UNIVERSITY OF PÉCS

Doctoral School of Chemistry

# Spatial and temporal distribution of particulate matter ($PM_{2.5}$, $PM_{10}$) trace element concentration (As, Cd, Ni, Pb) and human health effect in Romania

**PhD thesis**

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## Introduction

Air pollution is a problem that is as old as mankind, in the beginning the level of air pollution was a local or at most a regional problem, but today it is a global problem. The drastic increase in air pollution is due to the rate of economic development.

Population growth and the consequent increase in anthropogenic activities (transport, energy production, industry, etc.) are all major contributors to the current high levels of pollution. In relative terms, the burning of fossil fuels represents the most significant burden on the environment. Air pollutants include both anthropogenic and natural air pollutants. In a global effort to reduce emissions, a number of remedial measures have been introduced, such as divesting from fossil fuels and adopting a more environmentally conscious approach to life.

The harmful effects of air pollution on human health have been proven by numerous studies and research, both in the short- and long-term, contributing to the development of diseases and premature death. It can be said that air pollution has a significant negative impact on the environment, public health and, indirectly, on the economy, and it is therefore highly advisable to introduce integrated policy programs aimed at reducing and eliminating the causes and consequences of air pollution.

Short-term symptoms of exposure to air pollution include itchy eyes and nose, sore throat, wheezing, coughing, shortness of breath, chest pain, headache, nausea and respiratory tract infections. Long-term exposure to high concentrations of $PM_{10}$ can cause a range of health effects and premature death, such as lung cancer, cardiovascular disease, chronic respiratory diseases and allergies. $PM_{10}$ can reach the bronchial and alveolar regions of the trachea, $PM_{2.5}$ enters the lungs and accumulates, and ultrafine particulate matter can even penetrate blood vessels, thus damaging human health. The World Health Organization (WHO) reports that air pollution is one of the greatest environmental risks to human health, with 22% of illnesses and deaths being linked to air pollution.

Furthermore, in Romania, many factors contribute to the variation of air pollution levels, such as the complex natural and socio-geographical diversity, the diversity of topography: all types of topography (plains, hills, plateaus, mountains) are found in almost equal proportions. The Carpathian Mountains range forms the backbone of the country. Meanwhile, the location and development of towns are linked to the diversity of the subsoil. All these economic, geographical, and social factors have an impact on particulate matter emissions. Furthermore, there is evidence of cross-border pollution in the region, both natural and anthropogenic.

## 1. Research objectives

My research studies the temporal variation of particulate matter ($PM_{2.5}$ and $PM_{10}$) concentrations in Romania in regional and national contexts, and comparative studies on their health effects were conducted. The objectives of the multi-stage research were the following:

- Spatial and temporal evolution of particulate matter ($PM_{2.5}$ and $PM_{10}$) in all regions of Romania and calculation of relative risk (RR).
- Analysis of spatial and temporal variations of As and heavy metals (Cd, Ni, Pb) from $PM_{10}$ in all Romanian regions and assessment of hazard quotient (HQ) and cancer risk (CR) in children and adults for three different exposure routes: inhalation, ingestion and dermal absorption.
- Comparative study on the distribution of trace element concentrations of $PM_{10}$ and estimation of their health effects in one of the most industrially polluted areas of Romania, Copșa Mică in Sibiu County.
- Temporal analysis and health effects of heavy metals (Cd, Ni, Pb) in fine ($PM_{2.5}$) and coarse ($PM_{10}$) particulate matter in Bucharest.
- Investigation of the effectiveness of $PM_{10}$ concentration reduction by precipitation under windless conditions in the Ciuc Basin, taking into account precipitation intensity, duration and meteorological parameters.
- Finally, an epidemiological study was conducted to examine how meteorological parameters and $PM_{10}$ particulate matter concentrations in the Ciuc Basin have evolved over time in comparison with daily admissions to the County Emergency Hospital in Miercurea Ciuc for respiratory and cardiovascular diseases.

## 2. Materials and methods

The study examined the spatial and temporal evolution of $PM_{10}$ concentration and its As and heavy metals (Cd, Ni, Pb) concentration in all regions of Romania. Daily data were provided from the Romanian air quality monitoring network. The concentration of the particles was determined by the gravimetric method according to the EN 12341 standard. The reference method for measuring As, Cd, Ni and Pb concentrations was carried out according to SR EN 14902 "Ambient air quality from the fraction of $PM_{10}$ particulate matter".

In order to analyze the spatial and temporal variation of air pollution levels, the following statistical methods were used: descriptive statistics, box plot analysis, heat map, Pearson, Spearman correlation, principal component analysis, Hierarchical Cluster Analysis and HYSPLIT combined with concentration weighted trajectories (CWT). To study the health effects, short- and long-term health risk assessment models for $PM_{2.5}$ and $PM_{10}$ particulate matter were calculated:

$$RR = exp[\beta(X - X_0)] \quad (1)$$

where: $X$ - is the annual average concentration of $PM_{10}$ ($\mu g\ m^{-3}$), $X_0$ - is the background concentration of $PM_{10}$ (10 $\mu g\ m^{-3}$), $\beta$ - is the risk coefficient.

$$RR = [(X + 1)/(X_0 + 1)]\beta \quad (2)$$

where: $X$ - is the annual average concentration of $PM_{2.5}$ ($\mu g\ m^{-3}$), $X_0$ - is the background concentration of $PM_{2.5}$ (3 $\mu g\ m^{-3}$), and $\beta$ - is the risk coefficient.

Health risk indicators for As and heavy metals (Cd, Ni, Pb) in particulate matter were calculated for three different routes: inhalation, ingestion and dermal absorption:

$$CDI_{ing} = (C \times IngR \times EF \times ED \times CF)/(BW \times AT) \quad (3)$$

$$EC_{inh} = (C \times ET \times EF \times ED)/AT \quad (4)$$

$$DAD_{derm} = (C \times SA \times AF \times EV \times ABS \times EF \times ED \times CF)/(BW \times AT) \quad (5)$$

where: $CDI_{ing}$ - daily chemical intake by ingestion, $EC_{inh}$ - inhalation exposure concentration, $DAD_{derm}$ - dose absorbed through the skin.

The risk assessment was analysed separately for non-carcinogenic and carcinogenic effects. Non-carcinogenic risk is characterised by the hazard quotient (HQ) and carcinogenic effects by the cancer risk (CR). The HQ and CR risks for As and heavy metals in $PM_{10}$ can be expressed using the following equations for inhalation, ingestion and dermal contact:

$$HQ_{ing} = CDI/RfDo \quad (6)$$

$$HQ_{inh} = EC/(RfCi \times 1000\ \mu g\ mg^{-1}) \quad (7)$$

$$HQ_{derm} = DAD/(RfDo \times GIABS) \quad (8)$$

$$CR_{ing} = CDI \times SFo \quad (9)$$
$$CR_{inh} = IUR \times EC \quad (10)$$
$$CR_{derm} = DAD \times (SFo/GIABS) \quad (11)$$

For the CR assessment, chemicals were considered to pose a carcinogenic risk if the CR values were between $10^{-4}$ and $10^{-6}$.

To study the washout effect of precipitation, the effectiveness of the atmospheric cleaning effect was investigated for rainfall of different intensities and durations: windless < 1 m $s^{-1}$; three different rainfall intensity levels (low: 0.2-0.4 mm $h^{-1}$, moderate: 0.4-3.9 mm $h^{-1}$, and heavy: > 3.9 mm $h^{-1}$; rainfall duration 1-6 h). The analyses were carried out separately for the cold (October-March) and warm (April-September) periods, respectively.

As a concluding study, an epidemiological study was conducted to compare the evolution of $PM_{10}$ concentrations in the Ciuc Basin with respiratory, cardiovascular and meteorological parameters and patients admitted to the County Emergency Hospital in Miercurea Ciuc. The seven studied diseases were: 1. lung cancer - LC (ICD-10 codes C33-C34); 2. acute myocardial infarction - AMI (ICD-10 code I21); 3. ischaemic heart disease - IHD (ICD-10 codes I20-I25, except I21); 4. chronic cardiopulmonary disease - CCP (ICD-10 code I27.9); 5. Upper respiratory tract infections - URTI (ICD-10 code J00-J06); 6. Pneumonia - P (ICD-10 code J12-J18); 7. Chronic obstructive pulmonary disease - COPD (ICD-10 code J44). Hospital admissions for men and women were analysed for five age groups ([0-5], (5-14], (14-40], (40-60] and 60+ years). The following meteorological parameters were studied and calculated: air temperature, precipitation, relative humidity, temperature-humidity index (THI), wind chill equivalent diagram index (WCT).

**3. Results**

In the PhD thesis, the spatial and temporal distribution of trace element concentrations (As, Cd, Ni, Pb) in particulate matter ($PM_{2.5}$, $PM_{10}$) and their impact on human health in Romania is presented.

The achieved results are summarized in the following points:

1. *Time series analysis of the particulate matter and trace elements*

There is a significant difference in the temporal distribution of the particulate matter and the trace elements. The seasonal pattern of $PM_{10}$ and the trace elements As, Cd and Pb shows high concentrations in winter and low concentrations in summer, except for Ni, which shows the opposite trend. The seasonal variation of pollutants is strongly correlated with various factors, mainly meteorological parameters and emission sources, therefore the higher $PM_{10}$ concentration in winter can be explained by residential heating and the presence of unfavorable meteorological conditions, such as thermal inversion, fog and low boundary layer height. On the other hand, the elevated Ni concentration in summer is mainly associated with industrial production and traffic intensity.

2. *Spatial evolution of particulate matter and trace elements*

The highest concentrations of trace elements were identified in Sibiu County, As: 1.69 ng $m^{-3}$, Cd: 2.32 ng $m^{-3}$, Ni: 5.19 ng $m^{-3}$, Pb: 0.292 ng $m^{-3}$, mainly in Copșa Mică, one of the most polluted regions in Romania. Air pollutant concentrations in Copșa Mică are several times the national average: Pb 10.5 x, Cd: 4 x, As and Ni 2.5 x. The highest concentrations of particulate matter ($PM_{10}$) are found in Iași County and Bucharest > 32 µg $m^{-3}$.
The ratio of $PM_{10}$ and fine particles also shows a high variability (0.52-0.76). The highest ratio (0.76) was found in the most polluted region of Bucharest, indicating that a large part of $PM_{2.5}$ comes from industrial emissions, and high values were also observed in the developed industrial regions (NW, NY), where the $PM_{2.5}/PM_{10}$ ratio is high (0.73). The lowest ratio was found in the DK region (0.52).

3. *Health effects of particulate matter and trace elements:*

The average relative risk of $PM_{10}$ for all-cause mortality was 1.020 (±0.0024), ranging from 1.017 in the Western region to 1.025 in the Bucharest region. Furthermore, a high relative risk of heart and lung cancer was observed, mainly due to $PM_{2.5}$ exposure, with national averages of 1.26 (±0.023) and 1.42 (±0.037), respectively.
Based on hazard ratio and cancer risk calculations, inhalation HQ values in Romania were

higher than the safe limit (=1) for both adults and children. The highest non-carcinogenic risk was detected for Cd (9.53). Ni also showed a non-carcinogenic risk with a value of 1.92. Taking into account that the sum of the three trace elements (HI) reached $1.22\times10^{1}$, inhalation of a mixture of trace elements no longer poses a non-carcinogenic health risk. The cancer risk values calculated for Romania for inhalation increased to $2.00\times10^{-5}$ for children and $8\times10^{-5}$ for adults, indicating a cumulative cancer risk from exposure to mixtures of elements by inhalation. For Copșa Mică the cumulative cancer risk via inhalation in adults and children was $9.34\times10^{-5}$ and $2.34\times10^{-5}$, respectively.

*4. The study of the wash-out effect of particulate matter*

The concentration of $PM_{10}$ in the rainy periods was lower than in the case of the non-precipitation period, and there was a noticeable difference in the average $PM_{10}$ concentrations in the cold and warm periods: 2.8 μg $m^{-3}$ and 2 μg $m^{-3}$, respectively. The greatest decrease in $PM_{10}$ concentration was observed in all cases investigated in the cold season with low and medium rainfall intensity, after 6 hours of continuous rain (35.61%, 32.46%).

*5. Results of the epidemiological study*

The diseases are most common in the over-60 age group, accounting for 41.3% of all cases. The second highest number of referrals was observed in children under 5 years of age, with 28.9%, followed by patients (40-60 years), accounting for 20.4%. The most common disease was upper respiratory tract infections (URTI) with 13.55%, with children aged [0-5] years being the most affected age group with 9.12%. The next most common disease was COPD with 12.5%, followed by LC with 9.01%, CCP with 6.99%, IHD with 6.73% and AMI with 2.5%. A significant difference was found in the number of referrals between men and women. Using Pearson's correlation, a moderate correlation was found between $PM_{10}$ and P (0.51) and $PM_{10}$ and URTI (0.39). $PM_{10}$ showed a moderate negative correlation with boundary layer (-0.61). When determining the source areas for particulate matter, concentration-weighted trajectory analysis (CWT) highlights the main potential source areas that influence regional $PM_{10}$ concentrations. The main potential source areas are located to the north, northwest and southeast. Ukraine and the Republic of Moldova are identified as moderate source areas.

**Thesis points:**

The PhD thesis presents the spatial and temporal distribution of particulate matter and its trace elements in Romania, highlighting the industrially most polluted region Copșa Mică, and the capital Bucharest. In addition, the precipitation washout mechanisms and epidemiological studies were studied in the Ciuc Basin.

1. Regarding the temporal evolution of particulate matter, the seasonal variation of pollutants is strongly correlated with various factors, mainly meteorological parameters and emission sources.
2. The relative risk attributed to $PM_{2.5}$ (cardiovascular disease and lung cancer) was significantly higher than the relative risk attributed to $PM_{10}$ for all-cause mortality. The total trace elements in fine particulate matter ($PM_{2.5}$) had a higher enrichment than in coarse particulate matter ($PM_{10}$).
3. The annual concentrations of trace elements in Copșa Mică are several times higher than the national average (As: 2.64 times, Cd: 4.01 times, Ni: 2.44 times, Pb: 10.52 times).
4. The potential carcinogenic risk from inhalation and dermal absorption exceeded the acceptable level of carcinogenicity, indicating an increased risk of cancer in adults in the study area. Because of the longer exposure, adults were more likely to have a carcinogenic risk than children.
5. The largest decreases in $PM_{10}$ concentrations were observed in the cold season, with low and moderate rainfall intensities, after 6 hours of continuous rainfall in all cases studied.
6. Short-term exposure to airborne particulate matter in the Ciuc Basin was positively associated with upper respiratory tract diseases and pneumonia. Long-term exposure showed that $PM_{10}$ had a negative effect on cardiovascular disease, but with a significant delay in dose-response. In addition, $PM_{10}$ and comfort indices (WCT, THI) play an important role.

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