between 0.52 and 0.76, respectively. The relative risk generated by PM_{10} in all-cause mortality had a significant variation between the regions, a relative risk of 1.017 in case of Bucharest and1.025 for western regions, with an average of 1.020 (± 0.002). According to our observations, a positive relative risk was identified in the case of cardiopulmonary and lung cancer morbidity mainly attributed to $PM_{2.5}$ exposure, hence the resulted risk for the country average values was 1.26 (± 0.023) and 1.42 (± 0.037), respectively. The results revealed that the excess risk and attributable fraction for cardiopulmonary mortality can be reduced by 26.7% and 21.0%. Analyzing the evolution of particulate matters and the possible health impacts of $PM_{2.5}$ and PM_{10} in all region of Romania a strong positive correlation was observed. Since the distributions of PM in different region had significant variation, more investigation is required to understand and decipher the most important regional emission sources for each region. In order to address this issue an in-depth investigation should separately analyze the regional characteristics of air pollution.

1. Introduction

It is widely accepted and supported by scientific evidence, that air pollution is a major global public health risk factor even in the XXI[st] century, when more links are revealed by research studies between a number of serious diseases among various age groups and air pollution. There is a strong correlation between air pollution and increased morbidity and mortality as well; according to the World Health Organization (WHO) report [41], air pollution is responsible for seven million people's death worldwide every year. Among the air pollutants, particles (PM) are considered as being the most dangerous substances released from different biogenic and anthropogenic sources or produced by secondary reactions taking place in the atmosphere [10,20,24,25]. Since $PM_{2.5}$ and PM_{10} have different physico-chemical properties the ratio between the fine and coarse particulate ($PM_{2.5}/PM_{10}$) can offer more details about the particulate source, origination process, and human health impact [19,2,38,39,4,7,9]. Coarse particles (PM_{10}) can get in deep into the respiratory tract, causing a serious respiratory disease [13,17,24,30,5]. However, due to the smaller size, the fine particles can pass via the respiratory tract and accumulate in the lungs causing different respiratory diseases as well as lung cancer [12,14,27,33,37].

According to the literature, the increased PM concentration is associated with increased morbidity and mortality in the population of the European Union, as a consequence the $PM_{2.5}$ reduced the average life span by 8.6 months [31]. Furthermore, according to different research outcomes the decreasing the $PM_{2.5}$ concentration level by 10 μg m^{-3} can increase life time by 0.61 year [1,18,34,35]. The $PM_{2.5}$ has a higher toxicity than PM_{10} thanks to the inflammation-causing capacity and oxidative stress [40]. Risk evaluation is a widely used method to evaluate the elevated risk of health issues in individuals exposed to high

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concentrations of particulate matter. From region to region the PM concentration and chemical composition show significant variation, which mainly depends on the geographical position, specific climate condition, anthropogenic activities and combustion sources [11,15, 21–23,29,3].

The particulate matter (PM), especially those with an aerodynamic equivalent diameter smaller than 2.5 μm are seldom studied due to the restricted availability of $PM_{2.5}$ related data. Previous studies have analyzed the human health effects of $PM_{2.5}$ and PM_{10} in Central-Eastern Europe, especially in Romania [8,36], but the human health assessment is yet to be studied.

To address this issue, the air pollution data was collected between 2009 and 2018 in order to analyze and decipher the temporal and regional distribution of airborne particulate matters and to calculate the relative risk, excess risk, and attributable death in eight different regions in Romania.

2. Materials and methods

2.1. Sampling site

Romania is a southeastern European country and the sixth/most populous member state of the EU with a population of around 19 million. The air pollution, especially in large cities, represents major concerns and it is well known that both short- and long-term exposure can lead to a wide range of diseases. In the present study, the human health risk assessment of particulate air pollution ($PM_{2.5}$ and PM_{10}) during 2009–2018, was carried out for Romania. The daily data of course (PM_{10}) and fine ($PM_{2.5}$) particulate matter concentrations were followed in eight different regions (B - Bucharest, C – Central, NE – North-East, NW – North-West, S – South, SE – South-East, SW – South-West, W – West) between 2009 January and 2018 December, except $PM_{2.5}$ in the Bucharest region, where the data are available only from 2016. The region concentration was determined by averaging data from all stations in that region where the measurements coverage was higher than 75% in the study period (Table 1).

where: *Num* - represents the number of monitoring stations in each region; *$PM_{2.5}$ - Mon. st. descr.* and *PM_{10} Mon. st. descr.* represent the $PM_{2.5}$ and PM_{10} monitoring station's names, *B, C, NE, NW, S, SE, SW, W* represent the Bucharest, Center, North East, North West, South, South West and West regions, respectively.

The daily data were obtained from the National Environmental Monitoring Agency network (*www.calitateaer.ro*), in total 33 ($PM_{2.5}$) and 122 (PM_{10}) monitoring station data were processed (Fig. 1.). In order to determine the pollution level variation, temporal and regional distribution, descriptive statistics and time series analysis were used. The coarse and fine particulate ratio ($PM_{2.5}/PM_{10}$) was calculated for each region. In order to decipher the seasonal variation, the data were classified using a four-season classification as follows: *a.* Spring (March-May), *b.* Summer- warm period (June-August), *c.* Autumn (September-November), *d.* Winter- cold period (December-February).

Table 1
The monitoring stations in Romania.

Reg.	Num.	$PM_{2.5}$ Mon. st. descr.	Num.	PM_{10} Mon. st.descr.
B	4	$B_{1,5,6,7}$	8	$B_{1,2,3,4,5,6,7,8}$
C	4	BV_2, HR_1, MS_1, SB_1	12	$BV_{1,2,3,4}$, CV_1, HR_1, $MS_{1,2,3,}$ $SB_{1,3,4}$
NE	5	BC_1, BT_1, $IS_{1,}$ NT_1, SV_1	14	$BC_{1,2}$, BT_1, $IS_{2,4,5,6}$, $NT_{1,3}$, $SV_{1,2,3}$, $VS_{1,2}$
NW	4	BH_1, CJ_2, MM_2, SM_1	16	$BH_{1,2,4}$, BN_1, $CJ_{1,2,3,5}$, $MM_{1,2,3,4,5}$, SJ_1, $SM_{1,2}$
S	5	AG_2, GR_2, PH_2, $TR_{3,5}$	23	$AG_{1,2,3,4,6}$, $CL_{1,2,3}$, $DB_{1,2}$, $GR_{1,2,3}$, $IL_{1,2}$, $PH_{1,2,3,5,6}$, $TR_{1,2,4}$
SE	4	BR_2, BZ_1, CT_2, GL_2	20	$BR_{1,2,3,4}$, $BZ_{1,2}$, $CT_{1,2,3,4,5,7}$, $GL_{1,2,3,4}$, $TL_{1,2,3}$, VN_1
SW	4	$DJ_{2,6}$, MH_1, VL_1	11	$DJ_{1,2,3,5,6}$, $GJ_{1,2,3}$, MH_1 OT_1 VL_1
W	3	AR_2, CS_5, TM_2	18	$AR_{1,2,3}$, $CS_{1,2,3,4,5}$, $TM_{1,2,3,5,6}$
Total	**33**		**122**	

2.2. Health risk assessment (HRA)

2.2.1. Health risk assessment methodology for short-term effect of PM_{10}

In order the determine the short-term exposure to PM_{10}, the relative risk (*RR*) for all-cause mortality was calculated according to Ostro [32] (Eq. 1). The relative risk for all-cause mortality was calculated if the PM_{10} concentration was higher than the background level (10 μg m^{-3}). A risk function coefficient of 0.0008 was used (95% CI: 0.0006–0.0010).

$$RR = exp[\beta(X - X_0)] \tag{1}$$

where: *X*- represents the annual mean concentration of PM_{10} (μg m^{-3}), X_0- represents the background concentration of PM_{10} (10 μg m^{-3}), *β*- is the risk function coefficient.

2.2.2. Health risk assessment methodology for short-term effect of $PM_{2.5}$

The relative risk associated with $PM_{2.5}$ was calculated separately for cardiopulmonary and lung cancer mortality for habitants over 30 years old [32] using Eq. 2.

$$RR = [(X + 1)/(X0 + 1)]\beta \tag{2}$$

where: *X*- represents the annual mean concentration of $PM_{2.5}$ (μg m^{-3}), X_0- is the background concentration of $PM_{2.5}$ (3 μg m^{-3}), and *β*- is the risk function coefficient. The applied *β* coefficients for the cardiopulmonary and lung cancer mortality was 0.15515 (95% CI: 0.0562–0.2541) and 0.23218 (95% CI: 0.08563–0.37873), respectively.

Furthermore, using the determined relative risk (*RR*), the attributable fraction (*AF*) was calculated [32] (Eqs. 3–4).

$$AF = (RR - 1)/RR \tag{3}$$

The calculated *AF* value indicates deaths ratio from the respective disease, which could be avoid if the concentration levels were lower by 10 μg m^{-3} and 3 μg m^{-3} for PM_{10} and $PM_{2.5}$, respectively.

$$ER = (RR - 1) \tag{4}$$

The exposure to ambient $PM_{2.5}$ and PM_{10} was estimated as a population-weighted annual average in Romania. The calculated exposure to PM was used as input in the health impact assessment to determine the total number of premature deaths.

3. Results

3.1. Statistical analysis of the data

In the studied period (2009–2018), the average concentration of fine and coarse particular matter in the eight studied regions varied between 17.01 and 22.91 μg m^{-3} and 23.02–33.29 μg m^{-3}, respectively. In order to decipher the trends, descriptive statistical analyses were conducted for outdoor $PM_{2.5}$ and PM_{10} mass concentrations - determined by gravimetric method STAS 12341. The highest multiannual mean concentration of the $PM_{2.5}$ and PM_{10} was measured in the Bucharest region (22.91 μg m^{-3} and 33.29 μg m^{-3}), followed by SW (20.40 μg m^{-3} and 30.85 μg m^{-3}) (Table2). The results show that the mass percentage for coarse particles is higher than the fine particles in all regions.

Seasonal and spatial distribution of $PM_{2.5}$ and PM_{10} levels in the studied regions are presented in Fig. 2. The results revealed higher PM concentrations during the cold period, especially in January and December, while the lowest levels were recorded during summer and no significant differences were observed between regions. Quantitatively, the difference between the highest and lowest monthly PM concentration was 1.77 times for PM_{10}, and 2.76 times for $PM_{2.5}$ respectively.

Due to the different physico-chemical characteristics of coarse and

Fig. 1. Sampling regions (Romania). where: the numbers represent the regions, including Bucharest (8) as well: 1-North-East, 2-South-Est, 3-South, 4-South-West, 5-West, 6-North-West and 7-Central region.

Table 2
Descriptive statistical analysis.

	Region	min	25 P	med	75 P	max	mean	stdev	count	95% CI	CV
$PM_{2.5}$, $\mu g\ m^{-3}$	B*	0.94	13.76	19.21	28.39	129	22.91	14.7	917	21.95–23.86	0.64
	C	0.36	9.06	13.3	19.62	138.7	17.01	13.73	3502	16.56–17.47	0.81
	NE	2	11.79	16.37	23.12	119.5	19.24	11.47	3567	18.86–19.61	0.6
	NW	0	10.08	14.99	23.21	107.5	18.04	11.23	3533	17.67–18.41	0.62
	S	1.6	11.24	14.9	20.95	81.02	17.51	9.59	3564	17.19–17.82	0.55
	SE	0.58	8.3	11.34	15.66	143.6	13.38	8.88	3319	13.08–13.68	0.66
	SW	0.91	11.9	16.99	24.69	118.4	20.4	13.51	3392	19.95–20.85	0.66
	W	1	8.99	13.93	21.59	132.6	17.21	12.46	3330	16.79–17.64	0.72
PM_{10},$\mu g\ m^{-3}$	B	3	22.12	29.75	39.67	230.3	33.29	17.72	3562	32.70–33.87	0.53
	C	3.66	14.82	21.15	30.21	174.1	24.57	15.01	3651	24.08–25.05	0.61
	NE	5.57	19.62	25.71	33.28	120.8	27.69	11.99	3651	27.30–28.08	0.43
	NW	3.45	15.28	21.48	30.46	127.6	24.29	12.27	3649	23.90–24.69	0.51
	S	5.97	20.31	26.28	34.37	92.61	28.57	11.65	3651	28.20–28.95	0.41
	SE	2	18.76	22.99	28.1	93.38	23.95	7.61	3648	23.70–24.20	0.32
	SW	3.55	20.16	27.22	37.07	171.8	30.85	16.33	3631	30.32–31.38	0.53
	W	5.04	15.34	20.94	28.09	99.52	23.02	10.63	3647	22.68–23.37	0.46

where: *min* - minimum; *25 P* - 25th percentile; *med* - median; *75 P* - 75th percentile; *max* - maximum, *mean* - average, *stdev* - standard deviation; *count* - number of samples; *95% CI* - confidence interval; *CV* -coefficient of variation. * the data are available only from 2016.

fine particulate, the $PM_{2.5}/PM_{10}$ ratio was also calculated. The spatial distribution of the ten-year mean of $PM_{2.5}/PM_{10}$ ratios in eight Romanian regions is presented in Fig. 3. The results show significant spatial distribution differences between regions, with a wide variability of 0.52 and0.76. The highest ratio (0.76) was found in the most polluted region (Bucharest), indicating that high $PM_{2.5}$ contributions come from industrial emissions, which has also been found in the well-developed industrialized western regions (NW, W) with higher $PM_{2.5}/PM_{10}$ ratio (0.73).

3.2. Health risk assessment

The relative risk (RR), excess risk (ER) and an attributable fraction (AF) were calculated for all-cause mortality in case of each region using the daily PM_{10} data. The average relative risk caused by PM_{10} for all-cause mortality was 1.020 ($\pm$ 0.0024), with variability from 1.017 in the West region to 1.025 in the Bucharest region (Fig. 4).

A positive relative risk for cardiopulmonary and lung cancer disease was observed which is mainly attributed to $PM_{2.5}$ exposure; according to the national average values, the relative risk was 1.26 ($\pm$ 0.023) and 1.42 ($\pm$ 0.037), respectively (Fig. 5).

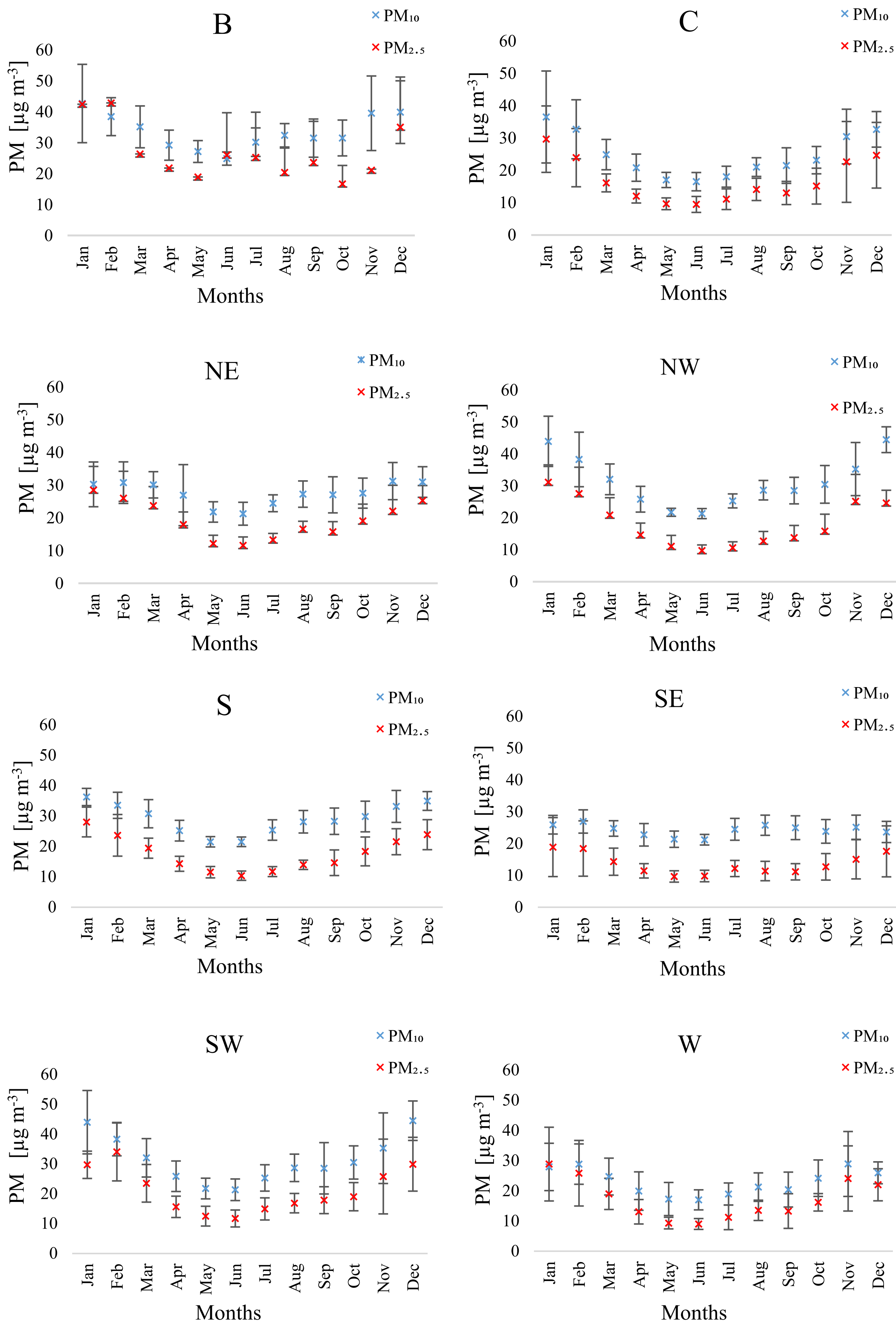

Fig. 2. Multiannual monthly mean $PM_{2.5}$ and PM_{10} concentration variation, averages are represented by blue and red *x*, and the ends of the whiskers represent the minimum and maximum standard deviations.

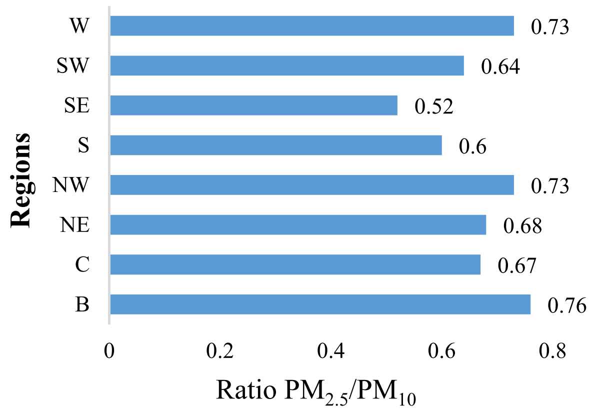

Fig. 3. The $PM_{2.5}/PM_{10}$ ratio variations in different regions.

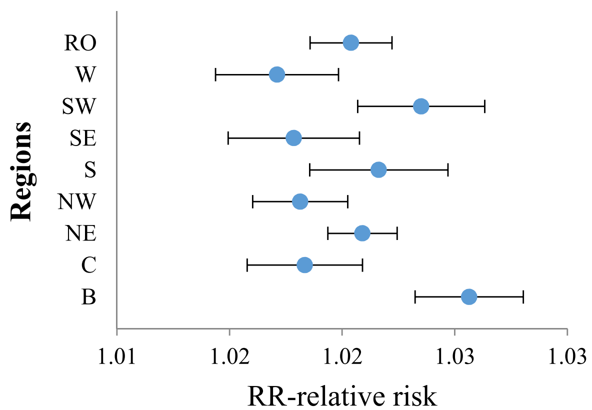

Fig. 4. PM_{10} all-cause mortality, where blue dots represent the means and the whiskers' ends show the standard deviations.

The calculated excess risk (ER) and the attributable fractions (AF) for all-cause mortality were evaluated for each region using daily PM_{10} data. The results revealed that the excess risks varied between 1.71% and 2.5% (Table 3).

Furthermore, the excess risk (ER) and attributable fraction (AF) for cardiopulmonary and lung cancer mortality were also determined for the long-term exposure to $PM_{2.5}$ and are presented separately (Tables 4–5). Results show that for cardiopulmonary mortality, the ER and AF in Romania varied between 21.4% and 32.6%, 17.5–24.6%, respectively.

4. Discussions

During the studied period the average $PM_{2.5}$ and PM_{10} concentrations were 1.82 and 1.35 times higher than the annually acceptable limit specified by the WHO Air Quality Standard. Over the years the PM concentrations show a strong seasonal variation, the maximum level was detected in the cold period, and the minimum in summer during the warm period. Therefore, the PM concentrations

Table 3
Human health risk calculation based on the PM_{10} concentration in different region for *all-cause mortality* associated with short-term PM_{10} exposure.

Region	ER (%)	ER ((95% CI)		AF (%)	AF (95% CI)	
B	2.56	2.42	2.71	2.5	2.36	2.64
C	1.83	1.02	1.02	1.8	1.65	1.95
NE	2.09	1.02	1.02	2.05	1.96	2.14
NW	1.81	1.02	1.02	1.78	1.65	1.91
S	2.16	1.02	1.02	2.12	1.93	2.3
SE	1.79	1.02	1.02	1.75	1.58	1.93
SW	2.35	1.02	1.03	2.3	2.13	2.46
W	1.71	1.02	1.02	1.68	1.52	1.85
RO	2.04	1.02	1.02	2	1.89	2.11

where: *B, C, NE, NW, S, SE, SW, W* represents the regions; *RO* - represents the country average; *ER* - excess risk; *AF* - attributable fraction and *95% CI* - confidence level.

Table 4
Human health risk calculation based on the $PM_{2.5}$ concentrations in different region for *cardiopulmonary mortality* associated with long-term exposure to $PM_{2.5}$.

Region	ER (%)	ER ((95% CI)		AF (%)	AF (95% CI)	
B	32.6	28.3	37	24.6	22.1	27
C	25.9	23.6	28.3	20.5	19	22
NE	28.5	27.5	29.6	22.2	21.6	22.8
NW	27.2	25.6	28.8	21.4	20.4	22.3
S	26.7	25.3	28.1	21.1	20.2	21.9
SE	21.4	18.9	23.8	17.5	15.9	19.2
SW	29.3	27.2	31.5	22.6	21.3	23.9
W	26.2	23.6	28.8	20.7	19	22.4
RO	26.7	25.3	28.1	21	20.2	21.9

Table 5
Human health effect calculation based on the $PM_{2.5}$ concentrations in different regions for *lung cancer* associated with long-term exposure to $PM_{2.5}$.

Region	ER(%)	RR ((95% CI)		AF (%)	AF (95% CI)	
B	52.6	45.1	60.1	34.4	31.2	37.5
C	41.2	37.3	45.2	29.1	27.1	31.1
NE	45.6	43.8	47.3	31.3	30.4	32.1
NW	43.3	40.6	46.1	30.2	28.9	31.5
S	42.5	40.1	44.9	29.8	28.6	30.9
SE	33.6	29.6	37.6	25	22.7	27.3
SW	47	43.3	50.6	31.9	30.1	33.6
W	41.6	37.3	45.9	29.2	27	31.5
RO	42.5	40.2	44.9	29.8	28.6	30.9

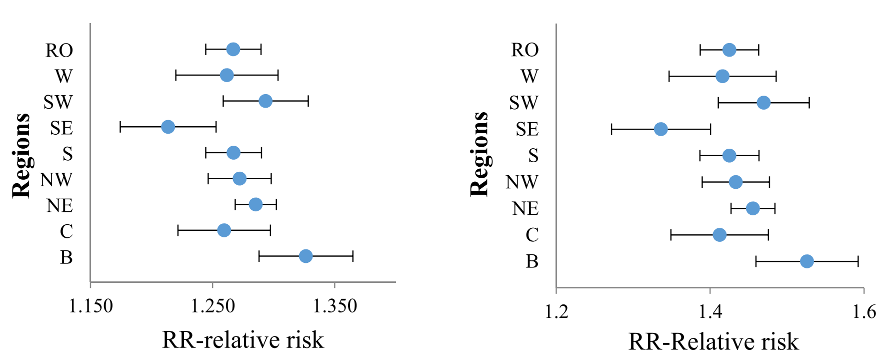

Fig. 5. $PM_{2.5}$ - cardiopulmonary disease (left) and $PM_{2.5}$ - lung cancer (right), where blue dots represent the means and the whiskers' ends show the standard deviations.

show a clear decline from spring to summer, reaching the lowest concentration in the warm period, while the highest level was observed in the winter period when biomass burning is significant due to the heating season and when vertical mixing is reduced [17,26,28,39,42].

As it was stated earlier, air pollution is more severe in densely populated cities and regions with industrial background and specific microclimate condition. The results revealed similar tendency in our study, with increased air pollution in Bucharest and the South-West region which is due to densely populated cities and strong industrial background nearby. On the other hand, the PM_{10} fraction partially could also be formed from the coagulation of fine particulates.

Analyzing the particulate matter concentration for a ten-year period in Romania the results clearly indicate a strong seasonal characteristic in all eight regions. According to our observations, the elevated pollution level is mainly ascribed to increased fossil burning and traffic; moreover, in the winter period, adverse meteorological circumstances like thermal inversion, frequent fog are also essential factors, especially in case of intra-mountain basin, hence favoring the accumulation of air pollutants [13,16]. Since the source of fine particles ($PM_{2.5}$) and coarse particles (PM_{10}) might be different; in order to decipher the main sources the $PM_{2.5}/PM_{10}$ ratio analysis is a well-known approach in the identification of particle pollution origin [43]. The results revealed highest ratio in regions with massive industrial background which indicates increased $PM_{2.5}$ contributions from industry. Furthermore, the relative risk calculations showed a positive risk for cardiopulmonary and lung cancer disease due to exposure to $PM_{2.5}$ and for all types of mortality in case of PM_{10}.

The higher excess risk was found in the Bucharest region which means that the habitants exposed to the actual PM_{10} concentration in Bucharest have more chance to experience different health issues by 2.56% than habitants in a group that is exposed to a background concentration of 10 $\mu g\ m^{-3}$ (PM_{10}). The lowest excess risk was found in the western regions with 1.71% more harmful effect compared to the background level, where is no industrial pollution. According to the calculated excess risk and attributional fraction, the all-cause mortality can be reduced by 2.04% and 2.00%, respectively, if the PM_{10} concentration levels are maintained at around 10 $\mu g\ m^{-3}$. If the annual concentration of $PM_{2.5}$ will be kept around 3 $\mu g\ m^{-3}$ the excess risk and attributional fraction for cardiopulmonary mortality will decrease by 26.7% and 21.0%, respectively.

Furthermore, the results are fairly similar to those reported by [6], according to their observations regarding the risk of air pollution in Lisbon, the lung cancer mortality rate could be prevented by 29.8% [6]. In case of Romania the excess risk and attributional fraction for lung cancer mortality can be prevented by 42.5% and 21.0%, respectively, if the $PM_{2.5}$ concentration levels will be kept around 3 $\mu g\ m^{-3}$.

Citizens of crowded cities (Bucharest, Iași, Brașov) have been exposed almost continuously to unhealthy levels of PM_{10} since 2007 and the measures taken to reduce air pollution have been ineffective, and this the main reason why Romania has now been condemned by European Commission. In the future, further analyses are necessary and will be carried out to examine emission sources and the geographical differences between the regions.

5. Limitations and strengths

The main limitation of this study was the use of the descriptive analytic methods. In order to address all aspects of the relation between different air pollutants and meteorological factors and the health adverse effects and health endpoint in the population further epidemiological studies are necessary. In this manner, we can decipher and understand the health effect mechanism of major air pollutants in different regions of Romania. By estimating the relative risk (RR), excess risk (ER) and attributional fraction (AF) during $PM_{2.5}$ and PM_{10} exposure a different aspect of air pollution have been illuminated, hence the results from our study can be used as support in the future for the development of environmental regulations and policies.

6. Conclusions

During the studied period (2009–2018), the average concentration of $PM_{2.5}$ and PM_{10} in the eight studied Romanian region was higher than the annually acceptable limit established by national and EU regulations. Significant differences were revealed between regions, namely, highest in the Bucharest region and lowest in the South-East region. Human health risks associated with exposure to particular matters ($PM_{2.5}$, PM_{10}) were estimated in the current study, and according to the results, the ratio between the fine and coarse particular matter in Romania warried between 0.52 and 0.76. The calculated relative risk for $PM_{2.5}$ (cardiopulmonary and lung cancer) was significantly higher than the relative risk caused by PM_{10} for all-cause mortality. Moreover, the relative risk calculated from $PM_{2.5}$ concentrations (1.26) was more than one order of magnitude higher than for the PM_{10} (1.02). The result showed that the exposure to particulate matters represent important potential risk for many health issues, which need to be minimized by environmental regulation. In the light of these facts, Romania still needs to improve its environmental protection policy and environmental protection actions as well, in order to reduce the emission of air pollutants with potential health effects.

CRediT authorship contribution statement

Katalin Bodor: Methodology, Validation, Formal analysis, Investigation, Resources, Writing – original Draft, **Róbert Szép:** Conceptualization, Methodology, Validation, Investigation, Supervision, **Zsolt Bodor**: Conceptualization, Methodology, Software, Formal analysis, Investigation, Visualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Collegium Talentum

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