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Analysis of the Particulate Matter Pollution in the Urban Areas of Croatia, EU †

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Abstract: Particulate matter (PM) comprises a mixture of chemical compounds and water particles found in the air. The size of suspended particles is directly related to the negative impact on human health and the environment. In this paper, we present an analysis of the PM pollution in urban areas of Croatia. Data on PM10 and PM2.5 concentrations were measured with nine instruments at seven stationary measuring units located in three continental cities, namely Zagreb (the capital), Slavonski Brod, and Osijek, and two cities on the Adriatic coast, namely Rijeka and Dubrovnik. We analyzed an hourly course of PM2.5 and PM10 concentrations and average seasonal PM2.5 and PM10 concentrations from 2017 to 2019. At most measuring stations, maximum concentrations were recorded during autumn and winter, which can be explained by the intensive use of fossil fuels and traffic. Increases in PM concentrations during the summer months at measuring stations in Rijeka and Dubrovnik may be associated with the intensive arrival of tourists by air during the tourist season, and lower PM concentrations during the winter periods may be caused by a milder climate consequently resulting in lower consumption of fossil fuels and use of electric energy for heating.

Keywords: particulate matter; PM2.5; PM10; air pollution; urban area; Croatia

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1. Introduction

Particulate matter (PM) consists of very small particles impregnated with a solution of acids, heavy metals, different organic and inorganic compounds, particles of dust and soil, etc. The WHO claims that 7 million people annually die premature deaths linked to the combined effects of indoor and outdoor air pollution [1].

Exposure to PM-polluted air is directly correlated to higher mortality rates and lower quality of life [2]. For this reason, the European Union has established a measuring network to monitor daily and annual PM10 concentrations, since PM10 is considered the most relevant for health risks. The monitoring in the United States is more focused on detecting PM2.5 concentrations because the studies conducted there showed that PM2.5 is related to anthropogenic emissions from biomass, combustion of fossil fuels, etc. [3]. From 2015, EU legislation implemented the US-based values to regulate the yearly average anticipated PM2.5 limit values to 25 $\mu g/m^3$ [4].

PM2.5 and PM10 may be responsible for a broad spectrum of adverse health issues, such as chronic obstructive pulmonary disease, asthma, and respiratory admissions [5], and increased mortality [6,7]. Children are especially affected by PM air pollution since they breathe more rapidly and are often closer to the ground. In this way, they inhale and absorb more pollutants. The WHO estimates that more than 90% of children are exposed to airborne pollutants every day. According to the Global Health Observatory (GHO) data in urban areas, the mean concentration of PM2.5 ranges from <10 to >100 μg/m^3, and the mean concentration of PM10 ranges from <10 to >200 μg/m^3 [8].

PM concentrations are usually determined by three approaches: (i) by measuring the concentration using gravimetric, optical, or quartz crystal microbalance principles; (ii) by measuring the size distribution with a scanning mobility particle sizer (SMPS); and (iii) by measuring particle charge size distribution with an electrical low pressure impactor (ELPI) spectrometer [9].

In this paper, we present the results of the hourly and seasonal average PM10 and PM2.5 concentrations in urban areas of Croatia obtained from the nine stationary measuring units located in three continental cities, namely Zagreb (the capital), Slavonski Brod, and Osijek, and two cities on the Adriatic coast, namely Rijeka and Dubrovnik, in the period from 2017 to 2019.

2. Materials and Methods

Nine instruments at seven locations in five cities in Croatia measured the PM2.5 and PM10 concentrations. The sampling interval was each hour for 24 h/day in the period from 2017 to 2019.

2.1. Locations

Data on PM10 and PM2.5 concentrations measured in the period from 2017 to 2019 at the stationary measuring units located in three continental cities, namely Zagreb, Slavonski Brod, and Osijek, and two cities on the Adriatic coast, namely Rijeka and Dubrovnik (Figure 1).

Figure 1. Marked measuring locations in urban areas of Croatia (smaller map [10], larger map [11]).

Stationary measuring units in Zagreb were Zagreb-1 (coordinates 45.800339° N, 15.974072° E), where PM10 was measured in the period from 2017 to 2019, and Zagreb PPI (coordinates 45.834372° N, 15.978394° E), where PM2.5 was measured in the period from 2017 to 2018.

Stationary measuring unit in Osijek was Osijek-1 (coordinates 45.558792° N, 18.698769° E), where PM10 was measured in the period from 2017 to 2019.

Stationary measuring units in Slavonski Brod were Slavonski Brod-1 (coordinates 45.159472° N, 17.995100° E), where PM2.5 was measured in the period from 2017 to 2019, and Slavonski Brod-2 (coordinates 45.149114° N, 18.023450° E), where PM10 was measured in the period from 2017 to 2019.

Stationary measuring unit in Rijeka was Rijeka-2 (coordinates 45.320794° N, 14.483511° E), where PM10 and PM2.5 were measured in the period from 2017 to 2018.

Stationary measuring unit in Dubrovnik was Dubrovnik airport (coordinates 42.553889° N, 18.284722° E), where PM10 and PM2.5 were measured during 2019.

2.2. Instrumentation

The Thermo Andersen ESM FH 62 I-R (ESM Andersen Instruments, Erlangen, Germany) is a beta-ray absorption monitor that measures a mass concentration of the suspended particles in ambient air. The samples are directly collected, and the particle mass is simultaneously measured during sampling by a dual-beam compensation method (to physically eliminate the influence of temperature and pressure) and a single filter-spot position. For this reason, it is used for stable long-term measurements [3]. This instrument was used to monitor the PM2.5 at Slavonski Brod-1 and PM10 at Zagreb-1, Osijek-1, Rijeka-2, and Dubrovnik airport. The instrument used in this study was calibrated every 6 months.

Two gravimetric devices from Sven Leckel (Germany) were used to measure PM concentrations. The first was a Small Filter Device model KFG LVS-3 that was used as a single filter gravimetric sampler. This model can be operated with controlled flow rates between 1.0 and 2.3 m^3/h with <2% deviation from the set point and continuous measurement for a minimum of 1 h and a maximum of 999 h. This instrument was used to monitor the PM2.5 at Zagreb PPI measuring station. The second was a sequential sampler SEQ47/50 that is equipped with PM2.5 and PM10 inlets that comply completely with the European PM2.5/PM10 standard reference sampler according to CEN EN 12341. This instrument was used to monitor the PM2.5 at Rijeka-2 measuring station.

PM2.5 was measured gravimetrically using a Derenda PNS 16T3.1/6.1 (Derenda, Germany). This instrument was used to monitor the PM10 at Slavonski Brod-2 measuring station.

The APDA-371 Ambient Dust Monitor (Horiba, Oberursel, Germany) automatically measures and records PM using the principle of beta-ray attenuation. It operates according to EU and EPA regulations and is also type-approved by TueV. It can operate independently for up to 60 days. This instrument was used to monitor the PM2.5 at Dubrovnik airport.

3. Results

Data on PM10 and PM2.5 concentrations were measured with nine instruments at seven stationary measuring units located in three continental cities, namely Zagreb (the capital), Slavonski Brod, and Osijek, and two cities on the Adriatic coast, namely Rijeka and Dubrovnik. The sampling interval was each hour for 24 h/day in the period from 2017 to 2019.

3.1. Average Hourly Concentrations

Average hourly PM concentration values were calculated by taking the average PM value of each hour during a one-year period. The average hourly PM10 concentrations at Zagreb-1 and Osijek-1 in the period from 2017 to 2019 are presented in Figure 2.

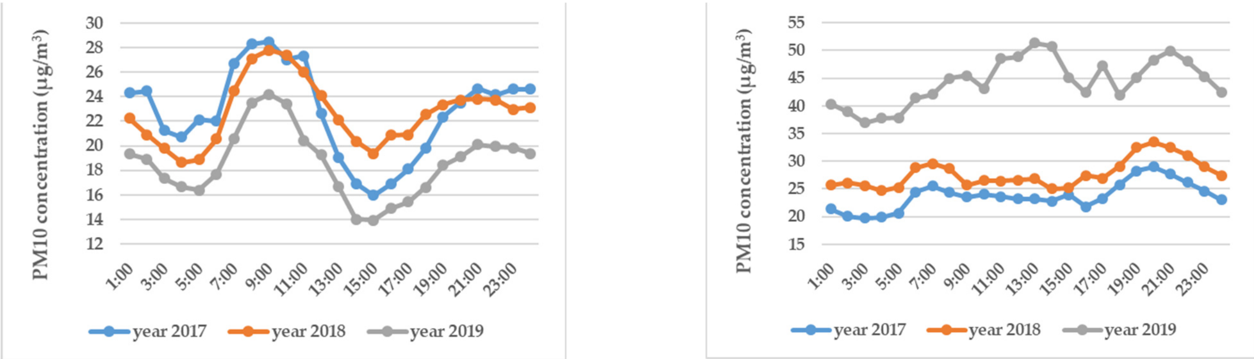

Figure 2. Average hourly PM10 concentrations at Zagreb-1 (**left**) and Osijek-1 (**right**) in the period from 2017 to 2019.

The average hourly PM2.5 concentrations at Slavonski Brod-1 and PM10 concentrations at Slavonski Brod-2 in the period from 2017 to 2019 are presented in Figure 3.

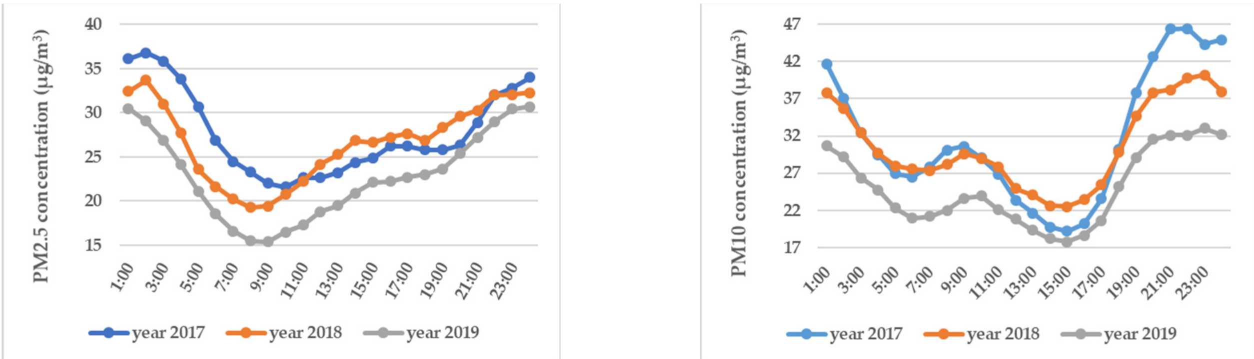

Figure 3. Average hourly PM2.5 concentrations at Slavonski Brod-1 (**left**) and PM10 concentrations at Slavonski Brod-2 (**right**) in the period from 2017 to 2019.

The average hourly PM10 concentrations at Rijeka-2 in the period from 2017 to 2018 and the average hourly PM10 and PM2.5 concentrations at Dubrovnik airport in 2019 are presented in Figure 4.

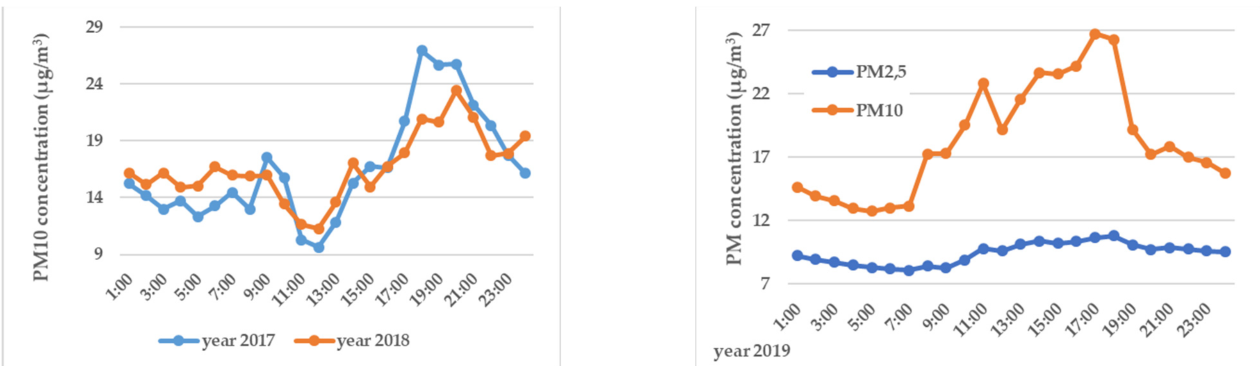

Figure 4. Average hourly PM10 concentrations at Rijeka-2 in the period from 2017 to 2018 (**left**) and average hourly PM10 and PM2.5 concentrations at Dubrovnik airport in 2019 (**right**).

3.2. Average Seasonal Concentrations

Average seasonal PM concentration values were calculated by taking the average PM value for each season for each year separately. The average seasonal PM10 concentrations at Zagreb-1 in the period from 2017 to 2019 and the average seasonal PM2.5 concentrations at Zagreb PPI in the period from 2017 to 2018 are presented in Figure 5.

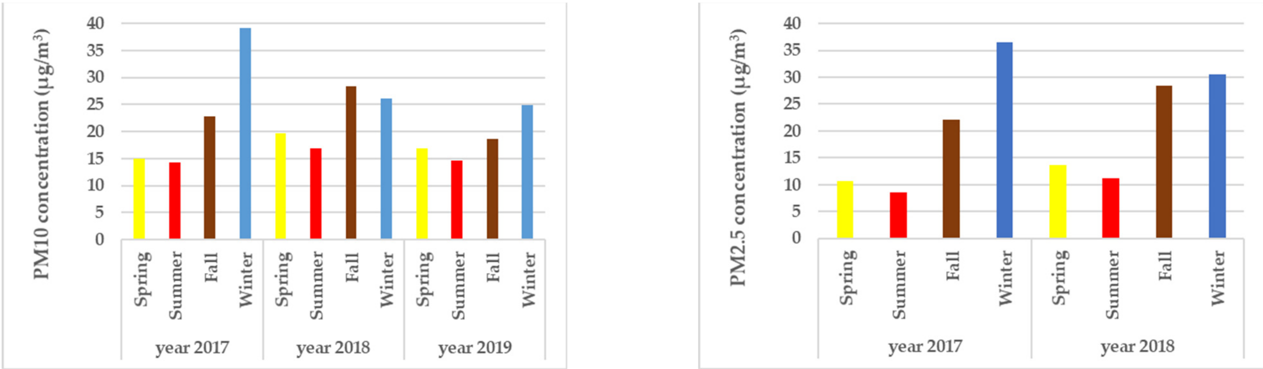

Figure 5. Average seasonal PM10 concentrations at Zagreb-1 in the period from 2017 to 2019 (**left**) and average seasonal PM2.5 concentrations at Zagreb PPI in the period from 2017 to 2018 (**right**).

The average seasonal PM10 concentrations at Osijek-1 in the period from 2017 to 2018 are presented in Figure 6.

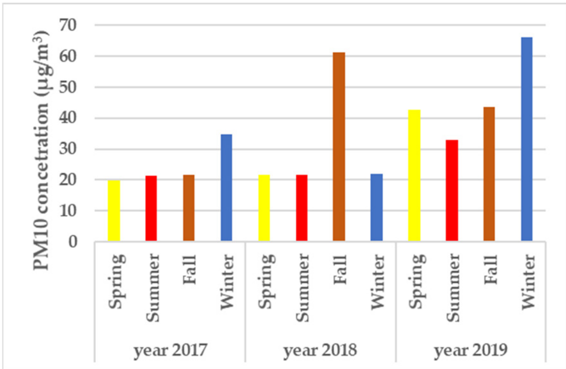

Figure 6. Average seasonal PM10 concentrations at Osijek-1 in the period from 2017 to 2018.

The average seasonal PM2.5 concentrations at Slavonski Brod-1 and PM10 concentrations at Slavonski Brod-2 in the period from 2017 to 2018 are presented in Figure 7.

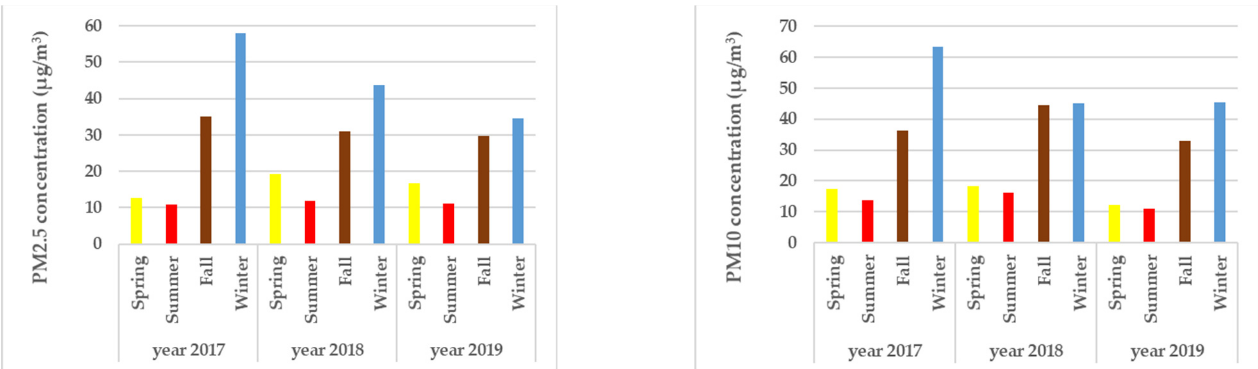

Figure 7. Average seasonal PM2.5 concentrations at Slavonski Brod-1 (**left**) and PM10 concentrations at Slavonski Brod-2 (**right**) in the period from 2017 to 2019.

The average seasonal PM10 and PM2.5 concentrations at Rijeka-2 in the period from 2017 to 2018 and at Dubrovnik airport in 2019 are presented in Figure 8.

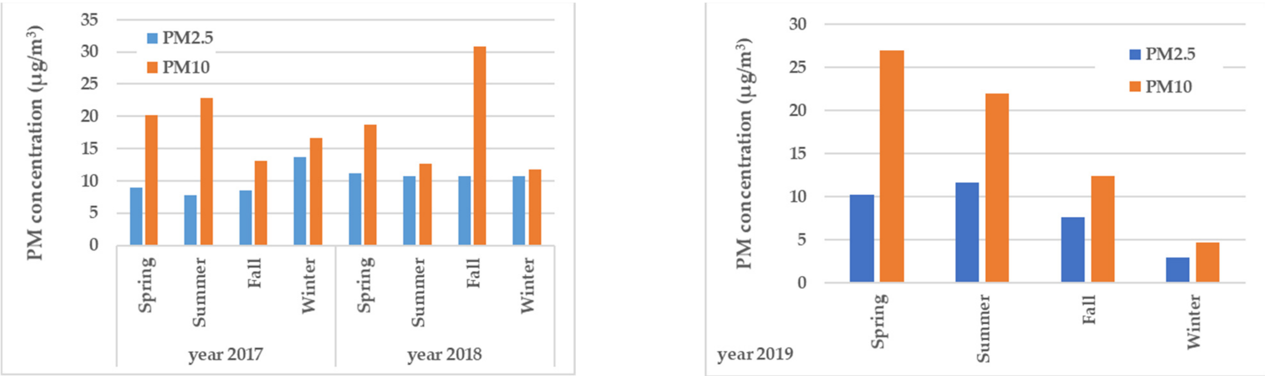

Figure 8. Average seasonal PM10 and PM2.5 concentrations at Rijeka-2 in the period from 2017 to 2018 (**left**) and at Dubrovnik airport in 2019 (**right**).

4. Discussion

4.1. Average Hourly Concentrations

The obtained average 24 h PM concentration values in the urban areas of Croatia show specific shapes and values for different cities and regions.

When observing the average 24 h values for the capital of Croatia, the city of Zagreb, at measuring station Zagreb-1 (Figure 2, left), it can be seen then there is a slight sinusoidal shape of the average PM10 values for 1 h interval within 24 h. The PM10 values started to increase from 5 a.m., achieving the maximum between 9 a.m. and 10 a.m. (28 µg/m^3 in 2017, 27 µg/m^3 in 2018, and 24 µg/m^3 in 2019) and then slowly decreasing until 3 p.m. After 3 p.m., the values started to increase, achieving maximal values at 8 p.m. (but still lower than daytime maxima). For all 3 observed years, the values were similar.

When observing the average 24 h PM10 values for the city of Osijek in the east of Croatia, at Osijek-1 measuring station, the values for 2017 and 2018 appear to be similar, reaching two maxima at 7 a.m. and 8 p.m., with values below 30 µg/m^3. In 2019, the values were much higher (all above 35 µg/m^3) with different trends, rising from the minimal values at 5 a.m. and reaching maximal values at 1 p.m. (52 µg/m^3).

When observing the average 24 h PM10 and PM2.5 values for the city of Slavonski Brod, the PM value trends look different than in previously considered cities. PM10 values (Figure 3, right) started to increase from 6 a.m. to 9 a.m. Then, the values slowly decreased until 4 p.m. After 5 p.m., values started to rapidly increase, reaching maxima at 9 p.m. (32 to 47 µg/m^3, from 2019 to 2017). The city of Slavonski Brod is located at the border with Bosnia and Herzegovina, which is a hard transit border and a border between the EU and outer Balkan countries. Heavy traffic, commuting, and a petrol plant near Slavonski Brod are the possible cause for high PM10 values in the late evening, and during the night. PM2.5 values have a similar trend (Figure 3, left), with the maxima at 2 a.m. (28 to 37 µg/m^3, from 2019 to 2017) followed by minima at 8 a.m. and then a slow increase up to late in the night. It can be noted that initial values of both PM10 and PM2.5 in 2019 are lower than in previous years.

When observing the average 24 h PM10 values for two cities on the Adriatic coast, at the sites of Rijeka and Dubrovnik airport, it can be seen that 24 h PM values were lower than in the continent. In the city of Rijeka (Figure 4, left), the PM values started to increase from 1 p.m., reaching maximal values in the evening at 6 p.m. (27 µg/m^3) and 8 p.m. (23 µg/m^3). During the night and early morning, the values were constant and low (15 µg/m^3). At the Dubrovnik airport, the PM10 values started to increase from 7 a.m., reaching maximal values at 5 a.m. (26 µg/m^3). PM2.5 values have similar trends but at much lower values (maxima at 10 µg/m^3).

4.2. Average Seasonal Concentrations

When observing the average seasonal PM10 values for the Zagreb-1 measuring station (Figure 5, left), a trend can be seen in the behavior of PM10 values: during summers, PM10 values are lowest (approx. 15 μg/m^3), while during winters, PM10 values are highest (maximum 39 μg/m^3 in winter 2017). The same trend can be observed for PM2.5 at Zagreb PPI (Figure 5, right) measuring station, with minimal values during summer (approx. 10 μg/m^3) and maximal values during winter (36 μg/m^3).

When observing the average seasonal PM10 values for the Osijek-1 measuring station (Figure 6), it can be seen that the values during springs and summers in 2017 and 2018 were similar (approx. 20 μg/m^3), but there was a great increase in the PM10 values in fall 2018 (61 μg/m^3). The PM10 values in 2019 were much higher than in previous years, with a trend similar to that seen in the city of Zagreb, with low values during summer (33 μg/m^3) and high values during winter (77 μg/m^3), but the values were much higher compared to the city of Zagreb.

When observing the average seasonal PM10 and PM2.5 values for Slavonski Brod (Figure 7), it can be observed that the lowest values appeared during summer (approx. 20 μg/m^3 for PM10 and approx. 12 μg/m^3 for PM2.5) and the highest values appeared during winter (58 μg/m^3 for PM10 in 2017 and 62 μg/m^3 for PM2.5 in 2017). This trend is similar to the PM trend observed for the city of Zagreb.

When observing the average seasonal PM10 and PM2.5 values for the Rijeka-2 measuring station (Figure 8, left), it can be seen that the highest PM10 values were in spring (20 μg/m^3) and summer (23 μg/m^3) in 2017 and spring (19 μg/m^3) and fall (31 μg/m^3) in 2018. During 2017, PM2.5 values were the lowest during summer (8 μg/m^3) and the highest during winter (14 μg/m^3), while in 2018, the PM2.5 values were approximately the same, at 11 μg/m^3.

When observing the average seasonal PM10 and PM2.5 values during 2019 for the Dubrovnik airport (Figure 8, right) measuring station, it can be seen that the highest PM10 values were during spring (27 μg/m^3), with decreasing tendency up to the winter (4 μg/m^3). The highest PM2.5 values were obtained during summer (12 μg/m^3), while the lowest values were obtained during winter (3 μg/m^3). Higher PM10 and PM2.5 values obtained spring and summer may be caused by the intensive touristic arrivals and heavier air traffic during these months.

5. Conclusions

Analysis of the average hourly concentrations of PM emissions showed that there is a difference between PM emissions in the capital city of Zagreb and other urban areas in Croatia, with the highest PM10 emissions during early morning and later evening.

During 2019, PM emissions in Osijek were noticeably higher than in the rest of the analyzed cities.

Heavy traffic, commuting, and border crossing at the international border crossing in Slavonski Brod between EU and other Balkan countries seem to influence the average hourly concentrations of PM emission, with the highest values observed during the late-night hours. At Slavonski Brod, the PM2.5 and PM10 emissions are correlated and have the same daily tendency.

Coastal cities Rijeka and Dubrovnik have the lowest PM emissions. As expected, at the Dubrovnik airport, the heavier air traffic increases the PM10 during working hours, from 7 a.m. to 7 p.m. PM2.5 has the same tendency, but with much lower emission values.

Cities in the continent obtained higher seasonal PM emission values during the fall and winter months when compared to the coastal cities. The lower PM10 and PM2.5 values during fall and winter months for the coastal cities are due to milder (sub-)Mediterranean climate and reduced amount of fossil fuel consumption during these months because electricity is the primary source for heating.

It can be noted that in the coastal cities, the average PM2.5 emissions were always much lower than average PM10 emissions, regarding the 24 h period or seasonal period.

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