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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 air. The size of suspended particles is directly related to the negative impact on human health and the environment. In this paper, we presented 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, Zagreb (the capital), Slavonski Brod and Osijek; and two cities at the Adriatic coast, Rijeka and Dubrovnik. We analyzed an hour 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 caused by a milder climate consequently resulting in lower fossil fuels consumption and the use of electric energy for heating.

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

#### 1. Introduction

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

The exposure to the PM polluted air is directly correlated to the higher mortality rates and lower quality of life [2]. For this reason, European Union has set the measuring network to monitor daily and annual PM10 concentrations, since PM10 is considered as the most relevant for health risks. US countries are more focused on detecting the PM2.5, since the studies there showed that PM2.5 is related to anthropogenic emissions like 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  $25~\mu\text{g/m}^3$ . [4]

PM2.5 and PM10 could been responsible for broad spectra of adverse health issues like chronic obstructive pulmonary disease, asthma and respiratory admissions [5] and increased mortality. [6], [7] Children are especially effected by the PM air pollution since they breath more rapidly and are

often closer to the ground. In this way, they inhale and absorb more pollutants. 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  $100 > \mu g/m^3$ , and from less than 10 to over  $200 \mu g/m^3$  for PM10. [8]

PMs are usually determinated by three approaches: i) by measuring the concentration using gravimetric, optical or quartz crystal microbalance principles, ii) by measuring the size distribution Scanning Mobility Particle Sizer (SMPS) and iii) by measuring particle charge size distribution by the Electrical Low Pressure Impactor (ELPI) spectrometer. [9]

In this paper, we presented the results of the hour 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, Zagreb (the capital), Slavonski Brod and Osijek; and two cities at the Adriatic coast, Rijeka and Dubrovnik in a 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. The sampling interval was each hour during 24 hrs/day, in a period from 2017 to 2019.

#### 2.1. Locations

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



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  $45,800339^{\circ}$  N,  $15,974072^{\circ}$  E) where PM10 were measured in a period from 2017 to 2019; and Zagreb PPI (coordinates  $45,834372^{\circ}$  N,  $15,978394^{\circ}$  E) where PM2.5 were measured in a period from 2017 to 2018.

Stationary measuring unit in Osijek was Osijek-1 (coordinates  $45,558792^{\circ}$  N,  $18,698769^{\circ}$  E) where PM10 were measured in a period from 2017 to 2019.

Stationary measuring unit in Slavonski Brod were Slavonski Brod-1 (coordinates 45,159472° N, 17,995100° E) where PM2.5 were measured in a period from 2017 to 2019; and Slavonski Brod-2 (coordinates 45,149114° N, 18,023450° E) where PM10 were measured in a period from 2017 to 2019.

Stationary measuring unit in Rijeka was Rijeka-2 (coordinates  $45,320794^{\circ}$  N,  $14,483511^{\circ}$  E) where PM10 and PM2.5 were measured in a 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*

Thermo Andersen ESM FH 62 I-R (ESM Andersen Instruments, Germany) is a beta-ray absorption monitor that measures a mass concentration of the suspended particles in ambient air. The samples are directly collected through and the particle mass was simultaneous measurement during sampling by a dual-beam compensation method (to physically eliminate the temperature and pressure influence) and a single filter–spot position. For this reason, it is used for stable long-term measurements. [3] The instrument was calibrated every 6 months. 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.

Two gravimetric devices from Sven Leckel (Germany) were used to measure PMs. First is 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 deviation from the set point: < 2%, and minimum 1 h – maximum 999 h continuous measurement. This instrument was used to monitor the PM2.5 at Zagreb PPI measuring station.

Second is a sequential sampler SEQ47/50 that is equipped with PM2.5 respectively PM10 inlet inlet complies 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.

PM 2.5 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.

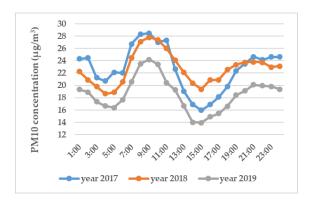
The APDA-371 Ambient Dust Monitor (Horiba, Germany) automatically measures and records PM using the principle of beta ray attenuation. It operates according to EU and EPA regulations and is also type approved by TueV. Is can operate independently up to 60 days. This instrument was used to monitor the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

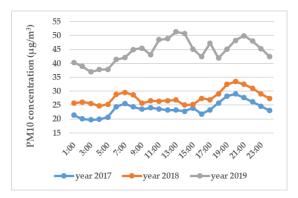
#### 3. Results

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

# 3.1. Average hour concentrations

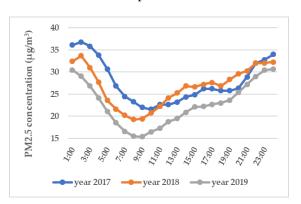
Average hour/day PM concentration values were calculated by taking the average PM value of each hour during one-year period. The average hour PM10 concentrations in Zagreb-1 and Osijek-1 in a period from 2017 to 2019 are presented in Figure 2.

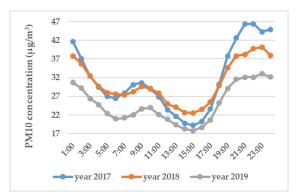




**Figure 2.** Average hour PM10 concentrations in Zagreb-1 (left) and Osijek-1 (right) in a period from 2017 to 2019.

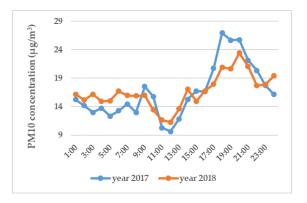
The average hour PM2.5 concentrations in Slavonski Brod-1 and PM10 concentrations in Slavonski Brod-2 in a period from 2017 to 2019 are presented in Figure 3.

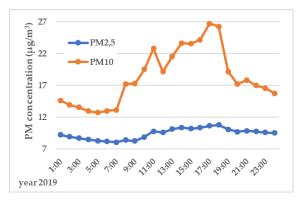




**Figure 3.** Average hour PM2.5 concentrations in Slavonski Brod-1 (left) and PM10 concentrations in Slavonski Brod-2 (right) in a period from 2017 to 2019.

The average hour PM10 and PM2.5 concentrations in Rijeka-2 and Dubrovnik airport in a period from 2017 to 2018 and during 2019, are presented in Figure 4.

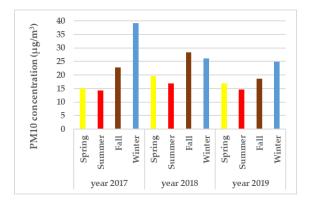


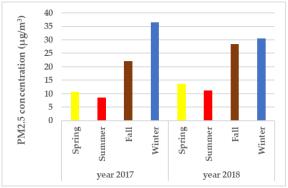


**Figure 4.** Average hour PM10 concentrations in Rijeka-2 (left) and PM10 and PM2.5 concentrations in Dubrovnik airport (right) in a period from 2017 to 2018 and during 2019, respectively.

#### 3.2. Average seasonal concentrations

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





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

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

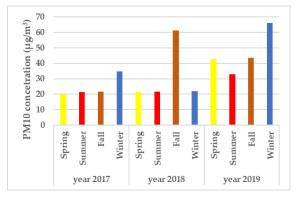
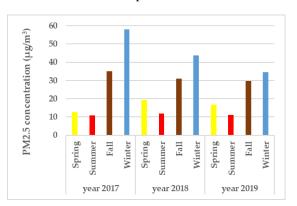
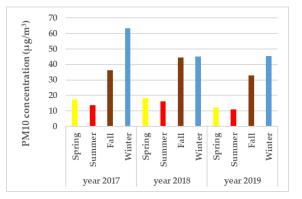


Figure 6. Average seasonal PM10 concentrations in Osijek-1 in a period from 2017 to 2018.

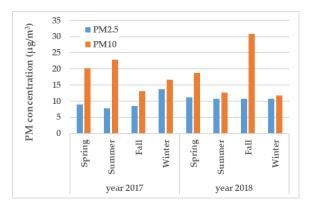
The average seasonal PM2.5 concentrations in Slavonski Brod-1 and PM10 concentrations in Slavonski Brod-2 in a period from 2017 to 2018 are presented in Figure 7.

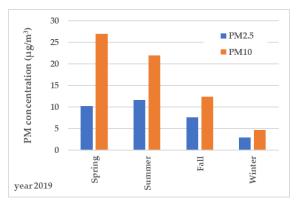




**Figure 7.** Average seasonal PM2.5 concentrations in Slavonski Brod-1 (left) and PM10 concentrations in Slavonski Brod-2 (right) in a period from 2017 to 2019.

The average seasonal PM10 and PM2.5 concentrations in Rijeka-2 and Dubrovnik airport in a period from 2017 to 2018 and during 2019 are presented in Figure 7.





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

#### 4. Discussion

### 4.1. Average hour concentrations

Obtained average 24-hour PM concentration values in the urban areas of Croatia shower specific shapes and values for different cities and regions.

When observing the average 24-hour 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-hour interval within 24 hours. The PM10 values started to raise from 5 a.m., achieving the maximum between 9 and 10 a.m. (28  $\mu$ g/m³ in 2017, 27  $\mu$ g/m³ in 2018, and 24  $\mu$ g/m³ in 2019), and then slowly decreasing until 3 p.m. After 3 p.m. the values started to increase achieving maxima values at 8 p.m. (but still lower than daytime maxima). For all three observed years, the values were similar.

When observing the average 24-hour PM10 values for the city of Osijek in the east of Croatia, at Osijek-1 measuring station, the values for 2017 and 2018 were similar, reaching two maxima; at 7 a.m. and at 8 p.m., with values below 30  $\mu$ g/m³. In year 2019 the values were much higher (all above 35  $\mu$ g/m³) with different trends, rising from the minima values at 5 a.m. reaching maximal values at 1 p.m. (52  $\mu$ g/m³).

When observing the average 24-hour PM10 and PM2.5 values for the city of Slavonski Brod, the PM vale trends look different than in previous cities. PM10 values (Figure 3, right) were starting to raise form 6 a.m. to 9.am. Then the values were slowly decreasing until 4 p.m. After 5 p.m. values start to rapidly increase reaching maxima at 9 p.m. (32 to 47  $\mu$ g/m³, 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 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 evenings, and during the night. PM2.5 values have a similar trend (Figure 3, left), with the maxima at 2.a.m. (28 to 37  $\mu$ g/m³, from 2019 to 2017), following the minimum at 8 a.m. and the slowly increasing 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-hour PM10 values for two cities at the Adriatic coast, cities of Rijeka and Dubrovnik airport, it can be seen that 24-hours PM values were lower than in the continent. In the city of Rijeka (Figure 4, left) the PM values started to raise from 1 p.m. reaching maxima values in the evening, at 6 p.m. (27  $\mu$ g/m³) and 8 p.m. (23  $\mu$ g/m³). During the night and early morning, the values were constant and low (15  $\mu$ g/m³). At the Dubrovnik airport, the PM10 values started to raise from 7 a.m., reaching maxima values at 5 a.m. (26  $\mu$ g/m³). PM2.5 values have similar trends but at much lower values (maxima at 10  $\mu$ g/m³).

### 4.2. Average seasonal concentrations

When observing the average seasonal PM10 values for the Zagreb-1 (Figure 5, left) measuring station there is a trend in PM10 values behaviour, during summers have the lowest PM10 values (approx. 15  $\mu$ g/m³), while during winters have the highest PM10 values (maximum 39  $\mu$ g/m³ in winter 2017). The same trend can be observed for PM2.5 at Zagreb PPI (Figure 5, right) measuring station, with minimal value during summer (approx. 10  $\mu$ g/m³) and maximal during winter (36  $\mu$ g/m³).

When observing the average seasonal PM10 values for the Osijek-1 measuring station (Figure 6) the values during springs and summers in 2017 and 2018 were similar (approx. 20  $\mu$ g/m³), but there was a great raise in the PM10 values in fall 2018 (61  $\mu$ g/m³). The PM10 values in 2019 were much higher than in previous years, with the trend similar to city of Zagreb, low value during summer (33  $\mu$ g/m³) and high values during winter (77  $\mu$ g/m³), but the values were much higher compared to the city of Zagreb.

When observing the average seasonal PM10 and PM2.5 values for the Slavonski Brod (Figure 7), it can be observed that the lowest values appeared during summer (approx.  $20 \mu g/m^3$  for PM10 and approx.  $12 \mu g/m^3$  for PM2.5) and the highest values during winter ( $58 \mu g/m^3$  for PM10 in 2017 and  $62 \mu g/m^3$  for PM2.5 in 2017). This trend is similar to the PM trend present at 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  $\mu g/m^3$ ) and summer (23  $\mu g/m^3$ ) in 2017 and spring (19  $\mu g/m^3$ ) and fall (31  $\mu g/m^3$ ) in 2018. During 2017 PM2.5 values were the lowest during summer (8  $\mu g/m^3$ ) and the highest during winter (14  $\mu g/m^3$ ), while in 2018 the PM2.5 values were approximately the same, at 11  $\mu g/m^3$ .

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  $\mu g/m^3$ ) with decreasing tendency up to the winter (4  $\mu g/m^3$ ). The highest PM2.5 values were obtained during summer (12  $\mu g/m^3$ ) while the lowest values were during winter (3  $\mu g/m^3$ ). Spring and summer obtained higher PM10 and PM2.5 values caused by the intensive touristic arrivals and heavier air traffic during these months.

## 5. Conclusions

Average hour concentrations of PM emissions analysis 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 international border crossing in Slavonski Brod between EU and other Balkan countries seems to influence the average hour concentrations of PM emission, with highest values during 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 emission of PMs. As expected, at the Dubrovnik airport the heavier air traffic raises 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 vales during fall and winter months, compared to the coastal cities. The lower PM10 and PM2.5 vales 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 since electricity is the primary source for heating.

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

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Conflicts of Interest: The authors declare no conflict of interest.

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