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# In-Depth Study of $PM_{2.5}$ and $PM_{10}$ Concentrations over a 12-Year Period and Their Elemental Composition in the Lignite Center of Western Macedonia, Greece

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**Abstract:** Western Macedonia, located in North-western Greece, is largely dominated by lignite mining and lignite-fired power plants, which are a significant part of the country's economy. However, the electricity generation and the related activities are among the greatest sources of air pollutants. In this study, we focus on the air quality of Western Macedonia based on measurements of Particulate Matter ($PM_{2.5}$ and $PM_{10}$) over a 12-year period (from 2010 to 2021) and a sampling of PM-bound trace elements over the course of 12 months (from December 2017 to November 2018). The analysis revealed an overall decrease of $PM_{2.5}$ and $PM_{10}$ concentrations over the study period. In general, the concentrations of PM exhibited seasonality patterns associated with the weather conditions and the local sources of air pollutants. These major sources of air pollution are the lignite mining processes, the emissions from the lignite-fired power plants, and the anthropogenic emissions from the biomass burning and heating systems. In addition, the analysis of the PM-bound trace elements revealed some differences; the elemental profiles of both $PM_{2.5}$ and $PM_{10}$ were quite similar, while the most abundant elements (Ca and Si) indicate that the main emission sources were related with the mining activities and the coal combustion.

**Keywords:** Western Macedonia; lignite-fired power plants; particulate matter (PM); PM-bound trace elements

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## 1. Introduction

In the context of the European Green Deal and in line with the EU's commitment to global climate action under the Paris Agreement in 2016, the EU aims to be 'climate-neutral' and reach net-zero greenhouse gas emissions by 2050. In order to achieve the target emissions reductions, the transition to renewable energy sources is required. The coal use is one of the greatest sources of greenhouse gas emissions and is one of the culprits in the deterioration of air quality. From the perspective of economy, coal is a traditional, reliable, and low-cost option for electricity production, which has been used since the early 1900s for commercial purposes.

On the other hand, coal-fired power plants release air pollutants such as sulphur dioxide ($SO_2$), nitrogen oxides (NOx), particulate matter (PM), as well as heavy metals (e.g., mercury), and fly ash. These pollutants have a great influence on human health, contributing to illnesses and premature deaths [1]. The use of lignite in coal-fired power stations without substantial pollution control technologies violates the EU Industrial Emissions Directive and is responsible for air pollution-related deaths. In Europe, the air pollution from coal power plants is responsible for 33,900 (95% CI: 33,000–37,600) excess deaths attributable to cardiovascular and respiratory diseases per year [2]. When the Paris climate agreement was signed in 2016, there were 324 coal-fired power stations in Europe. Since then, major power plants have been closed and will be shut down 50% by 2030. Although the emissions from large combustion plants in the EU have decreased over the period 2004–2019, their impact on the environment remains persistent.

In Poland, air pollutant measurements in the vicinity of the Adamów coal-fired power plant over the period 2009–2015 indicated a decrease in the concentrations of $SO_2$, CO, and $PM_{10}$, while $O_3$ and NO showed an increase [3]. However, the concentrations remained high with an average mean $PM_{10}$ value of 25.755 $\mu g/m^3$ and daily CO concentrations reaching values above the 1000 $\mu g/m^3$ and showing great temporal and seasonal variability [3]. In the North-Bohemian Brown Coal Basin, the presence of large combustion facilities and opencast brown coal mines affects the air quality, where the concentrations of $PM_{10}$ show a significant increase during the winter period, especially in areas located close to a quarry [4]. In Central Serbia, bulk precipitation samples collected from principal meteorological stations for the period 1998–2004, indicated high value of the ratio ($SO_4^{2-} / NO_3^-$), which was correlated with the air pollution from the coal-fired thermal power plants in the investigated area [5].

In Louisville, Kentucky, USA, $PM_{10}$ concentrations near coal-fired plants, even at levels below 20 $\mu g/m^3$ have been correlated with neurobehavioral disorders in children living in the vicinity of the power plants [6]. In China, coal-fired power plants are among the greatest sources of air pollution. Their spillover effects on public health are substantially contributing to the number of deaths and treatment costs, with the wind speed and direction playing a key role in these impacts [7].

Studies that have investigated the chemical composition and emissions from coal-fired power plants have found high concentrations of toxic heavy metals such as arsenic, chromium, mercury, nickel, cobalt, copper, mercury, iron, manganese, lead, selenium, and zinc [8–12]. These toxic metals are released during the mining and burning of coal, have a great impact on public health, contribute to air and soil pollution, and are disposed into the waterways, polluting the drinking water.

Greece is ranked among the top places for the lignite production in the European Union, Europe, and worldwide. During the last 40 years, lignite mining has experienced a gradual growth following the country's increasing electricity demand. From 1980 to 1990, the lignite production in Greece increased from 22.7 million tons to 49.9 million tons. Based on Chatzitheodoridis et al. [13], the year 2002 recorded the highest production of lignite in the history of Greece (70.3 million tons). However, in the last decade there has been a general decline in lignite production following the reduced demand for lignite power. This is due to the regulatory pressures to lower greenhouse gas emissions, competition from renewable energy sources and from lower natural gas prices, the increased energy imports, as well as the lower overall electricity demand as a result of the economic crisis [14,15].

The major lignite reserves are located in the region of Western Macedonia and in the municipality of Megalopolis, Arcadia (region of Peloponnese). These regions are currently operating the major coal-power plants of the country. In Megalopolis, Peloponnese, the lignite power plant has an initial installed capacity of 850 MW, while Western Macedonia has six lignite steam power plants with a total initial installed capacity of almost 4440 MW generated electricity [16]. Western Macedonia is largely dominated by lignite mining, lignite-fired power plants, and district heating systems, which are a significant part of the country's economy. The environmental pollution from these lignite power plants has been the subject of many studies during the last two decades [12,17–25]. The health impact of air pollution on lignite miners and citizens of the region has also been investigated by Sichletidis et al. [26–28] and Spyratos et al. [29]. Specifically, Sichletidis et al. [26] found that lignite miners in Eordea valley (Western Macedonia) had a high prevalence of atrophic rhinitis and upper respiratory system disorders due to excessive pollution by airborne particles (fly ash) and the high concentrations of chromium, nickel, cobalt, and lead in airborne dust. The detrimental effects of air pollution on the respiratory system of children in Western Macedonia have been investigated by Sichletidis et al. [27]. They found ahigh prevalence of rhinitis and infectious bronchitis in school children in Ptolemaida, which is a highly polluted region, compared with Grevena, which is a less polluted and non-industrialized region. The difference in the prevalence of the above-mentioned infections was about two-fold higher in Ptolemaida than in Grevena. Moreover, Spyratos et al. [29] conducted a 19-year cohort study among children in Ptolemaida and a follow up study of the initial subjects, who were permanent inhabitants of the town, concluding that air pollution correlated with nasal obstruction and chronic nasal symptoms. In general, the exposure to $PM_{10}$ was found to be a risk factor for the development of severe nasal obstruction [28], highlighting the fact that the upper respiratory system is severely affected by the air pollution emitted by lignite mining activities.

The scope of this study is the assessment of the long-term trends of PM concentrations in the Lignite Center of Western Macedonia in the context of the lignite phase-out plan for power generation, which is scheduled to shut down the coal power plants in the country. As we have mentioned, prior studies have established the impact of the coal-fired plants and the mining activities on the air quality of the region. Therefore, in this study, we used data on $PM_{2.5}$ and $PM_{10}$ concentrations from 10 Air Quality Monitoring Stations (AQMS) operated in the region, along with meteorological data (wind speed and direction) in order to determine the long-term trends of PM concentration levels and evaluate the air quality of the region. Moreover, daily PM samples were carried out in two different locations in the region, in the industrial area of the open-cast mines near the village of Pontokomi and in the rural area of the village of Petrana, in order to determine the concentrations of trace elements. It is important to determine the trace element concentrations in PM, because it is often enriched with potentially toxic trace elements. So, in this study, we also evaluate the seasonal characteristics and possible emission sources of the PM-bound trace elements concentrations.

## 2. Materials and Methods

### *2.1. Study Location*

Western Macedonia, located in North-western Greece, is divided into the regional units of Florina, Grevena, Kastoria, and Kozani. One of the major geologic lignite reserves in Greece that is exploited for energy purposes is located at the Kozani–Ptolemaida–Amyntaio–Florina axis in Western Macedonia. It covers an area of 9451 $km^2$ and comprises mostly mountainous and semi- mountainous areas. The area is well-known for its rich natural resources, such as fossil fuels (lignite), ores (asbestos, chromite, marble etc.), and forests (50% of its total land) that form ecosystems defined by rich biodiversity, as well as pastures, while it also has the greatest surface water potential in Greece (approximately 65% of the country) (https://tracer-h2020.eu/west-macedonia-greece-el53/, accessed on 20 September 2022).

As for 2020, four lignite power stations were operating in this region, namely "Agios Demetrios" (1500 MW), "Kardia" (1200 MW), "Ptolemaida" (700 MW), and "Amyntaio" (600 MW) (Government Committee SDAM, 2020). The intensive exploitation of the lignite deposits of Western Macedonia began in 1956 and escalated at a very fast pace, covering for decades most of the electricity consumption in Greece (Figure 1). Between the years 2001 and 2005 the lignite production in Western Macedonia exceeded 55 million tons per year followed by a decline from 50 million tons in 2006 to 45 million tons in 2012. Since 2013 there was a sharp decrease in the lignite production, reaching approximately 10 million tons in 2020 (Figure 1).

In accordance with the Hellenic National Meteorological Services (HNMS), the climate of Western Macedonia is mainly humid subtropical (Cfa), following the Köppen climate classification. Based on the climatic data for Kozani during the period 1955–2010, the monthly mean temperature ranged between 2.3 °C in January and 24.5 °C in July, while the minimum monthly temperature in January was −1.2 °C. The maximum monthly temperatures in July and August reached up to 29.6 °C. Figure 2 shows the average daily precipitation values for the center of the basin. The precipitation values were taken from the climate data storage of the Copernicus climate change service. Lower-than-average concentrations were measured during precipitation events. A detailed analysis of the climatological conditions in the regions of Florina, Kastoria, and Kozani is provided by Zoras et al. [30].

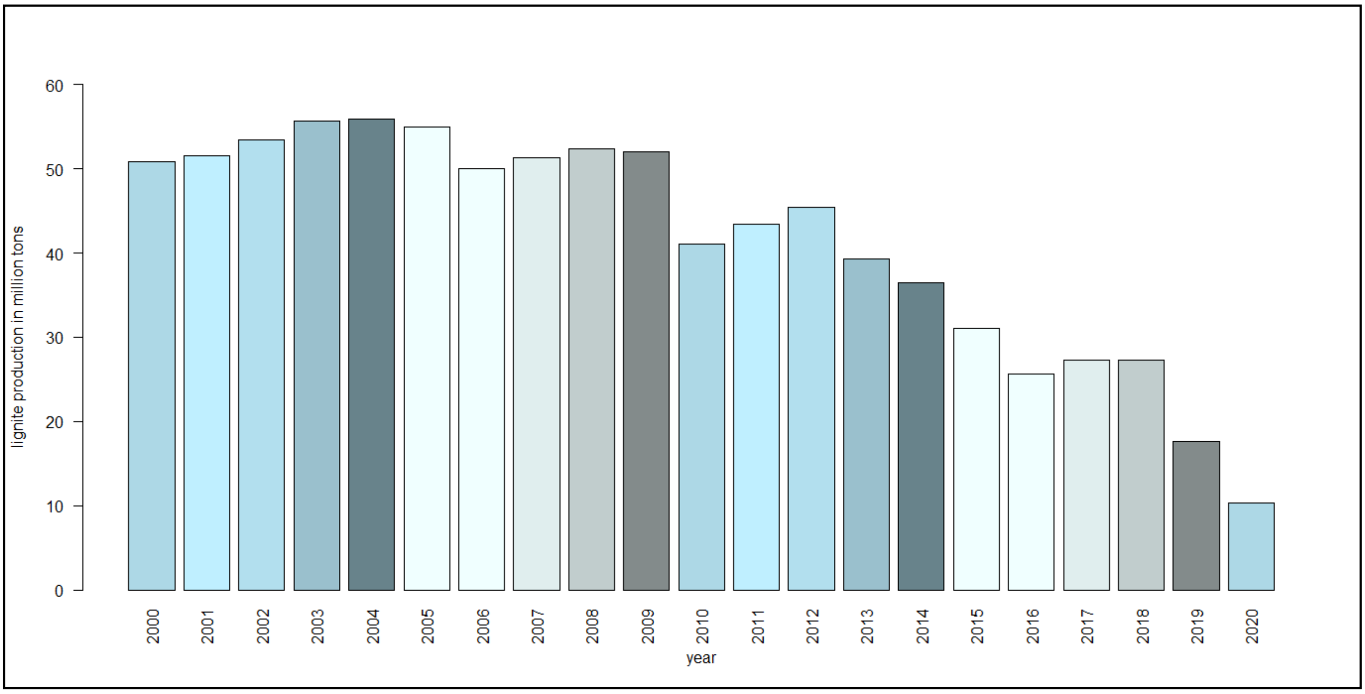



**Figure 1.** Lignite production (per million tons) in Western Macedonia from 2000 to 2020. Each bar in the barplot indicates the lignite production per year.

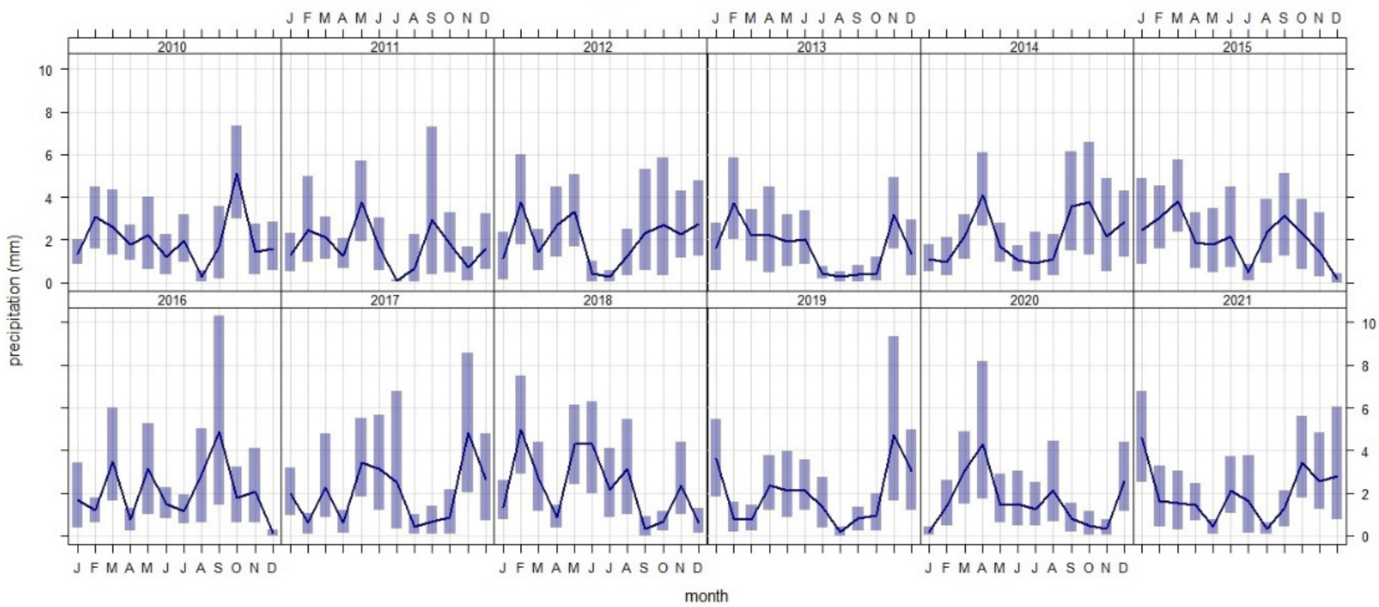



**Figure 2.** Mean monthly precipitation (indicated with blue lines) and 95% CI (confidence interval) in mean (indicated with blue shade bars).

## 2.2. Data

In this study, we used two datasets. The first dataset consists of hourly concentrations of $PM_{10}$ and $PM_{2.5}$ along with meteorological data (wind speed and direction) from 10 Air

Quality Monitoring Stations (AQMS) operated by the Lignite Center of Western Macedonia. The dataset of $PM_{2.5}$ and $PM_{10}$ covers a 12-year period, from 1 January 2010 to 31 December 2021. The measurements are available for all AQMS over the 12-year period, except for the AQMS at Oikismos, where measurements are available for the period from 1 January 2010 to 28 November 2019.

Figure 3 displays the location of stations and Table 1 shows the coordinates and altitude of each station. The second dataset consists of the concentrations of 17 $PM_{2.5}$-bound and 17 $PM_{10}$-bound trace elements (Al, Mn, Fe, Ni, Cu, Zn, Sn, Pb, Si, Mg, Cr, As, Na, K, Ca, Sr, Cd) of 72 samples from 2 locations (36 samples of $PM_{10}$ and 36 samples of $PM_{2.5}$). The sampling was carried out for a period of 12 months (December 2017 to November 2018) for 24 h ambient sampling. The samples were collected in the industrial area of the open-cast mines near the village of Pontokomi and in the rural area of the village of Petrana. These two receptor sites have different characteristics. Specifically, the distance of Pontokomi from the lignite-fired power plant "Kardia" is 2.5 km, while Petrana has a distance of 12.5 km from the lignite-fired power plant "Agios Demetrios".

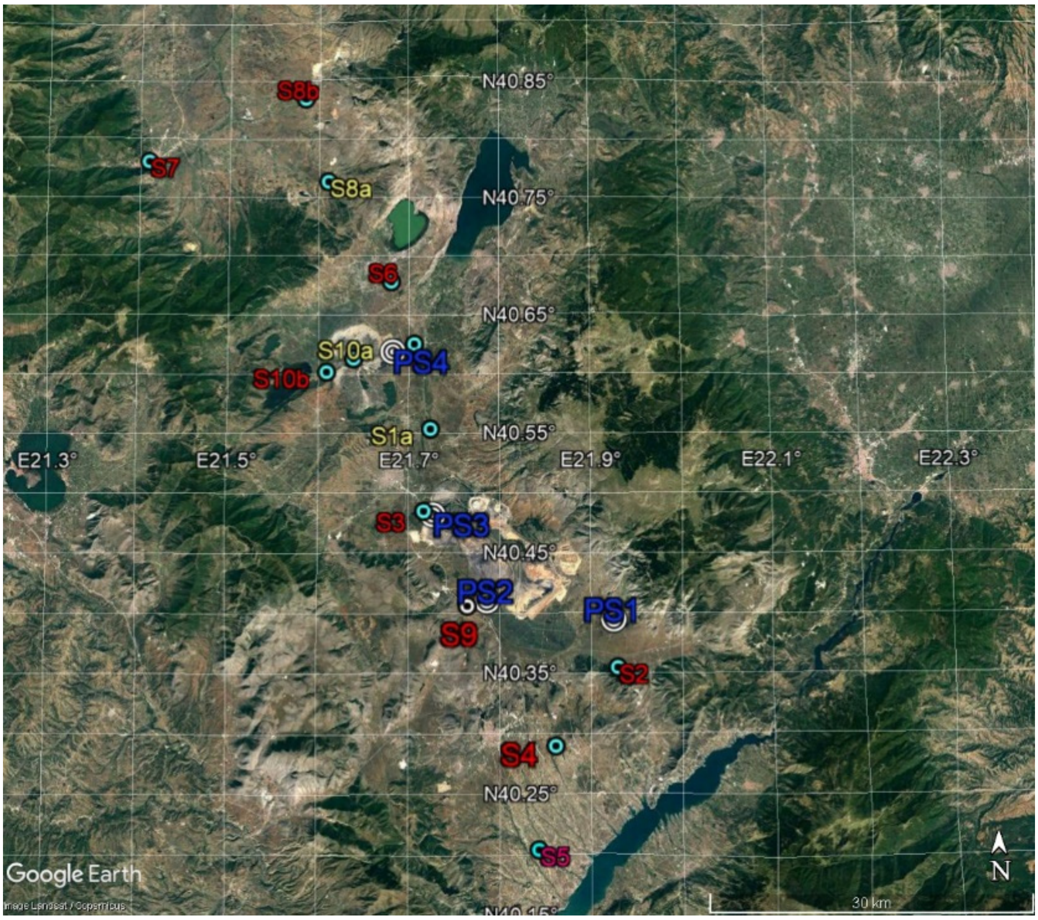



**Figure 3.** Location of stations (S) and power stations (PS) in Western Macedonia.

**Table 1.** Coordinates of stations (S) and power stations (PS).

| Name | Location | Latitude | Longitude | Altitude (m) |
|---|---|---|---|---|
| S1 | Filotas | 40.626056 | 21.707554 | 568 |
| S2 | Koilada | 40.355725 | 21.930784 | 686 |
| S3 | Oikismos | 40.485181 | 21.718224 | 673 |
| S4 | Petrana | 40.290150 | 21.863800 | 614 |
| S5 | Komi | 40.203969 | 21.843391 | 415 |
| S6 | Amyntaio | 40.678970 | 21.681830 | 628 |
| S7 | Florina | 40.782096 | 21.410366 | 659 |
| S8 | Vevi-Meliti | 40.835500 | 21.586800 | 677 |
| S9 | Pontokomi | 40.406530 | 21.768110 | 702 |
| S10 | Anargyroi | 40.602222 | 21.610000 | 611 |
| PS1 | Agios Demetrios | 40.393542 | 21.925377 | 680 |
| PS2 | Kardia | 40.408991 | 21.786542 | 693 |
| PS3 | Ptolemaida | 40.480864 | 21.727385 | 641 |
| PS4 | Amyntaio | 40.618154 | 21.682730 | 665 |

### *2.3. Sampling Techniques and Procedure*

The samples of the second dataset were collected using two low-volume samplers, LVS 3.1 and PNS16T-3.1 (Comde-Derenda GmbH, Stahnsdorf, Germany) with 47 mm glass fiber filters according to the EN12341 standard gravimetric measurement method for the determination of the $PM_{10}$ and $PM_{2.5}$ mass concentration. Analytical determinations of 17 trace elements (Al, Mn, Fe, Ni, Cu, Zn, Sn, Pb, Si, Mg, Cr, As, Na, K, Ca, Sr, and Cd) were performed using the microwave extraction procedure followed by quantitative analysis of graphite furnace atomic absorption spectrometry technique, using a ZEEnit 700—Analytic Jena GmbH (Jena, Germany) combined with Zeeman graphite furnace atomizer (South Holland, The Netherlands). For calculation of the calibration curve, the stock standard solution of the target element (Merck, Altoona, PA, USA) was used with various concentrations. The determination of the background contamination of metals was carried out by subtracting the field blank values from the concentrations. It is noted that the field blank values were minor, as they hovered below or around the detection limits specified by the method. Measurements were corrected accordingly. The limit of detection (LOD) values were calculated as 3 times the standard deviation, and the limit of quantitation (LOQ) values were calculated as 10 times the standard deviation of the results obtained from the analysis of 10 ppb stock standard solutions. The LOD values ($\mu g\ L^{-1}$) were Al (21.0), Mn (0.4), Fe (8), Ni (1.3), Cu (1.2), Zn (0.6), Sn (20.0), Pb (0.2), Si (5.0), Mg (0.2), Ti (40.0), V (50.0), Cr (0.1), As (20.0), Na (0.3), K (0.6), Ca (0.5), Sr (1.3), Cd (1.3).

A detailed description of the sampling techniques and procedure can also be found in the study by Evagelopoulos et al. (2022) [24].

### *2.4. Methodology*

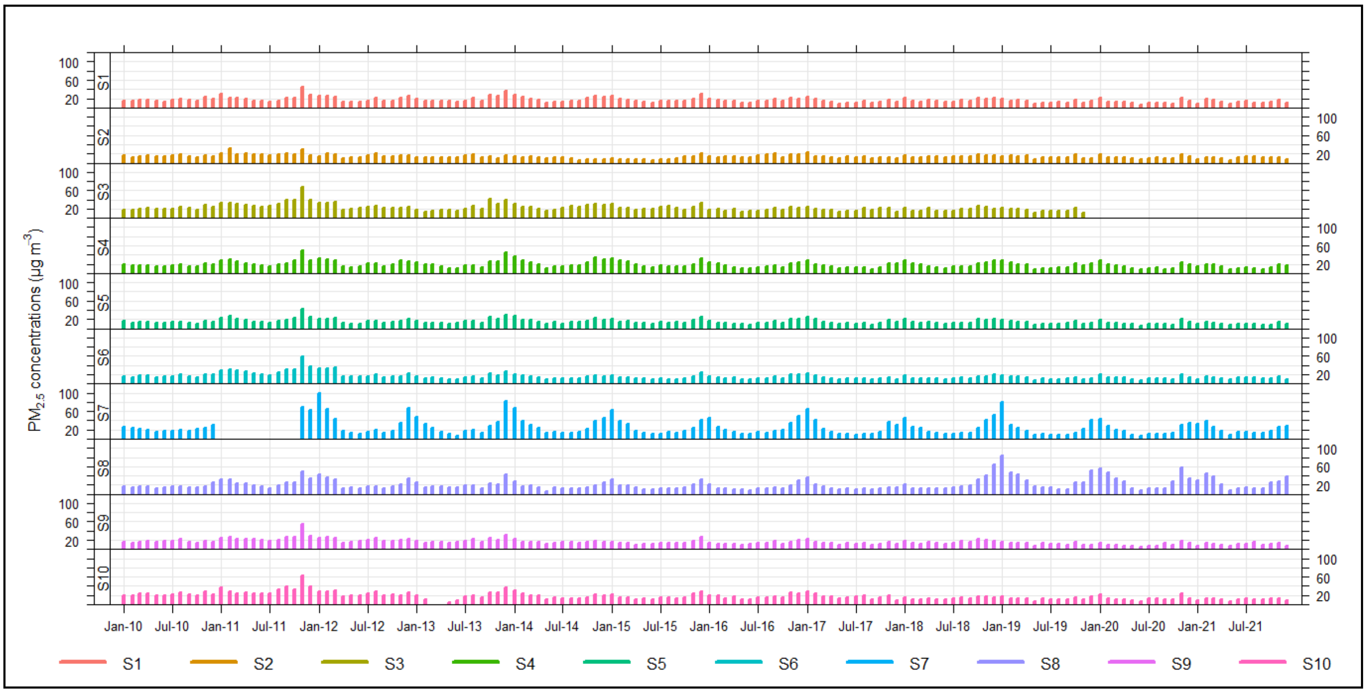
In this study, the analysis was performed in the computer software "R" by using the package "openair" (version: 12 November 2019) [31]. The package consists of many tools for importing and manipulating data, and can be used to undertake a wide range of analyses to enhance the understanding of air pollution data. Firstly, we analyzed the daily $PM_{2.5}$ and $PM_{10}$ concentrations at the 10 AQMS in order to find the mean monthly concentrations of $PM_{2.5}$ and $PM_{10}$ over the 12-year period. Then, based on the daily values of $PM_{2.5}$ and $PM_{10}$, the daily averages of the $PM_{2.5}/PM_{10}$ ratios were calculated. Secondly, we used the pollution roses to assess the dependence of $PM_{2.5}$ and $PM_{10}$ concentrations on wind speed and direction and find the potential source of air pollution in the region.

Thirdly, the time series of $PM_{2.5}$ and $PM_{10}$ concentrations were analyzed with the Theil–Sen estimator in order to estimate their long-term trends during the study period [31]. Lastly, we divided the dataset of $PM_{2.5}$-bound and $PM_{10}$-bound trace elements into a cold period (from 15 October to 14 March) and a warm period (from 15 March to 14 October) in order to compare the elemental constitutes of PM between the two sampling locations and between seasons.

## 3. Results and Discussion

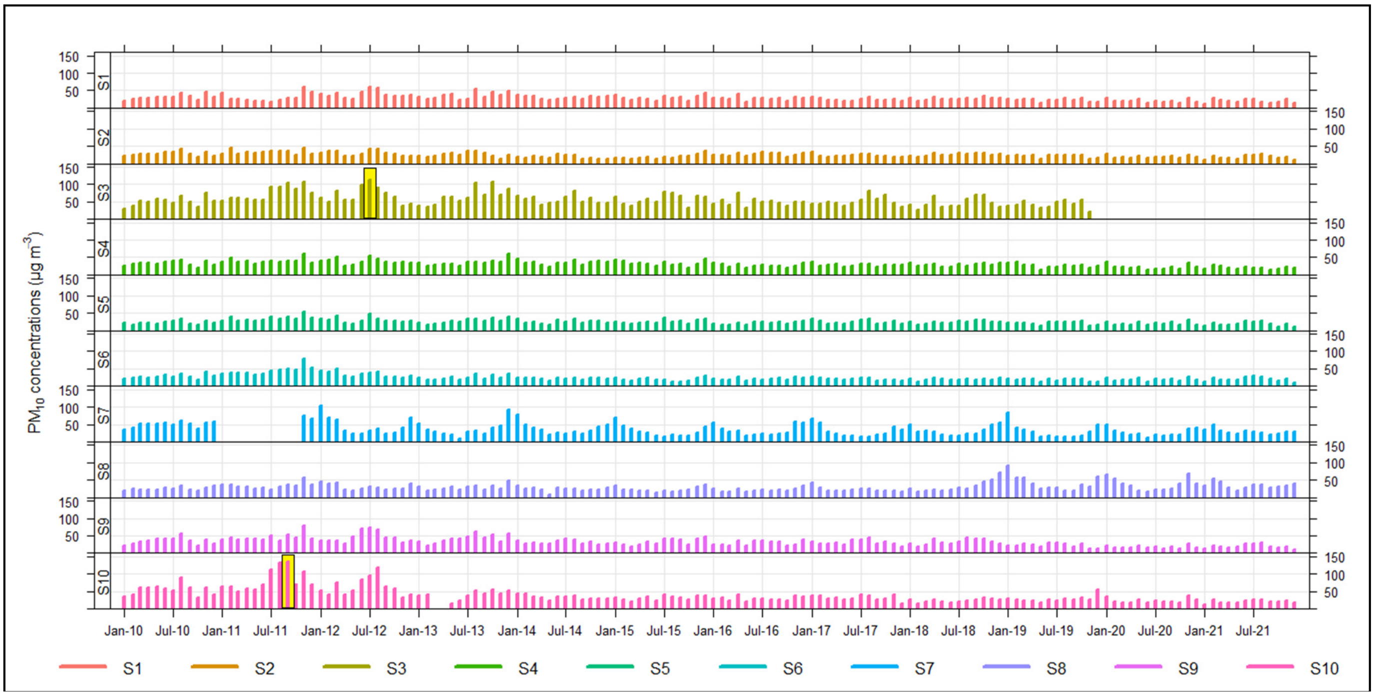
### *3.1. $PM_{2.5}$ and $PM_{10}$ Concentrations over a 12-Year Period in Western Macedonia*

Figure 4a,b show the monthly mean concentrations time series of $PM_{2.5}$ and $PM_{10}$ measured during the study period (2010–2021) at all AQMS of the region. At all AQMS there are clear seasonality patterns for $PM_{2.5}$ concentrations, with high levels during the cold period and low levels during the warm period. Besides the operation of lignite-fired power plants and mining activities, major contributors to high $PM_{2.5}$ concentrations during the winter period are the heating systems, residential wood combustion, and biomass burning. The AQMS are located in the broader area of the lignite basin, so they capture various sources of PM release (e.g., traffic emissions), not necessarily related to the mining and power generation [15]. In contrast to $PM_{2.5}$ concentrations, the monthly mean values of $PM_{10}$ concentrations reached the highest values during the winter months at the AQMS of Florina (S7) and Vevi-Meliti (S8), while the highest monthly mean concentrations of $PM_{10}$ were registered during the summer or autumn at the other AQMS. In general, the highest monthly mean values were calculated in August at the AQMS in Oikismos (S3) and Anargyroi (S10) based on the monthly mean values over the 12-year period. However, the highest mean monthly $PM_{10}$ concentrations were registered at the AQMS of Anargyroi (S10) in September 2011 (mean monthly value: 134 $\mu g/m^3$) and at the AQMS in Oikismos (S3) in July 2012 (mean monthly value: 112 $\mu g/m^3$).




(**a**)

**Figure 4.** *Cont.*




(**b**)

**Figure 4.** Monthly mean concentrations time series of (**a**) $PM_{2.5}$ and (**b**) $PM_{10}$ concentrations (μg/m$^3$) measured during the study period (2010–2021) at all AQMS of the region. In (**b**), the months with the highest monthly mean concentrations of $PM_{10}$ in AQMS S3 and S10 are indicated with yellow.

Previous studies have also concluded that the $PM_{10}$ concentrations in the region under study are higher during the warm period. For instance, the seasonal variations of concentration measurements of $PM_{10}$ in the commercial center of Kozani were analyzed by Triantafyllou et al. [32] for a 5-year period (1996–2000), who concluded that the mean and the median values are higher in the warm period (June–September) than in the cold period (November–February) and the transient period (March–May and October). This could be explained by the fact that there is a difference in the emission and deposition of pollutants among the months due to local meteorological factors. The precipitation, which is usually the main removal process of the atmospheric pollutants, is higher during the transient and cold period, which lowers their concentrations. Precipitation scavenging, also known as wet deposition as well as rainout and washout mechanisms, which are forms of scavenging, can effectively remove air particles from the atmosphere. Especially, large particles, such as $PM_{10}$, are most efficiently removed by washout, which is below-cloud capture of air pollutants by falling raindrops. Based on the climatic data for the period 1955–2010 in the meteorological station of Kozani of the HNMS (Hellenic Meteorological Services) weather network, the lowest monthly mean precipitation height (27.8 mm) was recorded in August. Based on the same climatic data, during the cold period and the transitional months, the precipitation height is the highest, with a high precipitation height (56.1 mm) in November as well as in May (54.7 mm).

Moreover, the high concentrations of coarser PM during the summer months are associated with the atmospheric conditions and prevailing strong winds, which favor the resuspension of soil and dust from dry surfaces [18,22,32]. Therefore, it seems that wind-induced resuspension is one of the most significant secondary sources of $PM_{10}$ during the warm period in the region [18,22,32].

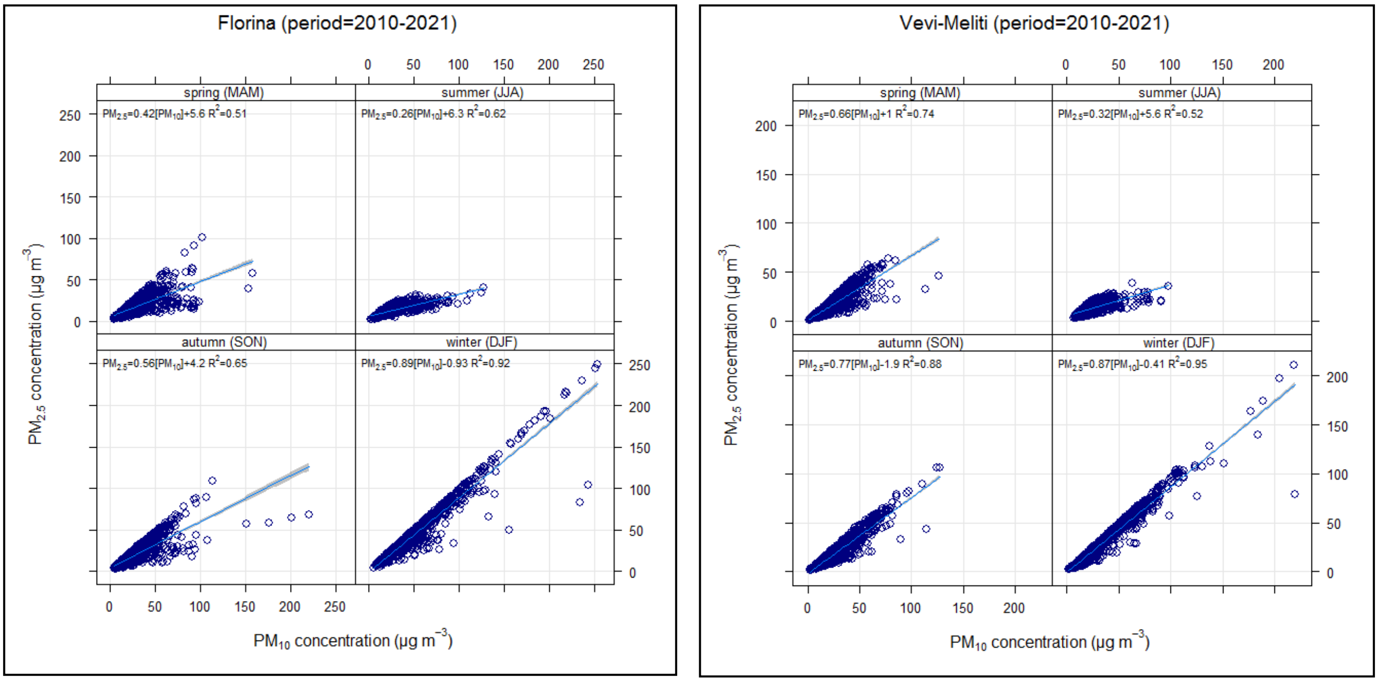
With reference to the averaged 24-h value over the study period, the AQMS in Florina, Vevi-Meliti, and Oikismos exhibited the highest mean value of $PM_{2.5}$. The mean values of $PM_{2.5}$ were 24.51 μg/m$^3$, 20.76 μg/m$^3$ and 22.50 μg/m$^3$ in Florina, Vevi-Meliti, and Oikismos. The mean values of $PM_{2.5}$ varied between 13.54 μg/m$^3$ in Koilada and 24.51 μg/m$^3$

in Florina. Similarly, the AQMS in Oikismos exhibited the highest concentrations of $PM_{10}$ with a mean value for the study period at 56.79 μg/m$^3$, while high values of $PM_{10}$ exhibited at the AQMS of Anargyroi with a mean value of 37.85 μg/m$^3$. The mean values of $PM_{10}$ varied from the approximate values of 24 μg/m$^3$ in Koilada and Amyntaio to 56.79 μg/m$^3$ in Oikismos.

In addition, the average ratios of $PM_{2.5}$ to $PM_{10}$ were assessed in order to characterize the underlying atmospheric and anthropogenic processes affecting PM concentrations within the local environment. Based on scientific literature, high ratios of $PM_{2.5}/PM_{10}$ indicate that anthropogenic sources contribute to particle pollution and small ratios indicate the involvement of coarse particles, which might be related to natural sources [33].

In our study, average $PM_{2.5}/PM_{10}$ ratios during the study period range between 0.48 in Oikismos and 0.70 in Florina and Vevi-Meliti. For Florina and Vevi-Meliti, the daily $PM_{2.5}/PM_{10}$ ratios were found to be from 0.16 to 1.0 and 0.21 to 1.0, respectively. During the study period, the daily average ratios of $PM_{2.5}/PM_{10}$ were above 0.9 for the 19% of the days in Florina while in Vevi-Meliti they were above 0.9 for the 16% of the days. These percentages were two-fold higher in Florina and Vevi-Meliti compared with the other sites where high daily ratios of $PM_{2.5}/PM_{10}$ were found for a lesser number of days. This indicates that fine particles comprised a large fraction in $PM_{10}$ in Florina and Vevi-Meliti. During the winter months, $PM_{10}$ concentration levels were well correlated with $PM_{2.5}$ concentration levels, with correlation coefficients ($R^2$) of 0.92 for Florina and 0.95 for Vevi-Meliti for the period 2010–2021 (Figure 5a,b). The very good correlation factor ($R^2$) equal to 0.95 suggests that $PM_{2.5}$ and $PM_{10}$ came from similar emission sources of particulate matter sources [34]. During the winter months, the concentrations of $PM_{10}$ and $PM_{2.5}$ reached up to 250 μg/m$^3$ in Florina and 200 μg/m$^3$ in Vevi-Meliti. In addition, high levels of PM were indicated in autumn, reaching up to 100 μg/m$^3$ in both sites. However, in spring and summer, the PM levels in both sites were below 100 μg/m$^3$ and 50 μg/m$^3$, respectively. Obviously, the increased PM levels during wintertime were mostly due to domestic heating and the increased electricity demand from the lignite-fired power plants. It is worth mentioning that there are district heating networks in Western Macedonia, which utilize the heat waste from lignite combustion in steam-electric power stations Public Power Corporation for covering the heating and hot water needs of the buildings (households, offices, and commercial). These networks are currently operated in Kozani, Ptolemaida, and the greater area of Amyntaio. District heating has the advantage of higher overall system efficiency and lower environmental impact. However, in Florina and Vevi-Meliti, district heating networks are currently not available and conventional/traditional heating systems are used instead.

It is well known that the air pollutants emitted from the power generation units and from the mining and transportation of lignite from the mines to the units have caused significant air pollution issues in the wider region of Western Macedonia. Based on studies and reports conducted with reference to this area, the mean measured $PM_{10}$ concentrations at the different sites in the periphery of the mine areas ranged from 38 μg/m$^3$ to 72 μg/m$^3$, during the cold and the warm periods of the year (November–December 2011 and August–September 2012) [22]. In addition, Valavanidis et al. [35] mentioned that the levels of $PM_{10}$ concentrations often exceeded the daily limit of 50 μg/m$^3$ at the air quality monitoring stations of Public Power Corporation in the lignite region of Western Macedonia, especially during the summer months. Most exceedances of the above-mentioned daily limit occurred at the station of Oikismos, where our analysis revealed similar results.




(**a**) (**b**)

**Figure 5.** Linear regression equations for each season yielding a best-fitting straight line for $PM_{2.5}$ to $PM_{10}$ at the AQMS of (**a**) Florina and (**b**) Vevi-Meliti for the study period (2010–2021).

It is evident that PM levels are associated with wind speed and direction, as well as PM, which can be transported by the wind, affecting neighboring areas. In Figure 6a,b, the monthly pollution increases for $PM_{10}$ concentrations in Oikismos during two different periods are shown. From 1 January 2010 to 9 November 2014, the air quality station was at a distance of 650 m from the lignite power plant of Ptolemaida, while for the period from 10 November 2014 to 13 November 2019 after a fire in a plant's unit, the lignite power plant shut down. During the first study period, the higher concentrations of $PM_{10}$ were indicated during the summer months and the prevailing winds were mainly NNE-NE (Figure 6a). During the second study period, the highest $PM_{10}$ concentrations were observed under the influence of SSE-SE and SE-ESE wind directions, while low $PM_{10}$ concentrations were observed for northerly winds (Figure 6b).

Another important process affecting the seasonal variations of the air pollutants is the resuspension, in which particles in soil and dust are blown by the wind up from the soil surface. During the warm season, the drying up of surfaces due to the scarcity of precipitation and low humidity levels exacerbate the processes of particle resuspension, especially under the influence of strong winds. In particular, in mining areas such as in the area under study, the soil cover structures of mining quarries and the tailings after the mining process, the resuspension mechanism was favored under the influence of wind and turbulent flow. Triantafyllou et al. [20] highlights the fact that an important source of inhalable particulates in the Kozani–Ptolemais Basin is the dust generated from mining operations (e.g., excavation), the transport by uncovered trucks, the deposition of lignite and fly ash, and the resuspension because of strong winds. These findings highlight the dependence of air pollutants on meteorological conditions. In addition, the seasonal variation of PM is correlated with the seasonal variation in power demand. It is worth mentioning that there is an increase in electric power demand during the summer period compared with spring, resulting in high emission levels of air pollutants [19,20].

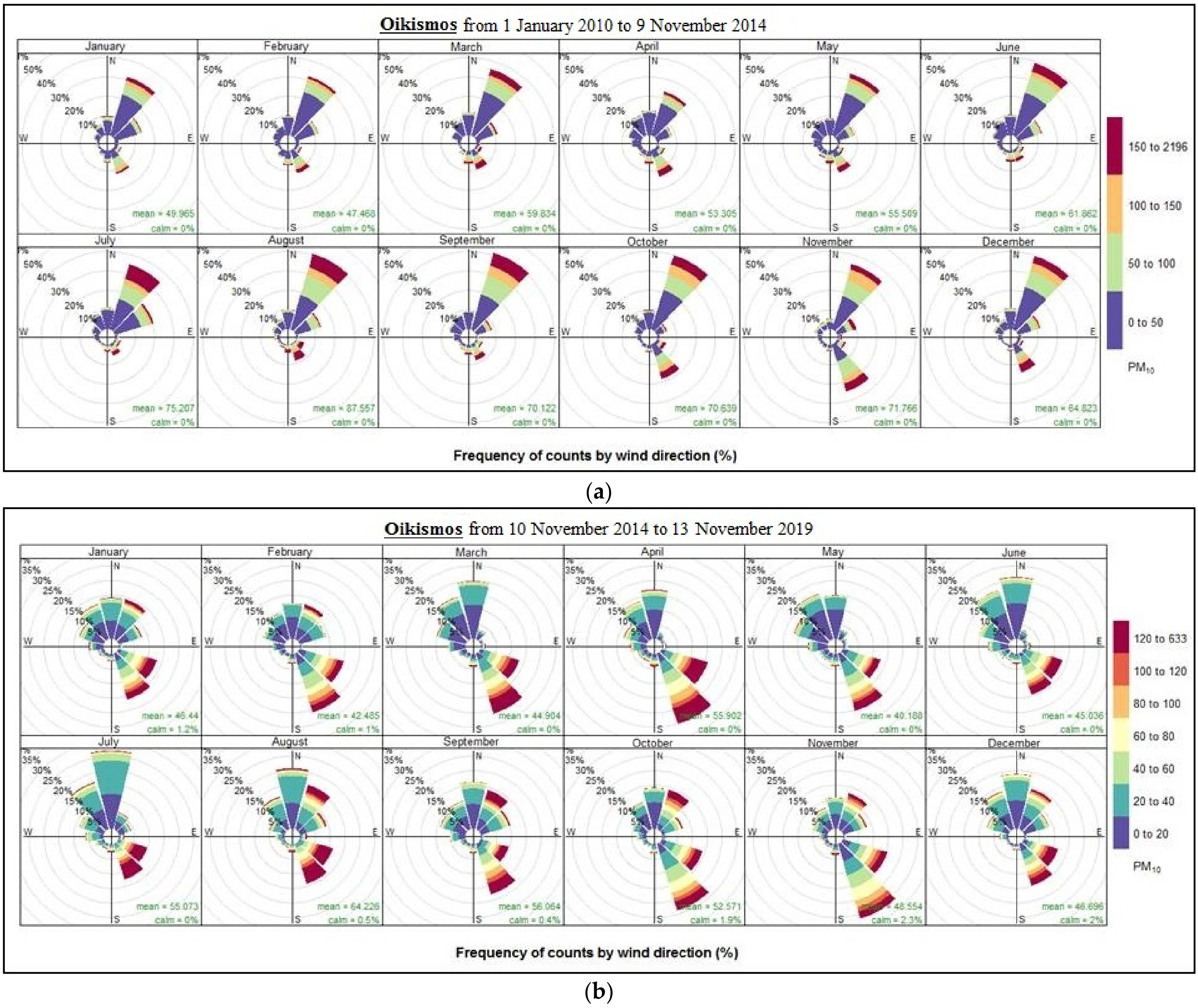



**Figure 6.** Monthly pollution increases for $PM_{10}$ concentrations at Oikismos AQMS for (**a**) the period from 1 January 2010 to 9 November 2014 and (**b**) the period from 10 November 2014 to 13 November 2019. The concentrations of PM are shown by the color scale legend.

Triantafyllou et al. [36] quantified the contribution of five lignite power plants and the opencast lignite mining, as well as the urban activities in the industrial axis, to the particulate pollution of the region. They found that the highest contribution of the power plants to $PM_{10}$ concentrations was found for Koilada and Pontokomi, which are located close to lignite power plants, while the lowest contribution was found for Petrana, which is an agriculture area at a distance of 12.5 km from the power plant "Agios Dimitrios".

With reference to meteorological conditions resulting in extremely high $PM_{10}$ concentrations, Triantafyllou et al. [32] identified the synoptic weather conditions favoring the air pollution episodes. Particularly, these episodes during the winter period are associated with a high-pressure system that covers the central and south Europe and stable/stagnant conditions and favors the accumulation of pollutants. In addition, they are correlated with high wind speed conditions, which result in dust resuspension, and conditions favoring the long-range dust transport from the Saharan desert. Moreover, elevated temperature inversions leads to pure dispersion conditions of the air pollutants.

Furthermore, Triantafyllou [37] investigated the prevailing meteorological conditions when $PM_{10}$ concentrations were greater than or equal to 150 $\mu g/m^3$ during the period

1991–1994 in Kozani at a distance of 8 km from “Kardia” lignite power station. The analysis revealed that the highest concentrations were associated with stagnant conditions, while the local circulation in the Aliakmon valley was found to be associated with the accumulation of pollutants. On the other hand, high wind speed resulted in dust resuspension from the ground to the air and therefore the transport of air pollutants was favored from neighboring sources [37]. With reference to the local circulation of the Aliakmon river valley, it should be mentioned that the basin is characterized by topographic complexity and a variety of physico-geographic characteristics, which are expected to induce local circulation patterns. In addition, the channeling of the regional or synoptic circulation through the gaps connecting the basin with the rest of the mainland of Northern Greece has a substantial role in the formation of the air flow patterns in the area affecting the dispersion of the air pollutants [38].

Karagiannidis et al. [39] analyzed the meteorological conditions that contributed to a prolonged particulate matter air pollution episode, which occurred in the terrain basin of Amyntaio–Ptolemais–Kozani in November 2009. From the 17 November to 20 November 2009, Southern Europe was dominated by an Omega block and therefore the surface wind field in the area was weak. The synoptic meteorological conditions in the area were associated with the evolution of the Omega block, which gradually transformed to a high-over-low pattern. As the surface wind remained weak, $PM_{10}$ and $PM_{2.5}$ concentrations increased, and when the wind field strengthened temporarily, the PM in air quality stations decreased [39].

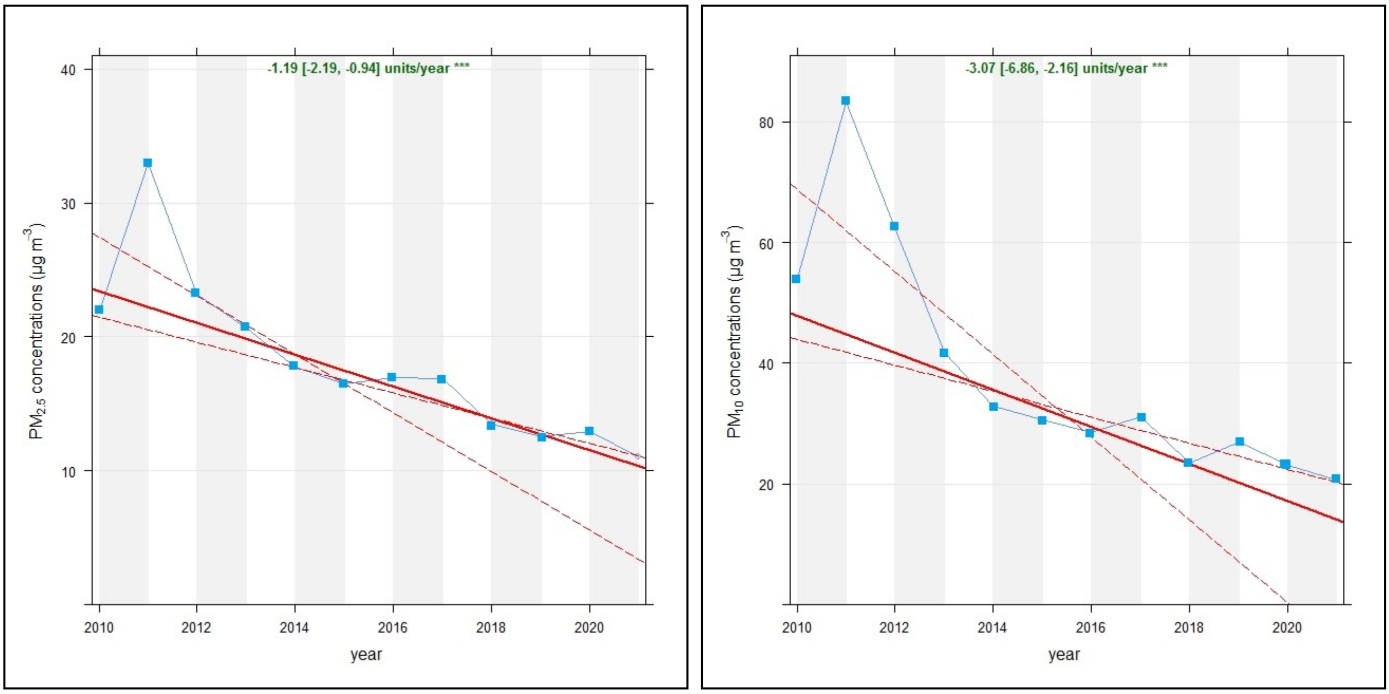
### *3.2. $PM_{2.5}$ and $PM_{10}$ Concentrations Trends at Petrana and Pontokomi AQMS*

Table 2 shows the Theil–Sen slope estimator for the trends of annual mean $PM_{2.5}$ and $PM_{10}$ concentrations along with the 95% confidence intervals of the slope at all AQMS. We used the option to deseasonalize the data first in order to provide a clearer indication of the overall trend on a monthly basis. The data are deseasonalized using the “stl” function (“stl” is the acronym for Seasonal and Trend decomposition using Loess). All Theil–Sen slopes show a decrease of $PM_{2.5}$ and $PM_{10}$ concentrations at all AQMS, besides Meliti AQMS. This could be due to the relocation of the AQMS in the year 2018. For example, the application of the Theil–Sen estimator to the $PM_{2.5}$ and $PM_{10}$ concentration trends at Arargyroi AQMS provide a slope of −1.19 [−2.19, −0.94] units/year and −3.07 [−6.86, −2.16] units/year, respectively. These trends for Arargyroi AQMS are also shown in Figure 7a,b. The plots show the deseasonalized annual mean concentrations of $PM_{2.5}$ and $PM_{10}$.

**Table 2.** Theil–Sen slope and 95% confidence intervals of the trend in average annual $PM_{2.5}$ and $PM_{10}$ concentrations ($\mu g/m^3$) at all AQMS of the region.

| AQMS Code | AQMS Name | Theil–Sen for $PM_{2.5}$ (Units/Year) | Theil–Sen for $PM_{10}$ (Units/Year) |
|---|---|---|---|
| S1 | Filotas | −0.74 [−1.02, −0.35] ** | −1.56 [−2.32, −0.82] *** |
| S2 | Koilada | −0.42 [−0.94, +0.01] $^+$ | −1.08 [−1.46, −0.04] $^+$ |
| S3 | Oikismos | −1.14 [−2.56, −0.25] * | −3.72 [−4.53, −0.32] * |
| S4 | Petrana | −0.82 [−1.16, −0.13] * | −1.8 [−2.11, −0.95] *** |
| S5 | Komi | −0.65 [−1.03, −0.03] * | −1.13 [−2.00, −0.16] * |
| S6 | Amyntaio | −0.51 [−1.75, −0.25] *** | −0.99 [−2.86, −0.26] * |
| S7 | Florina | −1.08 [−3.26, +0.06] $^+$ | −1.99 [−3.85, −0.87] *** |
| S8 | Meliti | +0.45 [−1.69, +1.6] | + 0.52 [−1.25, +1.85] |
| S9 | Pontokomi | −0.90 [−1.78, −0.5] *** | −2.4 [−3.28, −0.65] *** |
| S10 | Anargyroi | −1.19 [−2.19, −0.94] *** | −3.07 [−6.86, −2.16] *** |

Note: The symbols shown next to each trend estimate relate to how statistically significant the trend estimate is: $p < 0.001$ = ***, $p < 0.01$ = **, $p < 0.05$ = * and $p < 0.1$ = $^+$.



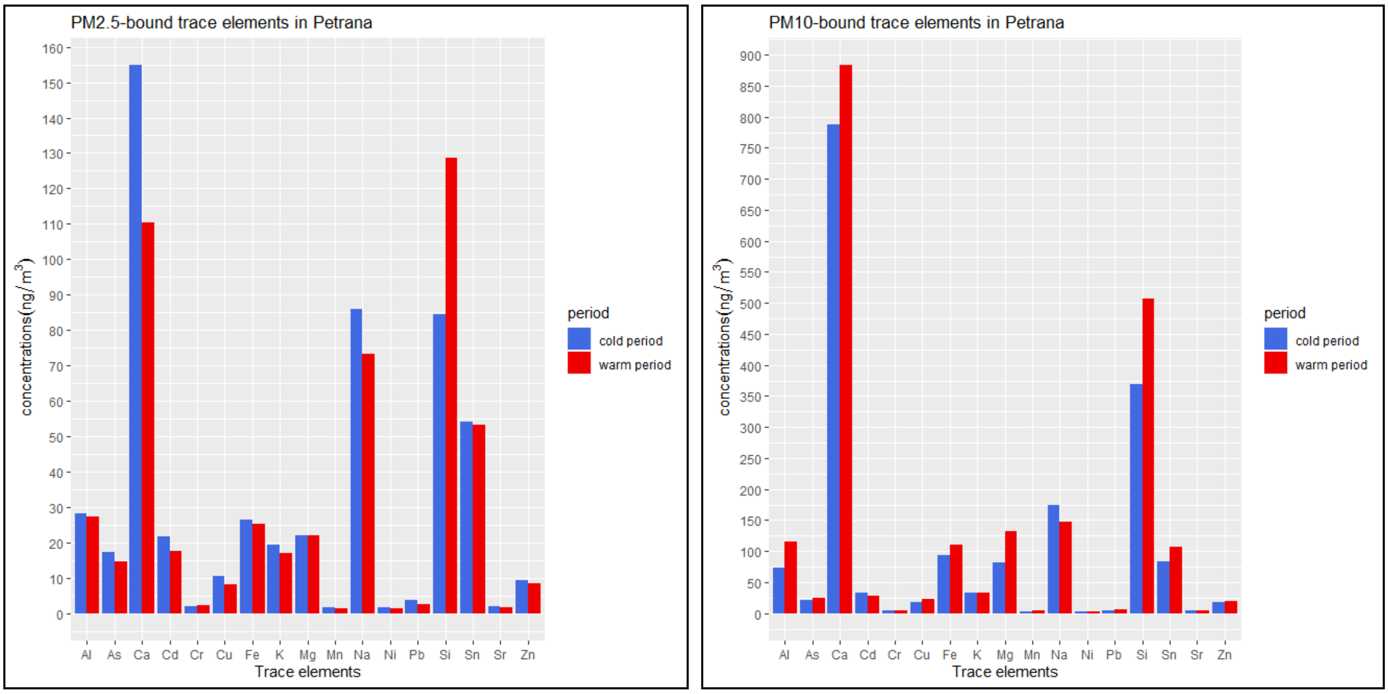

(**a**) (**b**)

**Figure 7.** Trends in (**a**) $PM_{2.5}$ and (**b**) $PM_{10}$ at Anargyroi AQMS. The plots show the deseasonalized annual mean concentrations of (**a**) $PM_{2.5}$ and (**b**) $PM_{10}$, with blue lines and squares. The solid red line shows the trend estimate and the dashed red lines show the 95% confidence intervals for the trend. The overall trend is shown at the top along with the 95% confidence intervals. The *** show that the trend is significant to the 0.001 level.

### *3.3. PM Sampling at Petrana (S4) and Pontokomi (S9)*

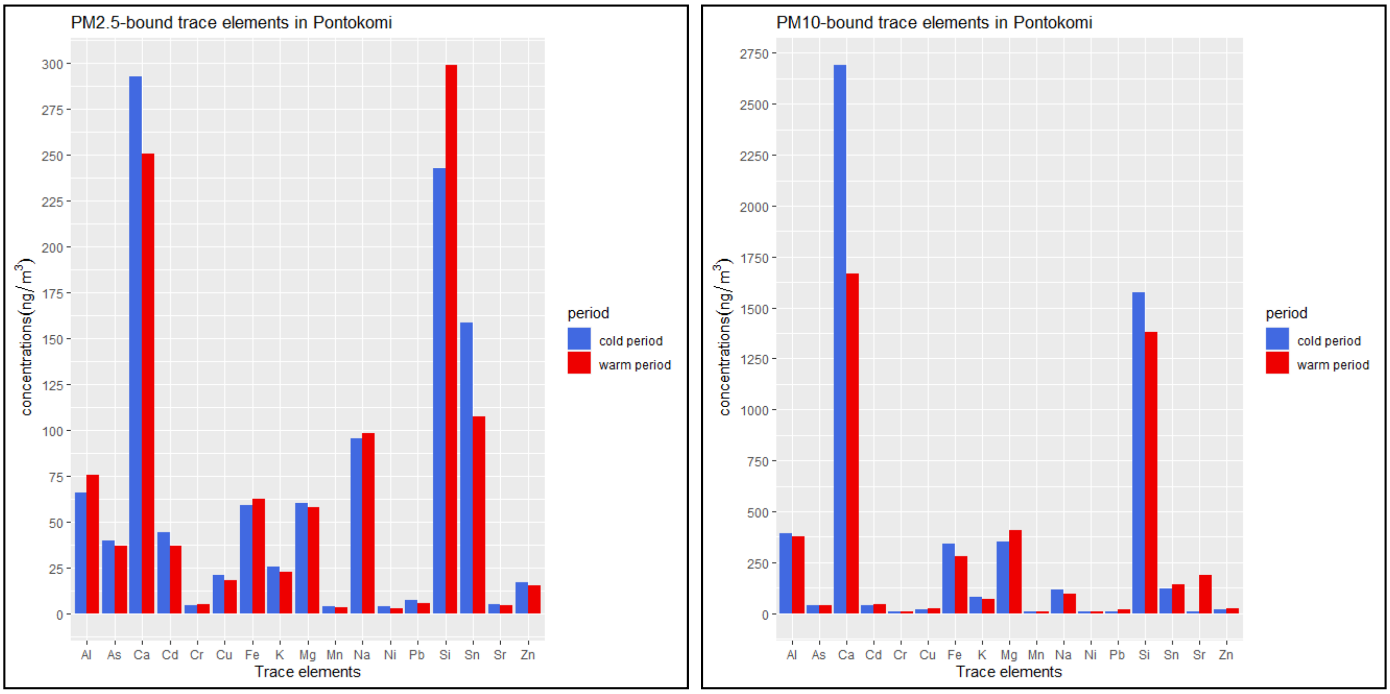
The concentrations of 17 $PM_{2.5}$-bound and 17 $PM_{10}$-bound trace elements (Al, Mn, Fe, Ni, Cu, Zn, Sn, Pb, Si, Mg, Cr, As, Na, K, Ca, Sr, and Cd) were measured during a sampling period that lasted for one year (from December 2017 to November 2018) in two sampling locations, Petrana and Pontokomi. There was a total of 144 samples, which were collected monthly with more than 2 samplings per month. These samplings were divided into a cold period, which lasted from 15 October to 14 March, and warm period, which lasted from 15 March to 14 October.

The mean concentrations of $PM_{2.5}$-bound and $PM_{10}$-bound trace elements exhibit differences between the sampling locations (Figures 8 and 9). At Pontokomi, the concentrations of trace elements are considerably higher compared to Petrana. The difference in the concentration levels of trace elements could be attributed to the emission sources of the two locations, suggesting the influence of the mining activities at Pontokomi. As we have mentioned, Pontokomi is located near the open-cast mines. Despite the differences in seasonal concentrations, the elemental profiles of both $PM_{2.5}$ and $PM_{10}$ were quite similar. At both locations, the most abundant elements with the highest concentrations are Ca and Si, followed by Na, Fe, Al, Mg, and Sn, with relative differences between the sampling locations and between the warm and cold period.




(**a**) (**b**)

**Figure 8.** Concentrations of (**a**) $PM_{2.5}$-bound and (**b**) $PM_{10}$-bound trace elements at Petrana during the warm and cold period of the year.




(**a**) (**b**)

**Figure 9.** Concentrations of (**a**) $PM_{2.5}$-bound and (**b**) $PM_{10}$-bound trace elements at Pontokomi during the warm and cold period of the year.

As illustrated in Figure 10a,b for Pontokomi, the trace elements Ca and Si contribute to 46% and 27% in $PM_{10}$ in the cold period, and 35% and 29% in $PM_{10}$ in the warm period, respectively. Similarly, the contribution of Ca and Si in $PM_{2.5}$ is 26% and 21% in the cold period, and 23% and 27% in the warm period (Figure 10c,d). Similar profiles are also indicated in Figure 11a–d for Petrana. However, Na seems to contribute with a higher percentage in the elemental profiles of both $PM_{10}$ and $PM_{2.5}$ at Petrana compared with Pontokomi (Figure 11a–d). In $PM_{10}$, the trace elements Ca and Si contribute to 44% and 20% in the cold period, and 41% and 24% in the warm period, respectively (Figure 11a,b). In $PM_{2.5}$, the contribution of Ca and Si is 28% and 15% in the cold period, and 21% and 25% in the warm period (Figure 11c,d). Moreover, K, Cd, Zn, and Cu have a relative minor contribution in elemental profiles while Mn, Pb, Cr, Ni, and Sr are the least abundant elements at both locations (Figures 10a–d and 11a–d).

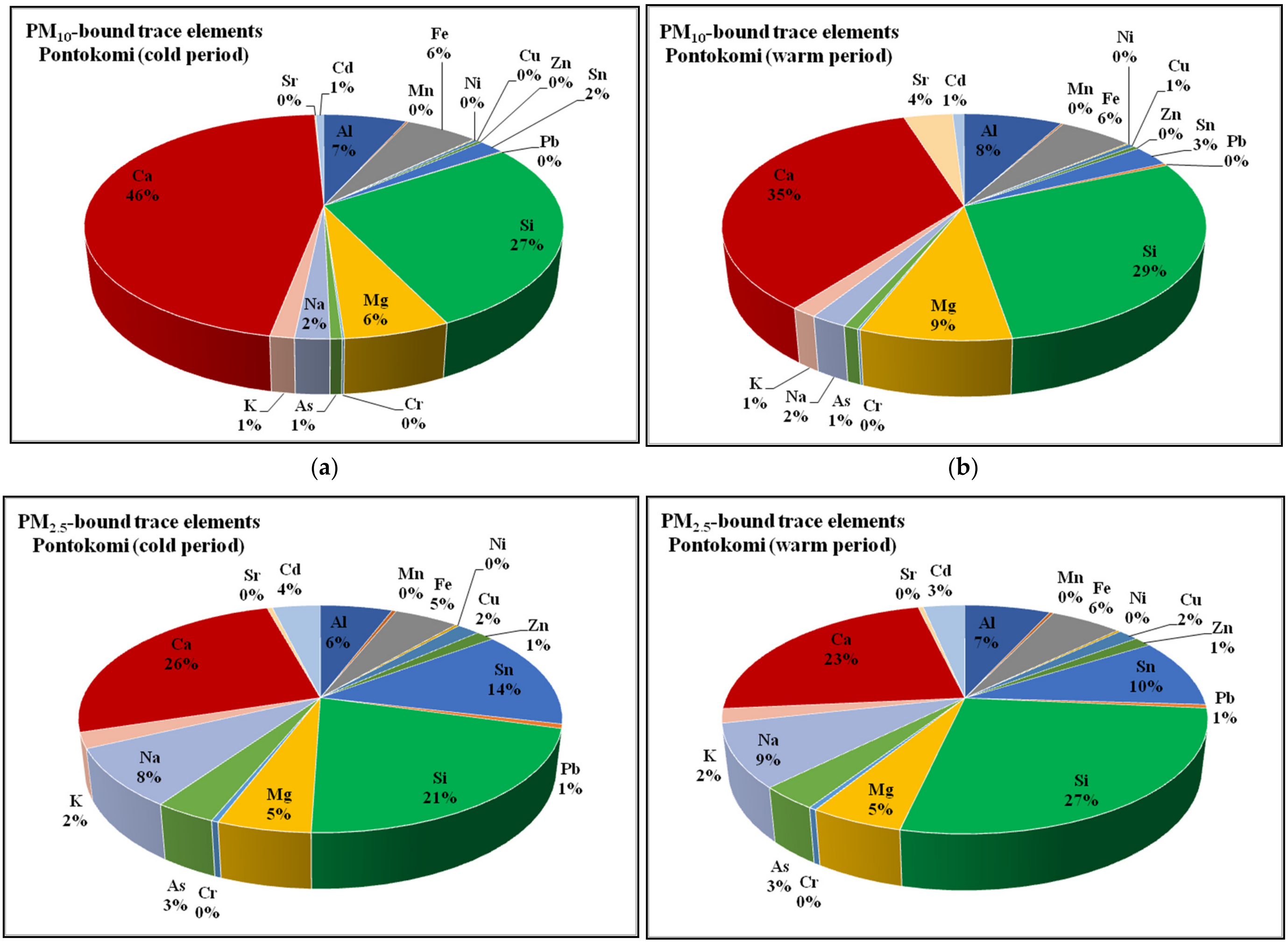



**Figure 10.** Pie charts for the percentage contribution of trace elements to (**a**) $PM_{10}$ in the cold period, (**b**) $PM_{10}$ in the warm period, (**c**) $PM_{2.5}$ in the cold period and (**d**) $PM_{2.5}$ in the warm period at Pontokomi.

Generally, Na, Mg, Al, Si, Cl, K, Ca, and Fe have a natural, crustal, and/or geogenic origin. Although the source of Si-rich particles is primarily associated with soil, they may have a natural origin (resuspension of soil dust and earth's crust) or anthropogenic origin (coal burning and construction activities). Similarly, Ca-rich particles may also have natural and/or anthropogenic-like resuspension of dust and windblown dust or crustal material from paved and unpaved roads as well as construction activities [40]. Previous studies have also found that in the lignite basin of Western Macedonia the most common trace elements are Ca and Si [18,22], while the differences in the relative abundances of Ca and Si in the geological profiles are due to the differences in the distribution of geological formations in the basin [22].

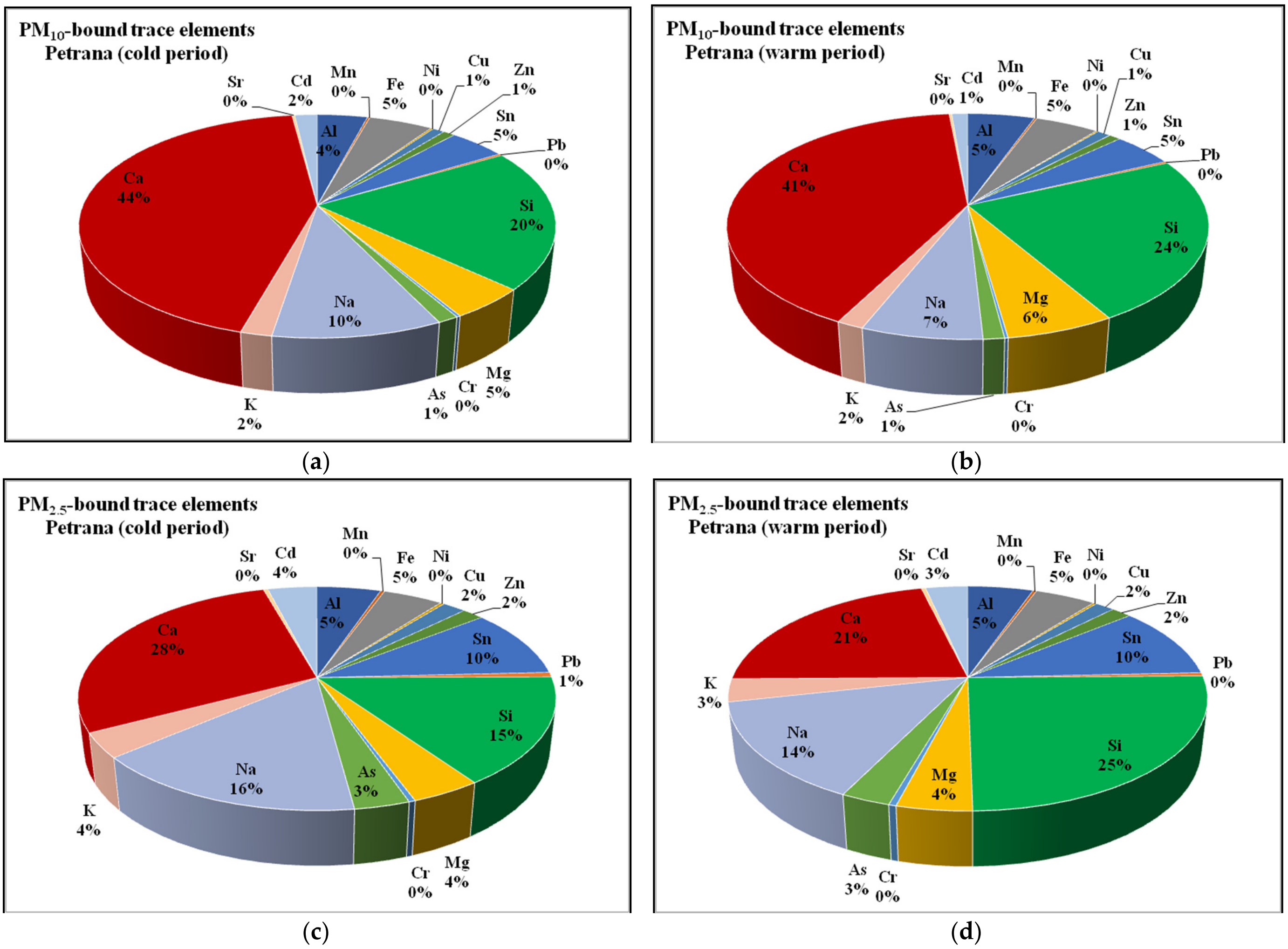



**Figure 11.** Pie charts for percent contribution of trace elements to (**a**) $PM_{10}$ in the cold period, (**b**) $PM_{10}$ in the warm period, (**c**) $PM_{2.5}$ in the cold period, and (**d**) $PM_{2.5}$ in the warm period at Petrana.

The low concentrations of As, Cd, Cu, Pb, and Zn indicate the near absence of vehicular emissions from traffic in the areas. In general, PM originating from traffic emission is often potentially enriched with trace elements such as As, Cd, Cr, Cu, Zn, Pb, and Ni [41,42].

In accordance with the scientific literature [43], the major elements (Ca, Fe, Mg, Al, Na, and K) mainly originate from a crustal source and are also the main components of coal fly ash. The trace elements (Mn, Co, As, Cd, and Pb) arise at high temperatures during the combustion of coal, while As is also used as a marker for coal combustion. It should be noted that Ca originates from lignite combustion since it exists in fly ash, mainly as lime, anhydrite, portlandite, calcite, etc. [43]. Based on the above, the main emission sources in the two locations are related with mining activities and coal combustion.

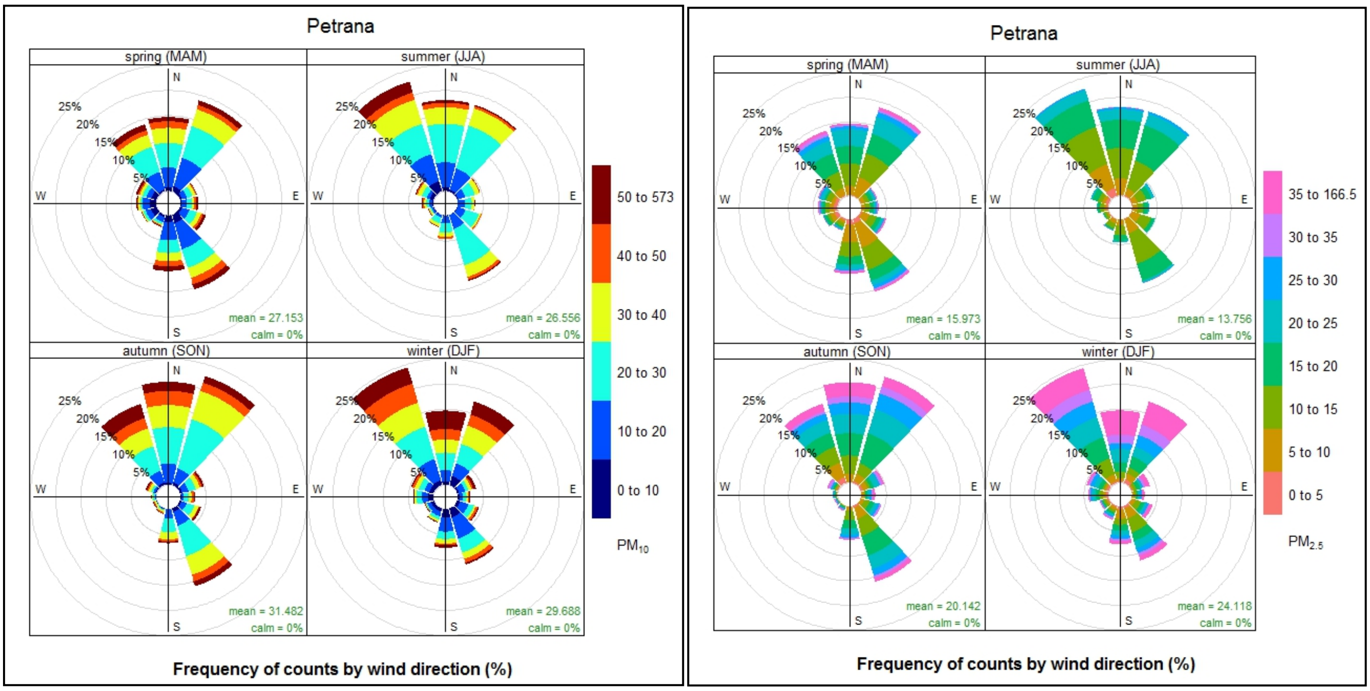
In addition, Iordanidis et al. [44] categorized the airborne particulates in the Florina-Ptolemais-Kozani basin according to their origin in geogenic, biogenic, anthropogenic (mainly fly ash released from lignite-fired power plants), and metalliferous categories. In particular, the geogenic category comprises mineral fragments, aggregates, and agglomerates of various composition derived from soils, sediments, and weathered rock. The mining activities in the region contribute greatly to the geogenic category. Characteristic natural particles in this category are the calcite crystal ($CaCO_3$) and silicon dioxide ($SiO_2$) as well as feldspars [(K, Na, Ca)$AlSi_3O_8$], phyllosilicate minerals, carbonates, and iron oxide-hydroxides. The fly ash category comprises aluminosilicate minerals, iron, and calcium-rich particles. In addition, fly ash can be constituted of potential toxic metals such as Cu and Zn. Fly ash comes from power plants, so larger fly ash-airborne particles are concentrated close to power stations, while fine and ultra-fine fly ash-airborne particles can be transported by the wind in other areas. The biogenic category includes aerosols with organic plant material, fungal hyphae, and spores. The metalliferous category mainly includes iron (Fe) and copper (Cu) enriched particles, denoting motor vehicle activity as its source. In addition, this category may be rich in toxic metals such as Zn and Cu.

Another study conducted by Garas et al. [43] in the same areas of Western Macedonia, applied the multivariate Positive Matrix Factorization (PMF) receptor model in order to identify the main ambient $PM_{10}$ sources that affect the urban sampling sites of the study. For the sampling sites in the city center of Kozani and in the city of Ptolemaida, they found six factors (sources) including two traffic sources (road dust and vehicle exhaust), soil dust, biomass burning, coal, and oil combustion. The source contribution for each site was different. In Ptolemaida, 30.8% of the source contribution came from coal combustion, while in Kozani the highest contribution was traffic, at almost 55%. With reference to the coal combustion source, which is the main focus of our study, it is characterized by the presence of trace elements Be, Ca, Cr, Ni, As, Cd, Pb, and Tl in Kozani and the trace elements As, Cd and Pb in Ptolemaida.

Over the period of sampling in the two locations, measurements of meteorological variables (temperature, humidity, wind speed, and direction) were performed. Table 3 shows the seasonal average values of temperature and humidity in both locations based on data over the 12-months of samplings (from 1 December 2017 to 30 November 2018). Figures 12a,b and 13a,b show the pollution increases of $PM_{2.5}$ and $PM_{10}$ based on data from the air quality monitoring stations at Petrana and Pontokomi from 1 December 2017 to 30 November 2018.

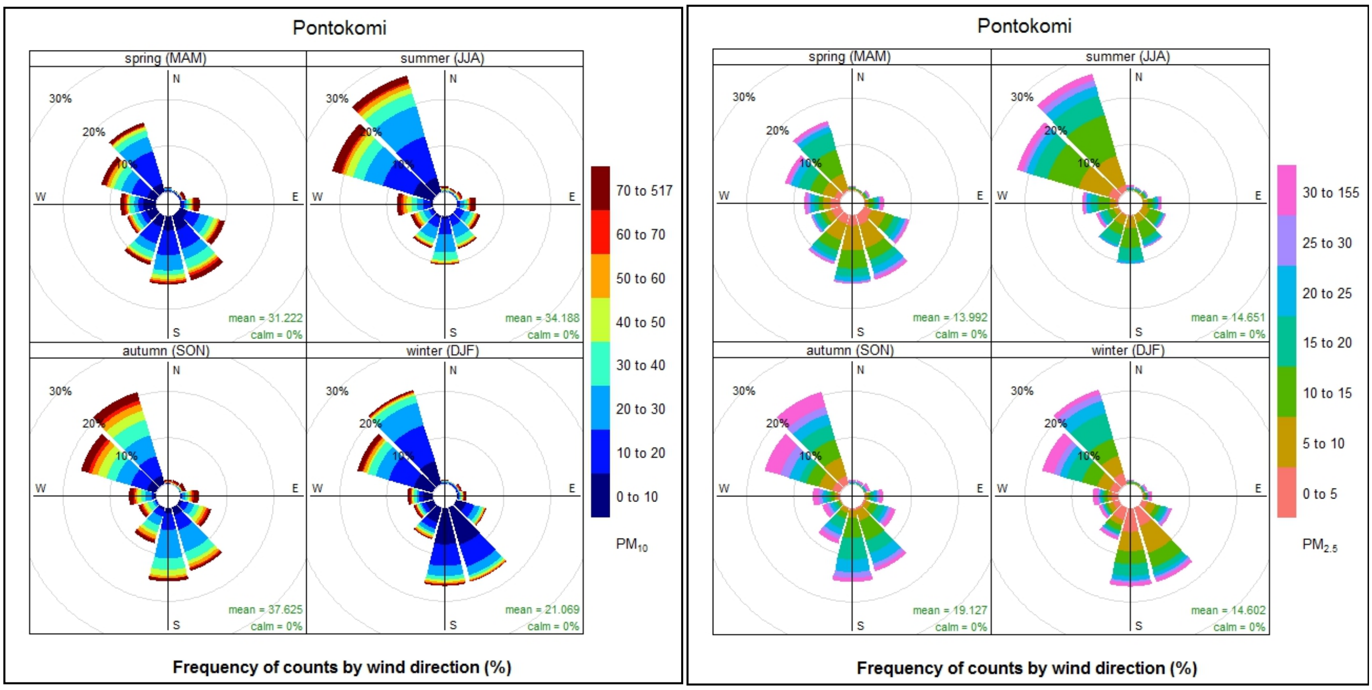
**Table 3.** Description of seasonal average values of meteorological variables during the sampling period (1 December 2017 to 30 November 2018).

| **Pontokomi $PM_{10}$** | **Temp (°C)** | **RH (%)** | **WS (m/s)** | **WD (Deg.)** |
|---|---|---|---|---|
| Winter | 5.59 | 84.50 | 3.24 | 223 |
| Spring | 15.48 | 63.00 | 2.70 | 192 |
| Summer | 22.27 | 62.14 | 2.34 | 264 |
| Autumn | 15.29 | 66.63 | 1.66 | 216 |
| Pontokomi $PM_{2.5}$ | Temp (°C) | RH (%) | WS (m/s) | WD (deg.) |
| Winter | 6.05 | 71.00 | 2.94 | 266 |
| Spring | 18.05 | 61.00 | 1.92 | 210 |
| Summer | 24.36 | 61.80 | 2.12 | 244 |
| Autumn | 13.93 | 70.85 | 2.27 | 208 |
| Petrana $PM_{10}$ | Temp (°C) | RH (%) | WS (m/s) | WD (deg.) |
| Winter | 4.94 | 78.50 | 1.19 | 127 |
| Spring | 14.56 | 63.45 | 1.27 | 157 |
| Summer | 21.74 | 59.71 | 1.20 | 127 |
| Autumn | 15.10 | 66.88 | 1.00 | 168 |
| Petrana $PM_{2.5}$ | Temp (°C) | RH (%) | WS (m/s) | WD (deg.) |
| Winter | 5.50 | 69.13 | 1.49 | 295 |
| Spring | 17.32 | 60.00 | 1.00 | 133 |
| Summer | 23.68 | 58.00 | 1.12 | 143 |
| Autumn | 14.05 | 68.85 | 1.12 | 102 |




(**a**) (**b**)

**Figure 12.** Seasonal pollution increases for (**a**) $PM_{10}$ and (**b**) $PM_{2.5}$ concentrations (µg/m$^3$) based on data from the AQMS Petrana from 1 December 2017 to 30 November 2018. The concentrations of PM are shown by the color scale legend.




(**a**) (**b**)

**Figure 13.** Seasonal pollution increases for (**a**) $PM_{10}$ and (**b**) $PM_{2.5}$ concentrations (µg/m$^3$) from the AQMS Pontokomi from 1 December 2017 to 30 November 2018. The concentrations of PM are shown by the color scale legend.

Figure 12a,b display the seasonal pollution increases for $PM_{10}$ and $PM_{2.5}$ concentrations in Petrana, respectively. The highest PM concentrations were found during autumn and winter. In general, wind data were mainly from the direction between NW-NNW and

N-NE, as well as SE-SSE, while much of the higher PM concentrations were associated with Northerly winds.

Figure 13a,b display the seasonal pollution increases for $PM_{10}$ and $PM_{2.5}$ concentrations at Pontokomi, respectively. In contrary to pollution increases for Petrana, at Pontokomi the seasonal variability is not clear given that relatively high concentrations were indicated throughout the year while high concentrations were also found during summer. In general, wind data were mainly from the directions WNW-NNW and S-SE. Much of the higher PM concentrations were associated with winds from the WNW-NNW direction in summer and autumn. For $PM_{2.5}$, the contribution of S-SE in autumn is substantial, with much of the higher $PM_{2.5}$ concentrations associated with both S-SE and WNW-NNW wind directions. Pontokomi is affected by the operation of two mines, located to the north and east, and by the lignite-fired power plant "Kardia", which is located to the east. The South Field mine, which is the largest in the area in terms of total excavations, is also located east of Pontokomi but at a further distance than the Kardia mine and power plant [15].

Another important aspect of the regional air pollution is the contribution of fly ash–airborne particles from the lignite power stations. Iordanidis et al. [21] found fly-ash in all sampling sites and sampling dates while fine and ultrafine particles were recorded mainly in remote areas and larger fly ash particles were mainly found in the vicinity of power stations. The prevailing wind disperses fly-ash as "fugitive dust", affecting adjacent and remote areas. In addition, the soil in the areas is contaminated due to the deposition of high amounts of fly ash released in the atmosphere for years, influencing the environmental quality through turbulent flow and dust suspension [45].

## 4. Conclusions

The temporal variations of $PM_{10}$ and $PM_{2.5}$ concentrations were assessed over a 12-year period (2010–2021) in the region of Western Macedonia, which is an area highly dominated by mining operations and lignite-fired power plants. These concentrations were correlated with meteorological parameters revealing the substantial contribution of wind speed and direction in their levels. The concentrations of PM exhibited seasonality patterns associated with the local prevailing weather conditions and the activity of lignite-fired power plants. In addition, in the context of the lignite phase-out plan for power generation, the PM levels seems to decrease over the study period, revealing the contribution of intense mining activities and coal-fired plants operation throughout the years.

In addition, the sampling of the concentrations of 17 $PM_{2.5}$-bound and 17 $PM_{10}$-bound trace elements (Al, Mn, Fe, Ni, Cu, Zn, Sn, Pb, Si, Mg, Cr, As, Na, K, Ca, Sr, and Cd) was conducted throughout the period of 12 months (December 2017–November 2018) in 2 sampling locations, Petrana and Pontokomi. The analysis revealed that the mean seasonal concentrations of $PM_{2.5}$-bound and $PM_{10}$-bound trace elements exhibit some differences, but the elemental profiles of both $PM_{2.5}$ and $PM_{10}$ were quite similar. The most abundant elements (Ca and Si) indicate that the main emission sources in the two locations were related with the mining activities and the coal combustion. In addition, the high PM levels correlated with winds blowing from locations of open-cast mines and coal-fired plants.

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