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# **Environmental Pollution**



journal homepage: www.elsevier.com/locate/envpol

# Health risk assessment of the European inhabitants exposed to contaminated ambient particulate matter by potentially toxic elements \*

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# ARTICLE INFO

Keywords: Air pollution Particulate matter Inhalation Health risks Cancer risk Potential toxic elements (PTEs) Europe

# ABSTRACT

 $PM_{10}$ -associated potential toxic elements (PTEs) can enter the respiratory system and cause health problems. In the current study, the health risk indices caused by  $PM_{10}$  inhalation by adults, children, and infants in 158 European cities between 2013 and 2019 were studied to determine if Europeans were adversely affected by carcinogenic and non-carcinogenic factors or not. The Mann–Kendall trend test examined  $PM_{10}$ 's increasing or decreasing trend. Random Forest analysis was also used to analyse meteorological factors affecting  $PM_{10}$  in Europe. Hazard quotient and cancer risk were estimated using  $PM_{10}$ -associated PTEs. Our results showed a decline in continental  $PM_{10}$  concentrations. The correlation between  $PM_{10}$  concentrations and temperature (-0.40), PBLH (-0.39), and precipitation were statistically strong (-0.21). The estimated Pearson correlation coefficients showed a statistically strong positive correlation between As & Pb, As & Cd, and Cd & Pb during 2013–2019, indicating a similar origin. PTEs with hazard quotient sbelow one, regardless of subpopulation type, posed no noncancerous risk to Europeans. The hazard quotient values positively correlated with time, possibly due to elevated PTE levels. In our study on carcinogen pollution in Europe between 2013 and 2019, we found unacceptable levels of As, Cd, Ni, and Pb among adults, children, and infants. Carcinogenic risk rates were highest for children, followed by infants, adult women, and adult men. Therefore, besides monitoring and mitigating PM concentrations, effective control of PM sources is also needed.

 $^{\star}\,$  This paper has been recommended for acceptance by Pavlos Kassomenos.

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#### https://doi.org/10.1016/j.envpol.2023.121232

Received 24 November 2022; Received in revised form 18 January 2023; Accepted 5 February 2023 Available online 10 February 2023 0269-7491/© 2023 Elsevier Ltd. All rights reserved.

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#### 1. Introduction

Air quality is affected by natural and anthropogenic activities. Atmospheric particulate matter is a health-threatening air pollutant (Almeida et al., 2020; WHO, 2005, 2021).

Globally, air quality has become the top environmental issue in recent decades. World Health Organization's acute  $PM_{10}$  exposure guideline is 45  $\mu g/m^3$ , and the chronic  $PM_{10}$  exposure guideline is 15  $\mu g/m^3$  (WHO, 2005, 2021). The current European Legislation sets the limit for  $PM_{10}$  values for human health protection at 50  $\mu g/m^3$  (daily average) and 40  $\mu g/m^3$  (annual average). To ensure health, short- and long-term values shouldn't be exceeded (WHO, 2021). Nevertheless, European cities have poor air quality due to high  $PM_{10}$  levels (EEA, 2016).

WHO reported three million premature deaths from ambient PM worldwide in 2016 (Lim et al., 2012). Epidemiological studies show that PM exposure can increase hospitalisation, outpatient visits, lung cancer, and respiratory and cardiovascular disease mortality (Kim et al., 2015; Lyu et al., 2017; Samoli et al., 2008; Mahmoodirad and Niroomand, 2020). A recently conducted systematic review reported a meta-analytic effect estimate of RR = 1.04 (95% CI:1.03–1.06) between PM<sub>10</sub> and all non-accidental mortalities per 10  $\mu$ g/m<sup>3</sup>, assuming a linear relationship (Chen & Hoek, 2020; WHO, 2021). According to GRADE (Grading of Recommendations Assessment, Development and Evaluation), the certainty of evidence was considered high. More specifically, Chen & Hoek's 2020 meta-analysis found that estimated Risk Ratios (RRs) of  $PM_{10}$  exposure (per 10 µg/m<sup>3</sup>) were 1.12 (95% CI:1.06–1.19) for respiratory, 1.06 (95% CI:1.01-1.10) for ischemic heart disease, and 1.08 (95% CI:1.04–1.13) for lung cancer mortality. The estimated RRs were also above 1 for COPD, stroke, and circulatory mortality (Chen & Hoek, 2020; WHO, 2021).

 $PM_{10}$  contains primary and secondary constituents in European cities (Holst et al., 2008; Pommier, 2021; Pommier et al., 2020). The primary compounds are elemental carbon, organic matter, sea salt, etc. In contrast, secondary compounds are generally formed in the atmosphere through the chemical reactions (Pommier, 2021; Pommier et al., 2020; Lau et al., 2020).  $PM_{10}$  particles can also contain PTEs from natural sources like the earth's crust (e.g., Fe, Ca, Ba, or Mn) or anthropogenic and natural sources (e.g., As, Pb, Cd, Zn, Ni, Cu, Cr or Hg). Season, climate, geography, and combustion sources affect PM concentration and composition (Panda et al., 2021; Valavanidis et al., 2008; Shafiee et al., 2019; Mahmoodirad et al., 2019).

Besides particle size distribution, numerous studies have investigated the association between particle chemical composition and human health risks, including carcinogenic and non-carcinogenic health risks (Almeida et al., 2020; Bello et al., 2017; Chalvatzaki et al., 2019; Curtis et al., 2006; Das et al., 2020; Lyu et al., 2017; Pinto et al., 2015; Romanazzi et al., 2014; Samek, 2016; Singh & Gupta, 2016; Guevara, 2016; WHO, 2005; 2021; Bello et al., 2017; López et al., 2017; Holst et al., 2008). A health risk assessment is necessary to estimate the adverse impacts of PM on human health in different environments to map the potential risk associated with everyday life exposure to atmospheric PM (Chalvatzaki et al., 2019; Acosta et al., 2011; Duan et al., 2014; Lee et al., 2013; Tan et al., 2017; Tong et al., 2020; Zhi et al., 2021; Niroomand et al., 2020a). The health risk assessment can help residents avoid certain activities to protect their health. Inhalable particle health risk assessments are not yet common.

To answer frequently asked questions related to the level of health risk associated with inhaling polluted air, the purpose of this novel study was to estimate the potential health risks to residents exposed to  $PM_{10}$  containing PTEs (As, Cd, Ni, and Pb), based on data accessibility and availability, across Europe between 2013 and 2019. In this study,  $PM_{10}$  particles were chosen over  $PM_{2.5}$  because (1)  $PM_{10}$  is a health-harming pollutant. The new WHO Air Quality Guidelines published in 2021 provide recommendations for six pollutants, one of which is  $PM_{10}$ , which significantly impacts health independent of  $PM_{2.5}$  (WHO, 2021).

(2) Although  $PM_{2.5}$  penetrates the lungs more deeply than  $PM_{10}$ , the impact of particulate matter exposure on human health depends on their chemical composition and not only their size. (3) We should also stress that  $PM_{10}$  particles correspond to particulate matter with an aero-dynamic diameter smaller than 10 m;  $PM_{2.5}$  particles are also included.

# 2. Materials and methods

#### 2.1. Study area

One hundred and fifty-eight cities were selected from Austria, Belgium, Croatia, Cyprus, Czech Republic, Denmark, Estonia, France, Germany, Gibraltar, Ireland, Italy, Latvia, Lithuania, Poland, Romania, Slovenia, Spain, Switzerland, and the UK (Fig. 1 & Table S1). These cities are selected based on a) data accessibility and availability and b) representatives of large city characteristics in the chosen country.

#### 2.2. Air quality & meteorological data

Air quality data, including daily mean concentrations of PM<sub>10</sub>. As (in  $PM_{10}$ ), Cd (in  $PM_{10}$ ), Ni (in  $PM_{10}$ ), and Pb (in  $PM_{10}$ ), as secondary data were obtained from the European environmental Agency database (https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm) for the studied cities (Fig. 1) in 2013-2019. In the current study, all the stations represent the urban background, and rural sites are not included in our study due to the lack of data. Regarding the hourly air quality data, only valid data for 20 h a day were averaged, representing the daily PM<sub>10</sub> concentration. In cases with more than one air quality station, we decided to choose the one which was most representative of the city's general urban air quality, with data coverage of at least 75%. It was also checked that missing values were not concentrated on any specific year or season. We did not consider the difference between the instruments and methods used for PM<sub>10</sub> nor the measuring of heavy metals across the study domain. It was assumed that the organisations measuring the data followed appropriate calibration methods (Morawska et al., 2021).

The meteorological data, including the daily mean, wind speed, wind direction, precipitation, temperature, mean sea-level pressure (MSLP), and planetary boundary layer height (PBLH), were obtained from ERA5 (ECMWF Reanalysis v5) reanalysis daily based data. ERA5 reanalysis data is produced by Copernicus Climate Change Service (C3S) at European Centre for Medium-Range Weather Forecasts (ECMWF).

All the spatial representation and interpolation techniques were carried out through the QGIS (Version 3.22.3) software (QGIS, 2022;



Fig. 1. The investigated domain in the current study between 2013 and 2019.

Niroomand et al., 2020a,b). The IDW (Inverse Distance Weighting) interpolation algorithm was applied with a spatial resolution of  $0.1^{\circ}$  by  $0.1^{\circ}$  for the output raster.

#### 2.3. p.m.<sub>10</sub> time series analysis

To analyse the significance of trends in long-term changes in PM<sub>10</sub> data, Mann–Kendall test ("(wq)" package in R) was applied, using the "seaKen" functions, for the seasonal Mann–Kendall trend tests (Kendall, 1949; Mann, 1945). The Sen's Slope estimator was also applied, from the same package (wq), to quantify the magnitude of changes in the PM10 concentrations (Silva Junior et al., 2018; Mahmoodirad and Sanei, 2016; Mirzaei et al., 2019; Niroomand et al., 2020b).

#### 2.4. Feature selection based on Random Forest analysis

The feature importance (FI) analysis was carried out via Random Forest in R to investigate the meteorological parameters impacting  $PM_{10}$  levels across Europe (Molla-Alizadeh-Zavardehi et al., 2014; Shafiee et al., 2021; Jamshidi et al., 2021). FI analysis was applied for meteorological parameters (temperature, mean sea-level pressure, planetary boundary layer height, wind speed, wind direction, and precipitation).

# 2.5. Health risk assessment

To investigate whether the health risk existed in the selected European cities, potential cancerous and non-cancerous risk rates using mean values of the contaminant contents were estimated below. Three exposure routes are specified by USEPA (United Environmental Protection Agency) in the risk assessment: inhalation, ingestion, and dermal contact (US EEA, 2019). Since inhalation is the most rapid exposure pathway, it was investigated in our study (Lim et al., 2012; US EEA, 2019; Holst et al., 2008). Therefore, US EPA (2007) methodology was deployed to evaluate the health risk via inhalation pathway due to being exposed to  $PM_{10}$  containing selected potentially toxic elements (US EPA, 2007). Both risk rates were calculated for As, Cd, Ni, and Pb.

# 2.5.1. Estimated daily intake

The daily intake was determined for four subpopulations of adult men & adult women (>7 years old), children (1 < 7 years old), and infants (0 < 1 year old) using Equation (1) as below (ATSDR, 2005):

$$EDI = (C \times IR \times AF \times F \times ED) / (BW \times AT)$$
<sup>(1)</sup>

 $AT = ED \times 365$ 

where *EDI*, *C*, *IR*, *AF*, *F*, *ED*, *BW*, and *AT* stand for estimated daily intake (mg/kg body weight per day), contaminant concentration (mg/m<sup>3</sup>), intake rate (m<sup>3</sup>/day), bioavailability factor (unitless), frequency of exposure (days/year), exposure duration (years), body weight (kg), and averaging time (days), respectively. Table S2 presents the variable used to estimate daily intake by different subpopulations.

#### 2.5.2. Potential carcinogenic risk of outdoor air inhalation

The probability of cancer development in European inhabitants was estimated based on the reported methods in the literature (Gruszecka-Kosowska, 2018; Chalvatzaki et al., 2019; Panda et al., 2021; Yousefi et al., 2021; Pachoulis et al., 2022). The dose-response correlation in the quantitative cancerous risk assessment is expressed in the potency slope, which calculates the probability of cancerous risk associated with an estimated exposure. The cancerous Risk (*R*) value is acceptable below 1.00E-06-1.00E-04 (Chalvatzaki et al., 2019; Megido et al., 2017), whereas the most tolerable risk rate is 1.00E-06.

The health risk vales of carcinogenic compounds were calculated according to equation (2) as below (US EPA, 2007):

$$R = EDI \times SF \tag{2}$$

where *R*, *EDI*, and *SF* stand for cancer risk (unitless), estimated daily intake (mg/kg body weight per day), inhalation slope factor [(mg/kg body weight per day)<sup>-1</sup>], respectively. Table S3 presents the slope factor (*SF*) values in estimating carcinogenic risk for different substances (CalEPA, 2016; Garbero et al., 2011; Taiwo et al., 2017; US EPA, 2016; Yang et al., 2014).

#### 2.5.3. Potential non-carcinogenic risk of outdoor air inhalation

Noncarcinogenic risk means all adverse impacts on human health caused by exposure factors, excluding cancer. The allowable non-cancer risk is below 1, and values above 1 are unacceptable risks and must take necessary corrective actions to reduce the risk levels. The unacceptable values greater than 1 mean a higher probability of developing noncancerous impacts on human beings (US EPA, 1989).

The health risk values of non-carcinogenic compounds were calculated according to equation (3) as below (US EPA, 2007):

$$HQ = EDI/RfD \tag{3}$$

where *HQ*, *EDI*, and *RfD* stand for hazard quotient (unitless), estimated daily intake (mg/kg body weight per day), and reference dose (mg/kg body weight per day), respectively.

Table S3 presents the reference dose (*RfD*) values in estimating noncarcinogenic risk for different substances (CalEPA, 2016; Garbero et al., 2011; Taiwo et al., 2017; US EPA, 2016; Yang et al., 2014).

#### 2.5.4. Combined carcinogenic and non-carcinogenic risk rates

Equation (4) was used to estimate the total carcinogenic risk ( $R_t$ ) of the inhalation of many substances at the same time (US EPA, 2007):

$$R_t = R_1 + R_2 + \dots + R_n \tag{4}$$

Also, Equation (5) was used to estimate the total non-carcinogenic risk (*HI*) of the inhalation of many substances at the same time (US EPA, 2007):

$$HI = HQ_1 + HQ_2 + \dots + HQ_n \tag{5}$$

where 1-n: specified the number of air pollutants.

#### 3. Results

#### 3.1. Spatial and temporal variation of $PM_{10}$

Across Europe, the maximum permissible daily  $PM_{10}$  concentration recommended by WHO (45 µg/m<sup>3</sup>) was exceeded between 2013 and 2019. The mean annual values are above the WHO recommended annual value of 15 µg/m<sup>3</sup> in a vast part of Europe during the studied period. Table 1 shows the highest number of  $PM_{10}$  events and daily  $PM_{10}$ concentrations, exceeding WHO guidelines in European cities between 2013 and 2019.

On the continental scale, there was a gradual decline in the total number of exceedances ranging from 6446 days in 2013–3721 days in 2019, which could be attributed to the increasing temperature during the year's cold period and heating reduction across Europe (Megaritis et al., 2013).

Figure S1 shows the monthly variations in the total number of  $PM_{10}$  events exceeding WHO recommended daily level in the study area from 2013 to 2019. Generally, the total monthly number of exceedances of the WHO limit was reduced between 2013 and 2019 (Figure S1). Table 2 shows the monthly analysis of the highest number of  $PM_{10}$  events and daily  $PM_{10}$  concentrations, exceeding WHO guidelines in European cities between 2013 and 2019. The provided information indicated that the highest measured levels were in the cold period of the year (Table 2) in our domain of study. The frequency of exceedance of the WHO threshold differs from its intensity in European cities. The longest events of high  $PM_{10}$  concentrations are mainly observed during wintertime

#### Table 1

The highest number of  $PM_{10}$  events and daily  $PM_{10}$  concentrations, exceeding WHO guidelines in European cities between 2013 and 2019.

|      | The highest n events. | umber of PM <sub>10</sub> | The highest daily PM <sub>10</sub> concentration. |  |  |  |  |  |  |  |
|------|-----------------------|---------------------------|---|--|--|--|--|--|--|--|
| Year | City                  | Number of<br>days         | City  | Daily $PM_{10}$ concentration (µg/m <sup>3</sup> ) |  |  |  |  |  |  |
| 2013 | Paris                 | 209                       | Venezia   | 313  |  |  |  |  |  |  |
| 2014 | Paris                 | 145                       | Nicosia   | 259  |  |  |  |  |  |  |
| 2015 | NowySacz              | 125                       | Nicosia   | 1137   |  |  |  |  |  |  |
| 2016 | Gornoslaski           | 128                       | Sevilla   | 335  |  |  |  |  |  |  |
| 2017 | Gornoslaski           | 124                       | Gornoslaski                                       | 370  |  |  |  |  |  |  |
| 2018 | Gornoslaski           | 128                       | Nicosia   | 325  |  |  |  |  |  |  |
| 2019 | Rybnik                | 106                       | Rybnik  | 272  |  |  |  |  |  |  |

(Table 2). The lower air temperature in the cold period, especially in December, January, February, and March, can be responsible for the highest number of exceedances in this year's period. On the other hand, only sporadic exceedances of  $PM_{10}$  daily limits are reported in summertime (Figure S1). For example, in September 2015, a recordbreaking dust storm that originated from dust sources located in Northern Syria and Iraq could increase the  $PM_{10}$  daily concentration above 1000 µg/m<sup>3</sup> (1137 µg/m<sup>3</sup> on September 8, 2015) (Mamouri et al., 2016).

In our work, a long-term analysis of  $PM_{10}$  concentrations showed a statistically significant reduction in most of the studied cities between 2013 and 2019, which could be attributed to the successful implemented clean air policies by the European Union. Fig. 2 shows the magnitude of changes via Sen's slope estimator in some selected cities. But a few cities, mainly in Spain and Poland, showed a statistically notable increase in  $PM_{10}$  concentration in the study period (Figure S2). Among the studied cities, Riga (Latvia), Gibraltar (Gibraltar), and Lodz (Poland), with Sen's slopes of -2.04, -1.78, and -1.75, respectively, showed the highest magnitude of reduction between 2013 and 2019. While Elblag (Poland), Poznan (Poland), and Malaga (Spain), with Sen's slopes of 1.15, 0.91, and 0.87, respectively, had the highest magnitude of increase in  $PM_{10}$  concentration during the study period (Figure S2).

#### 3.1.1. The impact of meteorological parameters on $PM_{10}$ concentrations

In our study, to work out the dependence of  $PM_{10}$  concentrations on meteorological parameters, the Pearson correlation coefficient and feature importance via RF analysis was determined over the period from 2013 to 2019 (Figure S3). The study domain was divided into Western, Eastern, Northern, and Southern Europe due to the variations in weather conditions across Europe (Fig. 2). Fig. 4 illustrates the meteorological parameters over Europe between 2013 and 2019..

According to the RF analysis, the main influencing factor(s) in Northern, Southern, Western, and Eastern Europe was precipitation (0.20), temperature (0.30), temperature (0.23), and temperature (0.34), respectively (Figure S3).

On the continental scale, the estimated Pearson correlation coefficients showed a statistically strong negative correlation (p-value below 0.05) between  $PM_{10}$  concentrations and temperature (-0.40), PBLH (-0.39), and precipitation (-0.21). Wind speed had a statistically negative significant correlation (p-value below 0.05) with  $PM_{10}$  concentrations as well, but not strong.  $PM_{10}$  concentrations had statistically positive correlation (p-value below 0.05) with MSLP (+0.20) and wind direction (+0.13). Regionally,  $PM_{10}$  concentrations showed higher vulnerability to temperature, MSLP, PBLH, wind direction, wind speed, and precipitation in Eastern (-0.63), Northern (+0.30), Eastern (-0.55), Northern (+0.30), Southern (-0.25), and Northern Europe (-0.34), with p-values below 0.05, compared to other parts, respectively.

# 3.2. Spatial and temporal variation of potentially toxic elements

Table 3 shows the maximum and minimum annual concentrations of PTEs in  $PM_{10}$  across Europe between 2013 and 2019. According to the results, the cities of Antwerpen (39.7 ng/m<sup>3</sup>), Kladno (6.7 ng/m<sup>3</sup>), Gibraltar (16.9 ng/m<sup>3</sup>), Antwerpen (618.7 ng/m<sup>3</sup>) had the highest annual values of As, Cd, Ni, and Pb, respectively from 2013 to 2019 (Table 3)..

Fig. 3 shows the spatial variation of the correlation between time and HQ values for As, Cd, Ni, and Pb in  $PM_{10}$ , respectively within the study area between 2013 and 2019.

Generally, a negative correlation between PTEs (As, Cd, Ni, and Pb) and time was observed. Results showed a positive correlation in Ni content within vast study cities (50 cities). As (in  $PM_{10}$ ), Cd (in  $PM_{10}$ ), Ni (in  $PM_{10}$ ), and Pb (in  $PM_{10}$ ) showed a positive correlation with time in some studied cities, mainly located in Germany (9 cities), Germany (3 cities), Poland (19 cities), and Germany & Spain (4 cities), respectively.

The strongest positive correlation between time and As (in  $PM_{10}$ ), Cd (in  $PM_{10}$ ), Ni (in  $PM_{10}$ ), and Pb (in  $PM_{10}$ ) was in Rouen (France, +0.98), Rouen (France, +0.90), Piotrkow Trubinalski (Poland, +0.88), and Alicante (Spain, +0.91), respectively. While cities of Antwerpen (Belgium, -0.96), Antwerpen (Belgium, -1.0), Castellon de la Plana (Spain, -0.98), and Saint Etienne (France, -0.97) showed the highest negative correlation with time regarding As (in  $PM_{10}$ ), Cd (in  $PM_{10}$ ), Ni (in  $PM_{10}$ ), and Pb (in  $PM_{10}$ ), respectively. Rouen (France), Berlin (Germany), and Belfast (UK) showed a positive correlation with time, meaning an increasing trend in all PTEs but with different values. There are also a few cities with correlation values near zero, showing neither positive nor negative correlation with time, which means there was not changes in the content of the studied elements (Fig. 3).

On the continental scale, the estimated Pearson correlation coefficients showed a statistically strong positive correlation between As & Pb (in  $PM_{10}$ ), As & Cd (in  $PM_{10}$ ), and Cd & Pb (in  $PM_{10}$ ) with values of 0.68, 0.52, and 0.48, respectively during 2013–2019, indicating the similarity between their sources. Regionally, the highest correlation between the aforementioned PTEs was observed in Western Europe, with values above 0.90.

# 3.3. Health risk assessment

# 3.3.1. Estimates of non-cancer risks

The non-cancer risk estimates (i.e., *HQ* value) were calculated based on the calculated intake rates and considering the reference values (*RfD*)

Table 2

The monthly analysis of the highest number of PM<sub>10</sub> events and daily PM<sub>10</sub> concentrations, exceeding WHO guidelines in European cities between 2013 and 2019.

|      | The highest numb | er of $PM_{10}$ events. |                | The highest daily PM <sub>10</sub> concentration. |           |  |  |  |  |  |
|------|------------------|-------------------------|----------------|---|-----------|--|--|--|--|--|
| Year | City             | Month                   | Number of days | City  | Month     | Daily $PM_{10}$ concentration (µg/m <sup>3</sup> ) |  |  |  |  |
| 2013 | Paris            | March                   | 28             | Venezia   | January   | 313  |  |  |  |  |
| 2014 | Nicosia          | December                | 26             | Nicosia   | March     | 259  |  |  |  |  |
| 2015 | Terni            | December                | 30             | Nicosia   | September | 1137   |  |  |  |  |
| 2016 | Vicenza          | December                | 30             | Sevilla   | February  | 335  |  |  |  |  |
| 2017 | Gornoslaski      | January                 | 27             | Gornoslaski                                       | January   | 370  |  |  |  |  |
| 2018 | Gornoslaski      | February                | 28             | Nicosia   | March     | 325  |  |  |  |  |
| 2019 | Verona           | January                 | 22             | Rybnik  | December  | 272  |  |  |  |  |



Fig. 2. The governing meteorological parameters including (a) wind profile & MSLP (Pa), (b) Temperature (oC), (c) PBLH (m), and Precipitation (mm/day) over Europe between 2013 and 2019.

Table 3 The maximum and minimum annual concentrations of potentially toxic elements in PM<sub>10</sub> across Europe between 2013 and 2019.

| Year | Max                     | Min            | Max                     | Min Max         |                         | Min                    | Max                     | Min            |  |
|------|-------------------------|----------------|-------------------------|-----------------|-------------------------|------------------------|-------------------------|----------------|--|
|      | As (ng/m <sup>3</sup> ) |                | Cd (ng/m <sup>3</sup> ) |                 | Ni (ng/m <sup>3</sup> ) |                        | Pb (ng/m <sup>3</sup> ) |                |  |
| 2013 | Glogow (16.0)           | Nicosia (0.2)  | Kladno (6.7)            | Alicante (0.1)  | Gibraltar (16.0)        | Alicante (0.1)         | Terni (73.1)            | Palma (1.1)    |  |
| 2014 | Antwerpen (35.2)        | Badajoz (0.1)  | Antwerpen (7.0)         | Alicante (0.1)  | Gibraltar (16.9)        | Kaunas (0.5)           | Antwerpen (392.7)       | Caceres (1.3)  |  |
| 2015 | Antwerpen (39.7)        | Badajoz (0.1)  | Kladno (3.0)            | Toledo (0.04)   | Charleroi (15.7)        | Ceske Budejovice (0.3) | Antwerpen (618.7)       | Caceres (1.5)  |  |
| 2016 | Antwerpen (31.2)        | Klaipeda (0.1) | Kladno (5.0)            | Toledo (0.01)   | Hamburg (14.2)          | Toledo (0.2)           | Antwerpen (490.4)       | Toledo (0.4)   |  |
| 2017 | Glogow (30.2)           | Klaipeda (0.1) | Antwerpen (4.2)         | Alicante (0.01) | Gibraltar (13.9)        | Klaipeda (0.2)         | Antwerpen (332.5)       | Klaipeda (0.6) |  |
| 2018 | Antwerpen (11.6)        | Zagreb (0.1)   | Kladno (3.9)            | Caceres (0.03)  | Gibraltar (12.9)        | Toledo (0.3)           | Antwerpen (175.7)       | Caceres (1.0)  |  |
| 2019 | Antwerpen (16.2)        | Klaipeda (0.1) | Antwerpen (3.8)         | Caceres (0.03)  | Gibraltar (13.0)        | Kaunas (0.4)           | Antwerpen (221.1)       | Madrid (0.3)   |  |

of doses. For the PTEs, hazard quotient values, regardless of the subpopulation type, were estimated to be smaller than 1, indicating no risk to adult men, women, children, and infants across Europe in the study period (Table S4 and S7).

The highest mean annual *HQ* values of exposures of PM-associated As to adult men, women, children, and infants were observed in Antwerpen, with values of 0.034, 0.040, 0.083, and 0.060, respectively, in 2015. HQ - Cd, HQ - Ni, and HQ - Pb had their highest values in Antwerpen (2014), Gibraltar (2014), and Antwerpen (2015), respectively, regardless the subpopulation type (Table S5). The total hazard values (*HI*) of exposures of PM-associated metals followed the same pattern for studied subpopulations during 2013–2019 (Figures S5-S8). Table S4 shows that the order of *HQ* values of exposures of PM-associated PTEs were Children, infants, adult women, and adult men, which could be attributed to the differences between estimated daily intake values of subpopulation types. Our estimated *HQ* values were close to the ones reported in similar studies, for example HQ - As = 0.063 and 0.0234 in northern Spain (cities of Asturias & Gijon) and Lisbon (Chalvatzaki

# et al., 2019; Megido et al., 2017).

Fig. 4 and S9-S11 show a general negative correlation with time in the HQ values irrespective to the subpopulation, but being exposed to PM-associated PTEs in the cities with a positive correlation of PTEs with time indicated an increasing non-carcinogenic risk following the order of HQ-Ni > HQ-As > HQ-Cd > HQ-Pb between 2013 and 2019. It means that the amount of evaluated PTEs can increase the non-cancer risks by increasing HQ values. Regarding HQ-Ni, many cities in Eastern Europe and some in Western and Southern Europe are more vulnerable than others, indicating growing concern from 2013 to 2019. In contrast, HQ-As showed more vulnerability in the Western and Southern European studied cities, while HQ-Pb among 4 studied non-cancer risks showed less concern during the study time frame.

The strongest positive correlation between time and *HQ-As, HQ-Cd, HQ-Ni,* and *HQ-Pb* was in Rouen (France, >0.95), Rouen (France, >0.90), Piotrkow Trubinalski (Poland, +0.85), and Alicante (Spain, >0.90), respectively. While cities of Antwerpen (Belgium, <-0.95), Antwerpen (Belgium, <-0.95), Castellon de la Plana (Spain, <-0.95),

# Table 4

The percent of European inhabitants at cancer risk from exposure to PM<sub>10</sub>-bound potentially toxic elements via inhalation from 2013 to 2019.

| Year        | C1 <sup>a</sup>  | C2 <sup>a</sup> | C3 <sup>a</sup>   | C4 <sup>a</sup> | C1 <sup>a</sup> | C2 <sup>a</sup> | C3 <sup>a</sup> | C4 <sup>a</sup> | C1 <sup>a</sup> | C2 <sup>a</sup> | C3 <sup>a</sup> | C4 <sup>a</sup> | C1 <sup>a</sup> | C2 <sup>a</sup> | C3 <sup>a</sup> | C4 <sup>a</sup> | C1 <sup>a</sup> | C2 <sup>a</sup> | C3 <sup>a</sup> | C4 <sup>a</sup> |
|-------------|------------------|-----------------|-------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|             | R-As             | ;               | $\overline{R-Cd}$ |                 |                 |                 |                 | R - Ni          |                 |                 |                 | R - Pb          |                 |                 |                 | R-Total         |                 |                 |                 |                 |
| Adult M     | en               |                 |                   |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| 2013        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2014        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2015        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2016        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2017        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2018        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| 2019        | 0                | 4               | 86                | 4               | 0               | 1               | 39              | 1               | 0               | 0               | 10              | 0               | 0               | 0               | 1               | 0               | 0               | 9               | 98              | 9               |
| Adult Women |                  |                 |                   |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| 2013        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2014        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2015        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2016        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2017        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2018        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| 2019        | 0                | 5               | 91                | 5               | 0               | 1               | 46              | 1               | 0               | 0               | 14              | 0               | 0               | 0               | 1               | 0               | 0               | 12              | 99              | 12              |
| Childrer    | <b>n</b> (1–7 ye | ars old)        |                   |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| 2013        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2014        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2015        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2016        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2017        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2018        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| 2019        | 1                | 17              | 99                | 18              | 0               | 5               | 80              | 5               | 0               | 0               | 46              | 0               | 0               | 0               | 3               | 0               | 1               | 39              | 100             | 40              |
| Infants     | (0–1 yea         | r old)          |                   |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| 2013        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2014        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2015        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2016        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2017        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2018        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |
| 2019        | 0                | 10              | 97                | 10              | 0               | 3               | 64              | 3               | 0               | 0               | 28              | 0               | 0               | 0               | 1               | 0               | 0               | 25              | 100             | 26              |

 $^{a}$  C1 = R  $\geq$  10E-4; C2 = 10E-5  $\leq$  R < 10E-4; C3= > 10E-6; C4= > 10E-5.



Fig. 3. The distribution of correlation strength between PTEs' concentrations (As, Cd, Ni, and Pb in PM<sub>10</sub>, respectively) and time across Europe between 2013 and 2019. Note: The legend equals to Pearson's coefficient of correlation value.



Fig. 4. The distribution of correlation strength between HQ-As, HQ-Cd, HQ-Ni, and HQ-Pb respective to adult men and time across Europe between 2013 and 2019. Note: The legend equals to Pearson's coefficient of correlation value.



Fig. 5. The distribution of correlation strength between *R*–*As*, *R*–*Cd*, *R*–*Ni*, and *R*–*Pb* respective to adult men and time across Europe between 2013 and 2019. Note: The legend equals to Pearson's coefficient of correlation value.

and Saint Etienne (France, <-0.95) had the highest negative correlation with time regarding *HQ-As, HQ-Cd, HQ-Ni* and *HQ-Pb*, respectively. Berlin (Germany) and Rouen (France) were the only cities showing a positive correlation in all *HQ* values with time, meaning an increasing trend in all non-cancer risks but with different values. In terms of Antwerpen (Belgium), according to Table S5, even though it has the highest annual values of PTEs but shows a negative correlation with time in *HQ* values, which means decreasing during the study time.

#### 3.3.2. Estimates of cancer risks

Carcinogenic risk rates (*R*) were calculated using estimated intake rates and the Slope Factor (*SF*) values. This study assumes an acceptable risk level to be one additional cancer case occurrence in the population of one million people (1.00E-06). The unacceptable risk levels were found in our study on carcinogenic pollution in the case of As (in PM<sub>10</sub>), Cd (in PM<sub>10</sub>), Ni (in PM<sub>10</sub>), and Pb (in PM<sub>10</sub>) in the vast part of Europe among adults (men & women), children, and infants between 2013 and 2019. On the continental scale, Table 2 shows the percentage of atcancer-risk people in respective studied subpopulations from 2013 to 2019.

The following order of *R* values of exposure of PM-associated PTEs, irrespective of the subpopulation type, to inhabitants, was observed: R-As > R-Cd > R-Ni > R-Pb (from highest to smallest *R*-value). Carcinogenic risk rates were higher among children, followed by infants, adult women, and adult men, which could be attributed to the differences between estimated daily intake values of subpopulation types (Table 4 and S6). Results showed that above 80% of European inhabitants were at carcinogenic risk related to As (R–As>10E-6), with elevated *R* values, above 95% in respective to children and infants. After As, Cd had higher carcinogenic risk rates among people, especially children and infants. Compared to As and Cd, Pb showed less vulnerability to carcinogenic risk rates. Table S6 shows only the three highest calculated *R* values in each year respective to studied subpopulations.

The total carcinogenic risk was highest in Antwerpen (Belgium, 2014) with the vales of 1.4E-04, 1.7E-04, 3.4E-04, 2.5E-04 for adult men, women, children, and infants, respectively (Figures S5-S8).

Regardless of the subpopulation type, carcinogenic risk rates followed a negative correlation with time (Fig. 5 and S12-S14). While some cities with a positive correlation indicated an increasing carcinogenic risk following the order of R–Ni > R–As > R–Cd > R–Pb between 2013 and 2019. It means that the evaluated amounts of PTEs over studied time (Fig. 3) can increase the cancer risk rates among inhabitants, regardless of a subpopulation, by increasing R values across Europe, indicating the necessity of taking practical controlling actions of PM sources along with controlling and mitigation of PM concentrations. Again, a vast number of cities in Eastern Europe, along with some in Western and Southern Europe, showed higher vulnerability to the carcinogenic risk rate of Ni, indicating growing concern from 2013 to 2019. Among four studied carcinogenic risk rates, R–Pb showed less concern during the study time frame (Fig. 5 and S12-S14).

The strongest positive correlation between time and R - As, R - Cd, R - Ni, and R - Pb was in Rouen (France, >0.95), Rouen (France, >0.90), Piotrkow Trubinalski (Poland, >0.85), and Alicante (Spain, >0.90), respectively. On the other hand, Antwerpen (Belgium, <-0.95), Antwerpen (Belgium, <-0.95), Castellon de la Plana (Spain, <-0.95), and Saint Etienne (France, <-0.95) showed the highest negative correlation with time regarding R - As, R - Cd, R - Ni, and R - Pb, respectively. Berlin (Germany) and Rouen (France), by positive correlations in all R values with time, showed increasing vulnerability to cancer risk among different subpopulations with different values.

# 4. Discussion

# 4.1. Spatial-temporal variation of PM<sub>10</sub> concentration

Despite introduced PM mitigation policies in Europe, similar studies

reported severe PM episodes across Europe, including Cracow and Warsaw (January 2006), Athens (December 2012), London (February 2014), and Paris (March 2014) (Reizer & Juda-Rezler, 2016). Polish cities recorded 77 p.m.<sub>10</sub> episodes between 2005 and 2012. In some PM<sub>10</sub> events, the daily EU air threshold was exceeded sevenfold in January 2009 and tenfold in January 2010. In January 2010, Jelenia Gora had the highest  $PM_{10}$  level, 480 g/m<sup>3</sup> (Reizer & Juda-Rezler, 2016).

A study investigating the  $PM_{10}$  levels in 17 European cities showed that central and east-central Europe had the highest  $PM_{10}$  levels in 2010–2014. Poland, Bulgaria, and Slovakia had the highest  $PM_{10}$  concentrations, while Finland had the lowest (Chlebowska-Styś et al., 2017). In early October 2020, a  $PM_{10}$  event was reported in Northern Europe. Several Norwegian air quality stations recorded daily mean  $PM_{10}$  values up to 97 g/m<sup>3</sup>, which exceeded reported mean values from the past four to 10 years (Groot Zwaaftink et al., 2021). The observed  $PM_{10}$  exceedance events are usually accompanied by a stable high-pressure atmospheric SLP (sea-level pressure) pattern, which favours thermal inversion and lowers the planetary boundary layer height (Czernecki et al., 2017; Holst et al., 2008; Nidzgorska-Lencewicz & Czarnecka, 2015).

A recent study examined Europe's  $PM_{10}$  temporal trend to determine the effectiveness of clean air policies (Beloconi & Vounatsou, 2021). In this study,  $PM_{10}$  concentration decreased by 36.5% (30.3%, 41.9%) between 2006 and 2019 (Beloconi & Vounatsou, 2021). In another study,  $PM_{10}$  emissions decreased by 1.7% per year between 2000 and 2017 in the EU-28 (Sicard et al., 2021). Despite a reduction in  $PM_{10}$ emissions in the EU during the study period, the percentage of EU-28 residents exposed to  $PM_{10}$  concentrations (40 g/m<sup>3</sup>) ranged from 18 to 44% in 2000–2010 to 13 to 30% in 2010–2017. EU threshold values were repeatedly violated, mainly in Eastern Europe (Guerreiro et al., 2014; Sicard et al., 2021). In urban EU-28 stations, annual average  $PM_{10}$ concentrations fell by 0.65 g/m<sup>3</sup> (EEA, 2019).

Wind erosion, mining and construction, agricultural land management, and traffic resuspension are Europe's leading sources of coarse PM emissions (Guevara, 2016). PM emissions are decreasing across Europe thanks to EU legislation focused on large point and road transport sources. The reduction was due to energy production and distribution improvements, gas abatement techniques, vehicle technologies related to "Euro" standards, and solvent storage and distribution (EEA, 2014; Sicard, Paoletti, et al., 2020; Vestreng et al., 2009).

But, the lack of regulations increases household, commercial, and institutional emissions (Gozzi et al., 2017; Guevara, 2016). These sectors contributed 43% of the total EU-28 p.m.<sub>10</sub> emissions in 2013. (EEA, 2019; Guevara, 2016). Another alarming issue is long-range transboundary air pollution caused by atmospheric circulation indicating that each country can be a producer and receiver (EEA, 2015; Gozzi et al., 2017).

# 4.2. Weather conditions and changes in PM<sub>10</sub> concentration

Air pollution levels vary depending on pollution source characteristics or discharged emissions, weather conditions, and the study area's physical geography (Czernecki et al., 2017; Volná & Hladk, 2020). Meteorological conditions determine pollutant intensity and dispersion (Volná & Hladk, 2020). The worst dispersion conditions for PM<sub>10</sub> air pollution occur at high atmospheric SLP (positive correlation), low precipitation (negative correlation), low wind speed (negative correlation), low air temperatures (negative correlation), and low PBLH (negative correlation) along with a wind direction from areas with a higher accumulation of pollution sources (Zhou et al., 2007) (Fig. 2 & S3). Wind profile affects horizontal dispersion and transport of pollutants, while air temperature and atmospheric stability affect vertical dispersion. Temperatures rise with height in stable weather, weakening vertical mixing. On the contrary, atmospheric instability promotes pollutant dispersion (Buchanan et al., 2002). So, improving the current

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knowledge about the existing correlation between meteorological conditions and elevated PM concentrations can help define health hazards' preventive measures in studied populated areas (ernikovsk et al., 2016; Shafiee et al., 2017; Volná & Hladk, 2020).

Besides, previous studies showed a positive correlation between  $PM_{10}$  and summer air temperature, e.g., higher agricultural emissions, and a negative correlation in winter, e.g., lower tertiary sector heating emissions, which means climate change reduces the benefits of PM precursor emission controls and raises PM levels (Barmpadimos et al., 2011; EEA, 2018). A recent study examined climate change's impact on Spain's air quality from 1993 to 2017. Most air pollutants changed significantly (Borge et al., 2019). Seasonal changes in  $PM_{10}$  levels up to 22 g/m<sup>3</sup> had the most significant impacts over the 25-year study period (Borge et al., 2019).

## 4.3. p.m. 10-associated potential toxic elements (PTEs)

PM<sub>10</sub> particles can also contain PTEs from natural and/or anthropogenic sources (Holst et al., 2008; Pommier, 2021; Pommier et al., 2020). The observed PTEs, in the current, are from anthropogenic sources such as public power and heat, residential combustion, industrial combustion and processes, road transport, non-road transport, and waste incineration (Pacyna et al., 2007; Schlutow et al., 2021). For As, Cd, and Ni, stationary fuel combustion is the main source, while gasoline combustion is the primary source (Pacyna et al., 2007). Pb is also found in vehicle exhaust and tyre-abrasion materials (Das et al., 2020; Sternbeck et al., 2002). EU-27 Cd and Pb emissions fell 33% and 44% between 2005 and 2019. (EEA, 2019). Manufacturing and extractive industries accounted for 57.6% and 61.8% of Cd and Pb emissions, respectively. Moreover, the energy supply sector's Cd emissions notably dropped 58.4%. Germany, Italy, and Poland contributed half of the EU's Pb, Cd, and Hg emissions in 2019 (EEA, 2019).

A significant positive correlation between Ni and As, Cr, with values of 0.46 and 0.51, respectively, was also reported (Batbold et al., 2021). Cr was mainly derived from vehicles and industrial emissions. While, As and Ni mainly come from coal combustion (Kursun Unver & Terzi, 2018). Pb permanent sources include lead-acid batteries, vehicle tyre abrasion, and urban brake wear (Batbold et al., 2021; Guttikunda et al., 2013).

Our study found a positive correlation between time and PTEs concentrations in some cities, indicating the need to control PM sources rather than overall PM concentrations. The development of monitoring devices capable of near-real-time chemical analysis can help authorities assess the health risks of air pollution in specific areas and identify anthropogenic and natural pollution sources (Godish et al., 2015). Current emission inventories must be improved to help solve present and future air quality issues. To address current and future issues, emission inventories should include species so far ignored, such as isotopes, heavy metals, and intermediate volatility organic compounds (IVOCs) that contribute to many chemical processes involving PM (Gozzi et al., 2017).

#### 4.4. p.m.10-associated potential toxic elements & human health

Urban air pollution increases mortality, non-cancerous and cancerous risks (Effatpanah et al., 2020; Leili et al., 2021; Morakinyo et al., 2017; Yousefi et al., 2022). Health risk assessments in Krakow, Poland, between 2007 and 2016 showed that non-carcinogenic risk levels were medium for inhaling  $PM_{10}$  (adults, children, and infants) (Gruszecka-Kosowska, 2018). The *HQ* values in Krakow relating to  $PM_{10}$  inhalation for men, women, children, and infants were 1.44, 1.72, 3.51, and 2.53, respectively (Gruszecka-Kosowska, 2018). On the contrary, our results showed low non-cancer risks in Krakow between 2013 and 2019, regardless of subpopulation type. This is likely due to a decreasing, but not statistically significant, trend in  $PM_{10}$  and a negative correlation between PTEs and time showing decreases over time.

For a short time, other studies in Athens, Kuopio, and Lisbon showed HQ values below 1, indicating no toxic effect. In Athens, inhaling certain heavy metals (As, Cd, Co, Cr, Mn, Ni, and Pb) posed higher risks (Chalvatzaki et al., 2019).

The estimated carcinogenic risk associated with polluted air showed that the carcinogenic risk of studied subpopulations in Krakow was not acceptable respective to As (in  $PM_{10}$ ), Cd (in  $PM_{10}$ ), Ni (in  $PM_{10}$ ), and Pb (in  $PM_{10}$ ) from 2007 to 2016. (Gruszecka-Kosowska, 2018). Children, infants, adult women, and men had total carcinogenic risks of 3.04E-04, 2.22E-04, 1.45E-04, and 1.22E-04 (Gruszecka-Kosowska, 2018). Chalvatzaki et al. (2019) found a low risk of cancer-related to Cd and Ni in Athens, Lisbon, and Kuopio, but *R*–*As* with values above 10E-6 showed a higher risk in Athens for all subpopulations (Chalvatzaki et al., 2019). It is also essential to add that Arsenic can cause bladder, lung, kidney, liver, skin, and prostate cancers (Martin et al., 2014).

According to our study and others, children and infants face greater carcinogenic and non-carcinogenic risks. Epidemiologic studies show that polluted air highly threatens these subpopulations (Leili et al., 2021; Schwenk et al., 2003). The main suggested reasons include renal clearance, greater lung volume compared to body surface area, faster ventilation rate, and metabolic immaturity (Aliff et al., 2020; Daston et al., 2004).

Elevated PTEs exposure could notably increase cancer risk and other health endpoints, especially in populated areas. Among the most common health endpoints of toxic air pollution exposure are genotoxicity, mutagenic effects, nervous diseases, incremental lifetime risk, and cancer (blood, skin, bone, and lung) (Ekpenyong and Asuquo, 2017; Guerreiro et al., 2016; Idani et al., 2020; Kumar et al., 2016).

# 4.5. Strengths, limitations and suggestions for future studies

As a strength of this research study, its relevance lies in its scope, which includes 158 European cities to provide a current overview of the impact of inhaling polluted particulate matter on European citizens, distinguishing it from other relevant scientific works with a limited scope. For this reason, solid evidence supports general conclusions. The results allow evaluation of the long-term effectiveness of European air quality policies on health, using non-carcinogenic and carcinogenic risk as metric variables. In the context of public health, this research may offer air quality managers valuable clues about the need to control PM emission sources, reducing ambient air PM concentrations and, by extension, human exposure to potentially polluted particulate matter.

Covering a vast number of cities from different countries allows us to compare the outputs of conducted  $PM_{10}$  trend analysis,  $PM_{10}$ -associated PTEs (including As, Cd, Ni, and Pb), their temporal & spatial distribution, and health risk assessment and have a better understanding of the changes over time in  $PM_{10}$  concentrations, spatio-temporal distribution of  $PM_{10}$ -associated PTEs (including As, Cd, Ni, and Pb), and consequently non-cancerous & cancerous risks affecting four subpopulations (including adult men, women, children, and infants) across Europe between 2013 and 2019. Such a provided vast amount of information is vital in developing health-protective policies based on realistic exposure scenarios.

The dissemination of results can also increase public awareness, specifically among sensitive groups, about the atmospheric pollution damaging effects (in this case, assessing cancer incidence) and its associated factors. Current knowledge on the status of European inhabitants' exposure to particulate matter comprised of toxic elements at potentially polluted levels is paramount in terms of Public Health. In this sense, authorities may take necessary actions to reduce people's exposure to toxic air pollutants and decrease cancer risk.

As a leading limitation, this study does not cover the exposure of all citizens at the European level, despite its vast reach. Nevertheless, the ambitious objective proposed by the investigation group offers the most comprehensive published results concerning the exposure level of Europeans to potentially polluted particulate matter.

As future suggestions, more efforts are still necessary to systematically collect information on the existing relationship between particulate matter, constitutes, and toxicity in cancerous and/or non-cancerous risk estimation. In this respect, it is possible to use the hazard index to estimate the allowable amount of particulate matter as per the maximum permissible concentrations of each PTEs. Further laboratory and field studies are needed to explicitly collect the aforementioned information to incorporate PM-associated constitutes in the health risk assessment process. Additionally, risk estimation of toxic interactions of PTEs in different sub-mixtures is also required since there is very rare or no information available for such risk estimation. Only a few medical field-related studies showed that metals could interact with each other and interfere with each other's toxic effects (Choudhury & Mudipalli, 2008; Das et al., 2020). They investigated the combined toxicity of PTEs to rat models. Still, the collected information did not apply to human health risk estimation because human receptors' toxicity benchmarks are unavailable. Moreover, the present USEPA database does not include the reference dose values of the mixture of constitutes (U.S. EPA, 2004), for instant, Cd-Pb, Ni-Pb, and Ni-Cd, etc. So, it is challenging to assess cancerous and/or non-cancerous risks in the context of exposure to a mixture of PM-associated PTEs.

So further detailed studies are required to include the interaction of different metals in risk assessment, to provide guidelines including doseresponse data of metals' mixture to human health, and to reduce the associated uncertainty with risk estimation of exposure to the mixture of PM-associated PTEs. Besides, other concerns raising uncertainties can be attributed to the lack of indoor measurement of some specific pollutants since people spend most of their time indoors, where pollutant concentrations differ from outdoors (Guo et al., 2004). It is also essential to estimate the amount of time people spend indoors and outdoors, as well as pollution types and their concentrations, to have a more realistic health risk assessment. The accurate characterisation of the investigated population is also necessary to determine the inhalation exposure, such as body weight, sex, lung surface area, health condition, and lifestyle Gruszecka-Kosowska (2018); (Gruszecka-Kosowska, 2018; Yousefi et al., 2022; Lewandowska et al., 2019; Pachoulis et al., 2022; Colas et al., 2022). Therefore, corrective actions are required to reduce the risk rates below acceptable levels once health risk values are accurately estimated.

# 5. Conclusion

To investigate the adverse impacts of being exposed to  $PM_{10}$ -bound PTEs, the current study conducted health risk analyses associated with exposure to  $PM_{10}$ -bound PTEs, both cancerous and non-cancerous risks, in selected European cities from 2013 to 2019.

Despite the observed decreasing trend in the  $PM_{10}$  concentrations, the recommended allowable daily  $PM_{10}$  concentrations were exceeded in the vast part of Europe, especially during the cold period of the year. Based on our results, the cities of Antwerpen (39.7 ng/m<sup>3</sup>), Kladno (6.7 ng/m<sup>3</sup>), Gibraltar (16.9 ng/m<sup>3</sup>), Antwerpen (618.7 ng/m<sup>3</sup>) had the highest annual values of As, Cd, Ni, and Pb, respectively from 2013 to 2019. The correlation analysis between time and PTEs' concentrations showed a positive correlation with time in some cities.

The estimated hazard quotient by representing non-carcinogenic effects, with values less than one, showed no risk to adult men, women, children, and infants due to the inhalation of particle-bound PTEs. On the other side, calculated carcinogenic risk rates (R) showed unacceptable risk levels in the case of As (in PM<sub>10</sub>), Cd (in PM<sub>10</sub>), Ni (in PM<sub>10</sub>), and Pb (in PM<sub>10</sub>) in the vast part of Europe among adults (men & women), children, and infants between 2013 and 2019. The highest R values of exposure of PM-associated PTEs, irrespective of the subpopulation type, belong to As, while the smallest one was Pb. Carcinogenic risk rates were higher among children, followed by infants, adult women, and adult men. Regardless of the subpopulation type, carcinogenic risk rates followed a negative correlation with time. While some cities with a positive correlation indicated an increasing carcinogenic

risk following the order of R–Ni > R–As > R–Cd > R–Pb between 2013 and 2019. It means that the amount of evaluated PTEs can increase the cancer risk rates among inhabitants by increasing R values.

In conclusion, our results highlighted (a) the need to control PM sources rather than overall PM concentrations and (b) the importance of health impacts from exposure to air pollution. Current emission inventories must be improved to help solve present and future air quality issues. To address current and future issues, emission inventories should include species so far ignored, such as isotopes, heavy metals, and intermediate volatility organic compounds (IVOCs) that contribute to many chemical processes involving PM. Moreover, implementing accurate health risk assessment methods caused by exposure to ambient particles is essential in controlling and mitigating urban air pollution.

# Credit author statement

Parya Broomandi: Writing - Original Draft, Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data Curation, Data analysis.; Andrés Rodríguez Seijo: Data Curation, Methodology, Review & Editing.; Nasime Janatian: Data analysis, Data curation.; Aram Fathian: Data Curation, Formal analysis, software.; Aidana Tleuken: Data Curation, Formal analysis.; Kaveh Mohammadpour: Data Curation, Formal analysis.; David Galán-Madruga: Data analysis, Data curation, Formal analysis, Review & Editing.; Ali Jahanbakhshi: Data Curation, Investigation, Formal analysis.; Jong Ryeol Kim: Resources, Data Curation, Project administration, Funding acquisition.; Alfrendo Satyanaga: Resources, Data Curation, Project administration, Funding acquisition.; Mehdi Bagheri: Resources, Data Curation, Project administration, Funding acquisition.; Lidia Morawska: Supervision, Validation, Review & Editing.

# **Ethical approval**

Not applicable.

# Consent to participate

Not applicable.

# **Consent for publication**

Not applicable.

#### 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.

# Data availability

Data will be made available on request.

# Acknowledgements

The authors acknowledge the financial support for the NU project (Nazarbayev Research Fund SOE2017004 & 11022021CRP1512 & OPCO2021003). This research was also supported by the CIIMAR Strategic Funding UIDB/04423/2020, UIDP/04423/2020 through national funds provided by the Portuguese Foundation for Science and Technology (FCT) and MCTES and the co-funding by the FEDER, within the PT2020 Partnership Agreement and Compete 2020. Andrés Rodríguez-Seijo would like to acknowledge the FCT and CIIMAR for their contract under the Scientific Employment Stimulus-Individual 2017 program (CEECIND/03794/2017) and MCIN/AEI/UVigo for their contract under the "Actuación financiada por IJC2020-044197-I/MCIN/AEI/

10.13039/501100011033 y por la Unión Europea NextGenerationEU"/ PRTR.

#### Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.121232.

#### References

- Acosta, J.A., Faz, Á., Kalbitz, K., Jansen, B., Martínez-Martínez, S., 2011. Heavy metal concentrations in particle size fractions from street dust of Murcia (Spain) as the basis for risk assessment. J. Environ. Monit. : JEM 13 (11), 3087–3096. https://doi. org/10.1039/c1em10364d.
- Aliff, A., Abdul Wahab, A.A., Shahrol, M., Abd Wahil, M.S., Mun, W., Ja'afar, H., 2020. Health risk assessment among adult and children on potential air pollutants released from the petrochemical plant in Malaysia: the result of air modelling. Pertanika Journal of Science and Technology 28, 859–878.
- Almeida, T.S., Brancher, M., de Melo Lisboa, H., Franco, D., Maranhão, T.A., Borges, D.L. G., 2020. Direct analysis of particulate matter (PM10) for the determination of Be, Cd and Pb using high resolution-continuum source electrothermal atomic absorption spectrometry: assessment of the potential correlation between analyte content and meteorological pa. Spectrochim. Acta B Atom Spectrosc. 172, 105951 https://doi. org/10.1016/j.sab.2020.105951.
- ATSDR, 2005. Public Health Assessment Guidance Manual.
- Barmpadimos, I., Hueglin, C., Keller, J., Henne, S., Prevot, A., 2011. Influence of meteorology on PM10 trends and variability in Switzerland from 1991 to 2008. Atmos. Chem. Phys. 11, 1813–1835. https://doi.org/10.5194/acp-11-1813-2011.
- Batbold, C., Chonokhuu, S., Buuveijargal, K., Gankhuyag, K., 2021. Source apportionment and spatial distribution of heavy metals in atmospheric settled dust of Ulaanbaatar, Mongolia. Environ. Sci. Pollut. Control Ser. 28 (33), 45474–45485. https://doi.org/10.1007/s11356-021-13861-2.
- Bello, S., Muhammad, B.G., Bature, B., 2017. Total excess lifetime cancer risk estimation from enhanced heavy metals concentrations resulting from tailings in katsina steel rolling mill, Nigeria. J. Mater. Sci. Eng. 6, 338. https://doi.org/10.4172/2169-0022.1000338.
- Beloconi, A., Vounatsou, P., 2021. Substantial reduction in particulate matter air pollution across Europe during 2006–2019: a spatiotemporal modeling analysis. Environ. Sci. Technol. 55 (22), 15505–15518. https://doi.org/10.1021/acs. est.1c03748.
- Borge, R., Requia, W.J., Yagüe, C., Jhun, I., Koutrakis, P., 2019. Impact of weather changes on air quality and related mortality in Spain over a 25 year period [1993–2017]. Environ. Int. 133, 105272 https://doi.org/10.1016/j. envint.2019.105272.
- Buchanan, C.M., Beverland, I.J., Heal, M.R., 2002. The influence of weather-type and long-range transport on airborne particle concentrations in Edinburgh, UK. Atmos. Environ. 36 (34), 5343–5354. https://doi.org/10.1016/S1352-2310(02)00579-4.
- CalEPA, 2016. Office of Environmental Health Hazard Assessment Assessments. http://oehha.ca.gov/chemicals.
- Chalvatzaki, E., Chatoutsidou, S.E., Lehtomäki, H., Almeida, S.M., Eleftheriadis, K., Hänninen, O., Lazaridis, M., 2019. Characterization of human health risks from particulate air pollution in selected European cities. Atmosphere 10 (2), 96. https:// doi.org/10.3390/atmos10020096.
- Chen, J., Hoek, G., 2020. Long-term exposure to PM and all-cause and cause-specific mortality: a systematic review and meta-analysis. Environ. Int. 143, 105974 https:// doi.org/10.1016/j.envint.2020.105974.
- Chlebowska-Styś, A., Sówka, I., Kobus, D., Pachurka, Ł., 2017. Analysis of concentrations trends and origins of PM 10 in selected European cities. E3S Web of Conferences 17. https://doi.org/10.1051/e3sconf/20171700013.
- Choudhury, H., Mudipalli, A., 2008. Potential considerations & concern in the risk characterization for the interaction profiles of metals. Indian J. Med. Res. 128, 462–483.
- Colas, A., Baudet, A., Le Cann, P., Blanchard, O., Gangneux, J.-P., Baurès, E., Florentin, A., 2022. Quantitative health risk assessment of the chronic inhalation of chemical compounds in healthcare and elderly care facilities. Toxics 10 (3). https:// doi.org/10.3390/toxics10030141.
- Curtis, L., Rea, W., Smith-Willis, P., Fenyves, E., Pan, Y., 2006. Adverse health effects of outdoor air pollutants. Environ. Int. 32 (6), 815–830. https://doi.org/10.1016/j. envint.2006.03.012.
- Czernecki, B., Półrolniczak, M., Kolendowicz, L., Marosz, M., Kendzierski, S., Pilguj, N., 2017. Influence of the atmospheric conditions on PM10 concentrations in Poznań, Poland. J. Atmos. Chem. 74, 1–25. https://doi.org/10.1007/s10874-016-9345-5.
- Das, A., Singh, G., Habib, G., Kumar, A., 2020. Non-carcinogenic and carcinogenic risk assessment of trace elements of PM2.5 during winter and pre-monsoon seasons in Delhi: a case study. Exposure and Health 12 (1), 63–77. https://doi.org/10.1007/ s12403-018-0285-v.
- Daston, G., Faustman, E., Ginsberg, G., Fenner-Crisp, P., Olin, S., Sonawane, B., Bruckner, J., Breslin, W., McLaughlin, T.J., 2004. A framework for assessing risks to children from exposure to environmental agents. Environ. Health Perspect. 112 (2), 238–256. https://doi.org/10.1289/ehp.6182.
- Duan, J., Tan, J., Hao, J., Chai, F., 2014. Size distribution, characteristics and sources of heavy metals in haze episod in Beijing. J. Environ. Sci. 26 (1), 189–196. https://doi. org/10.1016/S1001-0742(13)60397-6.

- EEA, 2014. European Environment Agency (2014) Air Quality in Europe 2014 Report. EEA Technical Report No 5/2014. https://doi.org/10.2800/22775.
- EEA, 2015. Air Quality in Europe 2015 Report. EEA Report 5/2015. European Environmental Agency, Copenhagen, Denmark. Available at: http://www.eea.euro pa.eu/publications/air-quality-in-europe-2015.
- EEA, 2016. Exceedance of Air Quality Limit Values in Urban Areas. Indicator Assessment. European Environment Agency, Copenhagen, Denmark. http://www.eea.europa. eu/data-and-maps/indicators/exceedance-of-airquality- limit-3/assessment-2.
- EEA, 2018. Global and European Temperature. European Environment Agency, Copenhagen, Denmark. Available at: https://www.eea.europa.eu/data-andmaps /indicators/global-and-european-temperature-4/assessment.
- EEA, 2019. European Environment Agency (2019) Air Quality in Europe 2019 Report. EEA Technical Report No 10/2019. https://doi.org/10.2800/822355.
- Effatpanah, M., Effatpanah, H., Jalali, S., Parseh, I., Goudarzi, G., Barzegar, G., Geravandi, S., Darabi, F., Ghasemian, N., Mohammadi, M.J., 2020. Hospital admission of exposure to air pollution in Ahvaz megacity during 2010–2013. Clinical Epidemiology and Global Health 8 (2), 550–556. https://doi.org/10.1016/j. cegh.2019.12.001.
- Ekpenyong, C.E., Asuquo, A.E., 2017. Recent advances in occupational and environmental health hazards of workers exposed to gasoline compounds. Int. J. Occup. Med. Environ. Health 30 (1), 1–26. https://doi.org/10.13075/ ijomeb.1896.00800.
- Garbero, V., Montalto, A., Lazovic, N., Salizzoni, P., Berrone, S., Soulhac, L., 2011. The impact of the urban air pollution on the human health: a case-study in turin. In: Steyn, D., Trini Castelli, S. (Eds.), Air Pollution Modeling And its Application XXI. NATO Science For Peace And Security Series C: Environmental Security. Springer, Dordrecht, pp. 729–732. https://doi.org/10.1007/978-94-007-1359-8119.
- Godish, T., Davis, W.T., Fu, J.S., 2015. Air Quality, fifth ed. CRC Press. https://doi.org/ 10.1201/b17341.
- Gozzi, F., Marcelli, A., Lucci, F., 2017. Current status of particulate matter pollution in Europe and future perspectives: A review 8, 1901–1909.
- Groot Zwaaftink, C., Aas, W., Eckhardt, S., Evangeliou, N., Hamer, P., Johnsrud, M., Kylling, A., Platt, S., Stebel, K., Uggerud, H., Yttri, K., 2021. What Caused a Record High PM<sub>10</sub> Episode in Northern Europe In October 2020? https://doi.org/10.5194/ acp-2021-496.
- Gruszecka-Kosowska, A., 2018. Assessment of the Kraków inhabitants' health risk caused by the exposure to inhalation of outdoor air contaminants. Stoch. Environ. Res. Risk Assess. 32, 485–499. https://doi.org/10.1007/s00477-016-1366-8.
- Guerreiro, C.B.B., Foltescu, V., de Leeuw, F., 2014. Air quality status and trends in Europe. Atmos. Environ. 98, 376–384. https://doi.org/10.1016/j. atmoseny.2014.09.017.
- Guerreiro, C.B.B., Horálek, J., de Leeuw, F., Couvidat, F., 2016. Benzo(a)pyrene in Europe: ambient air concentrations, population exposure and health effects. Environ. Pollut. 214, 657–667. https://doi.org/10.1016/j.envpol.2016.04.081.
- Guevara, M., 2016. Emissions of primary particulate matter. In: Harrison, R.M., Hester, R.E., Querol, X. (Eds.), Airborne Particulate Matter: Sources, Atmospheric Processes and Health. The Royal Society of Chemistry, pp. 1–34. https://doi.org/ 10.1039/9781782626589-00001.
- Guo, H., Lee, S.C., Chan, L.Y., Li, W.M., 2004. Risk assessment of exposure to volatile organic compounds in different indoor environments. Environ. Res. 94 (1), 57–66. https://doi.org/10.1016/S0013-9351(03)00035-5.
- Guttikunda, S.K., Lodoysamba, S., Bulgansaikhan, B., Dashdondog, B., 2013. Particulate pollution in ulaanbaatar, Mongolia. Air Quality, Atmosphere & Health 6 (3), 589–601. https://doi.org/10.1007/s11869-013-0198-7.
- Holst, J., Mayer, H., Holst, T., 2008. Effect of meteorological exchange conditions on PM10 concentration. Meteorol. Z. 17, 273–282. https://doi.org/10.1127/0941-2948/2008/0283.

Idani, E., Geravandi, S., Akhzari, M., Goudarzi, G., Alavi, N., Yari, A.R., Mehrpour, M., Khavasi, M., Bahmaei, J., Bostan, H., Dobaradaran, S., Salmanzadeh, S., Mohammadi, M.J., 2020. Characteristics, sources, and health risks of atmospheric PM10-bound heavy metals in a populated middle eastern city. Toxin Rev. 39 (3), 266–274. https://doi.org/10.1080/15569543.2018.1513034.

Jamshidi, M., Sanei, M., Mahmoodirad, A., Hoseinzadeh Lotfi, F., Tohidi, G., 2021. Uncertain SBM data envelopment analysis model: a case study in Iranian banks. Int. J. Finance Econ. 26 (2), 2674–2689.

- Kendall, M.A., 1949 [Pp. vii + 160. In: Maurice, G., Kendall, M.A. (Eds.), Rank Correlation Methods. Journal of the Institute of Actuaries, London: Charles Griffin and Co. Ltd., 42 Drury Lane, 1948. 18s.], pp. 140–141. https://doi.org/10.1017/ S0020268100013019, 75(1).
- Kim, K.-H., Kabir, E., Kabir, S., 2015. A review on the human health impact of airborne particulate matter. Environ. Int. 74, 136–143. https://doi.org/10.1016/j. envint.2014.10.005.
- Kumar, V., Kothiyal, N.C., Saruchi, Vikas, P., Sharma, R., 2016. Sources, distribution, and health effect of carcinogenic polycyclic aromatic hydrocarbons (PAHs) – current knowledge and future directions. Journal of the Chinese Advanced Materials Society 4 (4), 302–321. https://doi.org/10.1080/22243682.2016.1230475.
- Kursun Unver, I., Terzi, M., 2018. Distribution of trace elements in coal and coal fly ash and their recovery with mineral processing practices: a review. Journal of Mining and Environment 9 (3), 641–655. https://doi.org/10.22044/jme.2018.6855.1518.
- Lau, A., Shafiee, M., Smoot, G., Grossan, B., Li, S., Maksut, Z., 2020. On-sky SiPM performance measurements for millisecond to sub-microsecond optical source variability studies. Intl Soc Optical Eng 6 (4). https://doi.org/10.1117/1. jatis.6.4.046002.
- Lee, P.-K., Youm, S.-J., Jo, H.Y., 2013. Heavy metal concentrations and contamination levels from Asian dust and identification of sources: a case-study. Chemosphere 91 (7), 1018–1025. https://doi.org/10.1016/j.chemosphere.2013.01.074.

- Leili, M., Nadali, A., Karami, M., Bahrami, A., Afkhami, A., 2021. Short-term effect of multi-pollutant air quality indexes and PM(2.5) on cardiovascular hospitalization in Hamadan, Iran: a time-series analysis. Environ. Sci. Pollut. Res. Int. 28 (38), 53653–53667. https://doi.org/10.1007/s11356-021-14386-4.
- Lewandowska, A.M., Rudzki, M., Rudzki, S., Lewandowski, T., Laskowska, B., 2019. Environmental risk factors for cancer - review paper. Ann. Agric. Environ. Med. : AAEM 26 (1), 1–7. https://doi.org/10.26444/aaem/94299.
- Lim, S., Vos, T., Flaxman, A., Danaei, G., Shibuya, K., Adair-Rohani, H., Al-Mazroa, M., Amann, M., Anderson, H., Andrews, K., Aryee, M., Atkinson, C., Bacchus, L., Bahalim, A., Meng, Balakrishnan, K., Balmes, J., Barker, S., Baxter, A., Ezzati, M., 2012. A comparative risk assessment of burden of disease and injury attributable to 67 risk factors and risk factor clusters in 21 regions, 1990-2010: a systematic analysis for the Global Burden of Disease Study 2010. Lancet 380, 2224–2260. https://doi.org/10.1016/S0140-6736(12)61766-8.
- López, A., Yusà, V., Muñoz, A., Vera, T., Borràs, E., Ródenas, M., Coscollà, C., 2017. Risk assessment of airborne pesticides in a Mediterranean region of Spain. Sci. Total Environ. 574, 724–734. https://doi.org/10.1016/j.scitotenv.2016.08.149.
- Lyu, Y., Zhang, K., Chai, F., Cheng, T., Yang, Q., Zheng, Z., Li, X., 2017. Atmospheric sizeresolved trace elements in a city affected by non-ferrous metal smelting: indications of respiratory deposition and health risk. Environ. Pollut. 224, 559–571. https://doi. org/10.1016/j.envpol.2017.02.039.
- Mahmoodirad, A., Niroomand, S., 2020. A belief degree-based uncertain scheme for a biobjective two-stage green supply chain network design problem with direct shipment. Soft Comput. 24 (24), 18499–18519.
- Mahmoodirad, A., Sanei, M., 2016. Solving a multi-stage multi-product solid supply chain network design problem by meta-heuristics. Sci. Iran. 23 (3), 1429–1440.
- Mahmoodirad, A., Dehghan, R., Niroomand, S., 2019. Modelling linear fractional transportation problem in belief degree-based uncertain environment. J. Exp. Theor. Artif. Intell. 31 (3), 393–408.
- Mamouri, R.-E., Ansmann, A., Nisantzi, A., Solomos, S., Kallos, G., Hadjimitsis, D., 2016. Extreme dust storm over the eastern Mediterranean in September 2015: satellite, lidar, and surface observations in the Cyprus region. Atmos. Chem. Phys. 16, 13711–13724. https://doi.org/10.5194/acp-16-13711-2016.
- Mann, H.B., 1945. Nonparametric tests against trend. Econometrica 13 (3), 245–259. https://doi.org/10.2307/1907187.
- Martin, R., Dowling, K., Pearce, D., Sillitoe, J., Florentine, S., 2014. Health effects associated with inhalation of airborne arsenic arising from mining operations. In: Geosciences, pp. 128–175. https://doi.org/10.3390/geosciences4030128. Vol. 4, Issue 3.
- Megaritis, A., Fountoukis, C., Charalampidis, P., Pilinis, C., Pandis, S., 2013. Response of fine particulate matter concentrations to changes of emissions and temperature in Europe. Atmos. Chem. Phys. 13, 3423–3443. https://doi.org/10.5194/acp-13-3423-2013.
- Megido, L., Suárez-Peña, B., Negral, L., Castrillón, L., Fernández-Nava, Y., 2017. Suburban air quality: human health hazard assessment of potentially toxic elements in PM10. Chemosphere 177, 284–291. https://doi.org/10.1016/j. chemosphere.2017.03.009.
- Mirzaei, N., Mahmoodirad, A., Niroomand, S., 2019. An uncertain multi-objective assembly line balancing problem: a credibility-based fuzzy modeling approach. Int. J. Fuzzy Syst. 21 (8), 2392–2404.
- Molla-Alizadeh-Zavardehi, S., Mahmoodirad, A., Rahimian, M., 2014. Step fixed charge transportation problems via genetic algorithm. Indian Journal of Science and Technology 7 (7), 949–954.
- Morakinyo, O., Mokgobu, I., Mukhola, M., Engelbrecht, J., 2017. Health risk assessment of exposure to ambient concentrations of benzene, toluene and xylene in Pretoria West, South Africa. African Journal of Science, Technology, Innovation and Development 9, 1–8. https://doi.org/10.1080/20421338.2017.1352123.
- Morawska, L., Zhu, T., Liu, N., Amouei Torkmahalleh, M., de Fatima Andrade, M., Barratt, B., Broomandi, P., Buonanno, G., Carlos Belalcazar Ceron, L., Chen, J., Cheng, Y., Evans, G., Gavidia, M., Guo, H., Hanigan, I., Hu, M., Jeong, C.H., Kelly, F., Gallardo, L., Ye, C., 2021. The state of science on severe air pollution episodes: quantitative and qualitative analysis. Environ. Int. 156, 106732 https://doi.org/ 10.1016/j.envint.2021.106732.
- Nidzgorska-Lencewicz, J., Czarnecka, M., 2015. Winter weather conditions vs. air quality in Tricity, Poland. Theor. Appl. Climatol. 119 (3), 611–627. https://doi.org/ 10.1007/s00704-014-1129-8.
- Niroomand, S., Garg, H., Mahmoodirad, A., 2020a. An intuitionistic fuzzy two stage supply chain network design problem with multi-mode demand and multi-mode transportation. ISA (Instrum. Soc. Am.) Trans. 107, 117–133.
- Niroomand, S., Mosallaeipour, S., Mahmoodirad, A., 2020b. A hybrid simple additive weighting approach for constrained multicriteria facilities location problem of glass production industries under uncertainty. IEEE Trans. Eng. Manag. 67 (3), 846–854.
- Pachoulis, T., Panagopoulos, P., Dasopoulou, M., Balla, D., Stamatelopoulou, A., Manousakas, M., Eleftheriadis, K., Saraga, D., 2022. Population health risks assessment from air pollution exposure in an industrialized residential area in Greece. Atmosphere 13 (4). https://doi.org/10.3390/atmos13040615.
- Pacyna, E., Pacyna, J., Fudala, J., Strzelecka-Jastrzab, E., Hlawiczka, S., Panasiuk, D., Nitter, S., Pregger, T., Pfeiffer, H., Friedrich, R., 2007. Current and future emissions of selected heavy metals to the atmosphere from anthropogenic sources in Europe. Atmos. Environ. 41, 8557–8566. https://doi.org/10.1016/j.atmosenv.2007.07.040.
- Panda, U., Boopathy, R., Gadhavi, H.S., Renuka, K., Gunthe, S.S., Das, T., 2021. Metals in coarse ambient aerosol as markers for source apportionment and their health risk assessment over an eastern coastal urban atmosphere in India. Environ. Monit. Assess. 193 (5), 311. https://doi.org/10.1007/s10661-021-09057-3.
- Pinto, E., Soares, C., Couto, C.M.C.M., Almeida, A., 2015. Trace elements in ambient air at porto metropolitan area-checking for compliance with new European union (EU)

air quality standards. J. Toxicol. Environ. Health, Part A 78 (13–14), 848–859. https://doi.org/10.1080/15287394.2015.1051177.

- Pommier, M., 2021. Prediction of source contributions to urban background PM\$\_{10}\$ concentrations in European cities: a case study for an episode in December 2016 using EMEP/MSC-W rv4.15 Part 2: the city contribution. Geosci. Model Dev. (GMD) 14 (6), 4143–4158. https://doi.org/10.5194/gmd-14-4143-2021.
- Pommier, M., Fagerli, H., Schulz, M., Valdebenito, A., Kranenburg, R., Schaap, M., 2020. Prediction of source contributions to urban background PM\$\_{10}\$ concentrations in European cities: a case study for an episode in December 2016 using EMEP/NSC W rv4.15 and LOTOS-EUROS v2.0 – Part 1: the country contributions. Geosci. Model Dev. (GMD) 13 (4), 1787–1807. https://doi.org/10.5194/gmd-13-1787-2020.

QGIS, 2022. QGIS Geographic Information System (Version 3.22.3) Open-Source Geospatial Foundation Project. https://www.qgis.org/en/site/forusers/download.ht ml.

- Reizer, M., Juda-Rezler, K., 2016. Explaining the high PM10 concentrations observed in Polish urban areas. Air Quality, Atmosphere, & Health 9, 517–531. https://doi.org/ 10.1007/s11869-015-0358-z.
- Romanazzi, V., Casazza, M., Malandrino, M., Maurino, V., Piano, A., Schilirò, T., Gilli, G., 2014. PM10 size distribution of metals and environmental-sanitary risk analysis in the city of Torino. Chemosphere 112, 210–216. https://doi.org/10.1016/j. chemosphere.2014.04.034.
- Samek, L., 2016. Overall human mortality and morbidity due to exposure to air pollution. Int. J. Occup. Med. Environ. Health 29 (3), 417–426. https://doi.org/ 10.13075/ijomeh.1896.00560.
- Samoli, E., Peng, R., Ramsay, T., Pipikou, M., Touloumi, G., Dominici, F., Burnett, R., Cohen, A., Krewski, D., Samet, J., Katsouyanni, K., 2008. Acute effects of ambient particulate matter on mortality in Europe and North America: results from the APHENA study. Environ. Health Perspect. 116 (11), 1480–1486. https://doi.org/ 10.1289/ehp.11345.
- Schlutow, A., Schröder, W., Scheuschner, T., 2021. Assessing the relevance of atmospheric heavy metal deposition with regard to ecosystem integrity and human health in Germany. Environ. Sci. Eur. 33 (1), 7. https://doi.org/10.1186/s12302-020-00391-w.
- Schwenk, M., Gundert-Remy, U., Heinemeyer, G., Olejniczak, K., Stahlmann, R., Kaufmann, W., Bolt, H.M., Greim, H., von Keutz, E., Gelbke, H.P., 2003. Children as a sensitive subgroup and their role in regulatory toxicology: DGPT workshop report. Arch. Toxicol. 77 (1), 2–6. https://doi.org/10.1007/s00204-002-0416-9.
- Shafiee, M., Behnamian, H., Feghhi, S.A. hossein, 2017. A study of wake potentials for the pick-ups in storage ring. J. Instrum. 12 https://doi.org/10.1088/1748-0221/12/ 12/T12006. T12006-T12006.
- Shafiee, M., Fedorov, D., Grossan, B., Kizheppatt, V., Smoot, G., 2021. A readout system for microwave kinetic inductance detectors using software defined radios. J. Instrum. 16, P07015 https://doi.org/10.1088/1748-0221/16/07/P07015.
- Shafiee, M., Grossan, B., Hu, J., Colantoni, I., Smoot, G., 2019. Design optimization of a 10 kilopixel optical band microwave kinetic inductance detector. J. Instrum. 14 https://doi.org/10.1088/1748-0221/14/12/p12011. P12011–P12011.
- Sicard, P., Agathokleous, E., De Marco, A., Paoletti, E., Calatayud, V., 2021. Urban population exposure to air pollution in Europe over the last decades. Environ. Sci. Eur. 33 (1), 28. https://doi.org/10.1186/s12302-020-00450-2.
- Sicard, P., Paoletti, E., Agathokleous, E., Araminienė, V., Proietti, C., Coulibaly, F., De Marco, A., 2020. Ozone weekend effect in cities: deep insights for urban air pollution control. Environ. Res. 191, 110193 https://doi.org/10.1016/j.envres.2020.110193.
- Silva Junior, C.H.L., Almeida, C.T., Santos, J.R.N., Anderson, L.O., Aragão, L.E.O.C., Silva, F.B., 2018. Spatiotemporal rainfall trends in the Brazilian legal amazon between the years 1998 and 2015. Water 10, 1220. https://doi.org/10.3390/ w10091220.
- Singh, D.K., Gupta, T., 2016. Source apportionment and risk assessment of PM1 bound trace metals collected during foggy and non-foggy episodes at a representative site in the Indo-Gangetic plain. Sci. Total Environ. 550, 80–94. https://doi.org/10.1016/j. scitotenv.2016.01.037.
- Sternbeck, J., Sjödin, Å., Andréasson, K., 2002. Metal emissions from road traffic and the influence of resuspension – results from two tunnel studies. Atmos. Environ. 36, 4735–4744. https://doi.org/10.1016/S1352-2310(02)00561-7.
- Taiwo, A.M., Awomeso, J.A., Taiwo, O.T., Oremodu, B.D., Akintunde, O.O., Ojo, N.O., Elegbede, O.O., Olanrewaju, H.H., Arowolo, T.A., 2017. Assessment of health risks associated with road dusts in major traffic hotspots in Abeokuta metropolis, Ogun state, southwestern Nigeria. Stoch. Environ. Res. Risk Assess. 31 (2), 431–447. https://doi.org/10.1007/s00477-016-1302-y.
- Tan, W., Du, H., Liu, L., Su, T., Liu, X., 2017. Experimental and numerical study of ammonia leakage and dispersion in a food factory. J. Loss Prev. Process. Ind. 47, 129–139. https://doi.org/10.1016/j.jlp.2017.03.005.
- Tong, R., Liu, J., Wang, W., Fang, Y., 2020. Health effects of PM2.5 emissions from onroad vehicles during weekdays and weekends in Beijing, China. Atmos. Environ. 223, 117258 https://doi.org/10.1016/j.atmosenv.2019.117258.
- U.S. EPA, 2004. Regional Screening Level. http://www.epa.gov/regio%0An9/super%20 fund/prg/.
- US EPA, 2007. Concepts, Methods and Data Sources for Cumulative Health Risk Assessment of Multiple Chemicals, Exposures and Effects: a Resource Document. EPA/600/R-06/013F.

US EPA, 2016. Integrated Risk Information System (IRIS) (Glossary).

Valavanidis, A., Fiotakis, K., Vlachogianni, T., 2008. Airborne particulate matter and human health: toxicological assessment and importance of size and composition of particles for oxidative damage and carcinogenic mechanisms. J. Environ. Sci. Health C Environ. Carcinog. Ecotoxicol. Rev. 26 (4), 339–362. https://doi.org/10.1080/ 10590500802494538.

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- Vestreng, V., Ntziachristos, L., Semb, A., Reis, S., Isaksen, I.S.A., Tarrasón, L., 2009. Evolution of NOx emissions in Europe with focus on road transport control measures. Atmos. Chem. Phys. 9 (4), 1503–1520. https://doi.org/10.5194/acp-9-1503-2009.
- Volná, V., Hladký, D., 2020. Detailed assessment of the effects of meteorological conditions on PM10 concentrations in the northeastern part of the Czech republic. Atmosphere 11, 497. https://doi.org/10.3390/atmos11050497.
- WHO, 2005. WHO Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur Dioxide: Global Update 2005 : Summary of Risk Assessment. World Health Organization, Geneva, Switzerland. https://apps.who.int/iris/handle/10 665/69477.
- WHO, 2021. WHO Global Air Quality Guidelines: Particulate Matter (PM2.5 and PM10), Ozone, Nitrogen Dioxide, Sulfur Dioxide and Carbon Monoxide. World Health Organization. https://apps.who.int/iris/handle/10665/345329. License: CC BY-NC-SA 3.0 IGO.
- Yang, Y., Fang, W., Xue, M., Xu, Z., Huang, C., 2014. TSP, PM10 and health risk assessment for heavy metals (Cr, Ni, Cu, Zn, Cd, Pb) in the ambience of the

production line for waste cathode ray tube recycling. J. Mater. Cycles Waste Manag. 18, 1–7. https://doi.org/10.1007/s10163-014-0331-1.

- Yousefi, H., Lak, E., Mohammadi, M.J., Shahriyari, H.A., 2022. Carcinogenic risk assessment among children and adult due to exposure to toxic air pollutants. Environ. Sci. Pollut. Control Ser. 29 (16), 23015–23025. https://doi.org/10.1007/ s11356-021-17300-0.
- Yousefi, R., Wang, F., Ge, Q., Shaheen, A., 2021. Long-term aerosol optical depth trend over Iran and identification of dominant aerosol types. Sci. Total Environ. 722, 137906 https://doi.org/10.1016/j.scitotenv.2020.137906.
- Zhi, M., Zhang, X., Zhang, K., Ussher, S.J., Lv, W., Li, J., Gao, J., Luo, Y., Meng, F., 2021. The characteristics of atmospheric particles and metal elements during winter in Beijing: size distribution, source analysis, and environmental risk assessment. Ecotoxicol. Environ. Saf. 211, 111937 https://doi.org/10.1016/j. ecoenv.2021.111937.
- Zhou, K., Ye, Y., Liu, Q., Liu, A., Peng, S., 2007. Evaluation of ambient air quality in Guangzhou, China. J. Environ. Sci. (China) 19, 432–437. https://doi.org/10.1016/ S1001-0742(07)60072-2.