

1 *Conference Proceedings Paper*

2 **Analysis of the particulate matter pollution in the** 3 **urban areas of Croatia, EU**

4 **Martina Habulan**¹, **Bojan Đurin**², **Anita Ptiček Siročić**¹, **Nikola Sakač**^{1,*}

5 ¹ Faculty of Geotechnical Engineering, University of Zagreb, HR-42000 Varaždin, Croatia;
6 nsakac@gfv.unizg.hr

7 ² Department for Civil Engineering, University North, HR-42000 Varaždin, Croatia; bdjurin@unin.hr

8 * Correspondence: nsakac@gfv.unizg.hr (N.S.)

9 Received: date; Accepted: date; Published: date

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

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

24

25 **1. Introduction**

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

30 The exposure to the PM polluted air is directly correlated to the higher mortality rates and lower
31 quality of life [2]. For this reason, European Union has set the measuring network to monitor daily
32 and annual PM10 concentrations, since PM10 is considered as the most relevant for health risks. US
33 countries are more focused on detecting the PM2.5, since the studies there showed that PM2.5 is
34 related to anthropogenic emissions like biomass, combustion of fossil fuels, etc. [3]. From 2015, EU
35 legislation implemented the US based values to regulate the yearly average anticipated PM2.5 limit
36 values to 25 µg/m³. [4]

37 PM2.5 and PM10 could be responsible for broad spectra of adverse health issues like chronic
38 obstructive pulmonary disease, asthma and respiratory admissions [5] and increased mortality. [6],
39 [7] Children are especially effected by the PM air pollution since they breath more rapidly and are

40 often closer to the ground. In this way, they inhale and absorb more pollutants. WHO estimates that
41 more than 90% of children are exposed to airborne pollutants every day. According to the Global
42 Health Observatory (GHO) data in urban areas, the mean concentration of PM_{2.5} ranges from 10<
43 100> µg/m³, and from less than 10 to over 200 µg/m³ for PM₁₀. [8]

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

48 In this paper, we presented the results of the hour and seasonal average PM₁₀ and PM_{2.5}
49 concentrations in urban areas of Croatia obtained from the nine stationary measuring units located
50 in three continental cities, Zagreb (the capital), Slavonski Brod and Osijek; and two cities at the
51 Adriatic coast, Rijeka and Dubrovnik in a period from 2017 to 2019.

52 2. Materials and methods

53 Nine instruments at seven locations in five cities in Croatia measured the PM_{2.5} and PM₁₀. The
54 sampling interval was each hour during 24 hrs/day, in a period from 2017 to 2019.

55 2.1. Locations

56 Data on PM₁₀ and PM_{2.5} concentrations measured in a period from 2017 to 2019 at the stationary
57 measuring units located in three continental cities, Zagreb, Slavonski Brod and Osijek; and two cities
58 at the Adriatic coast, Rijeka and Dubrovnik.



59

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

61 Stationary measuring units in Zagreb were Zagreb-1 (coordinates 45,800339° N, 15,974072° E)
62 where PM₁₀ were measured in a period from 2017 to 2019; and Zagreb PPI (coordinates 45,834372°
63 N, 15,978394° E) where PM_{2.5} were measured in a period from 2017 to 2018.

64 Stationary measuring unit in Osijek was Osijek-1 (coordinates 45,558792° N, 18,698769 ° E) where
65 PM10 were measured in a period from 2017 to 2019.

66 Stationary measuring unit in Slavonski Brod were Slavonski Brod-1 (coordinates 45,159472° N,
67 17,995100° E) where PM2.5 were measured in a period from 2017 to 2019; and Slavonski Brod-2
68 (coordinates 45,149114° N, 18,023450° E) where PM10 were measured in a period from 2017 to 2019.

69 Stationary measuring unit in Rijeka was Rijeka-2 (coordinates 45,320794° N, 14,483511° E) where
70 PM10 and PM2.5 were measured in a period from 2017 to 2018.

71 Stationary measuring unit in Dubrovnik was Dubrovnik airport (coordinates 42,553889° N,
72 18,284722° E) where PM10 and PM2.5 were measured during 2019.

73 2.2. Instrumentation

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

82 Two gravimetric devices from Sven Leckel (Germany) were used to measure PMs. First is a Small
83 Filter Device model KFG LVS-3 that was used as a single filter gravimetric sampler. This model can
84 be operated with controlled flow rates between 1,0 and 2,3 m³/h with deviation from the set point: <
85 2%, and minimum 1 h – maximum 999 h continuous measurement. This instrument was used to
86 monitor the PM2.5 at Zagreb PPI measuring station.

87 Second is a sequential sampler SEQ47/50 that is equipped with PM2.5 respectively PM10 inlet
88 inlet complies completely with the European PM2.5/PM10 standard reference sampler according to
89 CEN EN 12341. This instrument was used to monitor the PM2.5 at Rijeka-2 measuring station.

90 PM 2.5 was measured gravimetrically, using a Derenda PNS 16T3.1/6.1 (Derenda, Germany).
91 This instrument was used to monitor the PM10 at Slavonski Brod-2 measuring station.

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

96 3. Results

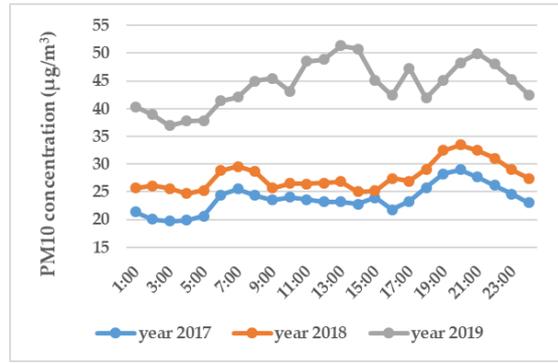
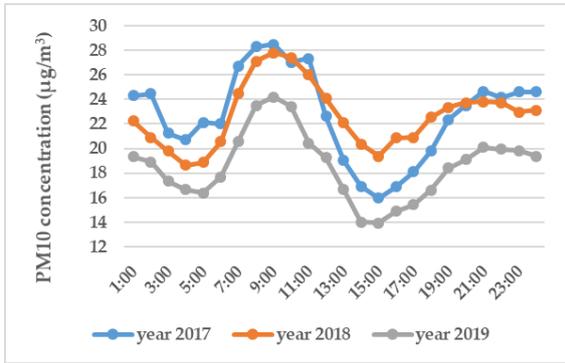
97 Data on PM10 and PM2.5 concentrations were measured with nine instruments at seven
98 stationary measuring units located in three continental cities, Zagreb (the capital), Slavonski Brod
99 and Osijek; and two cities at the Adriatic coast, Rijeka and Dubrovnik. The sampling interval was
100 each hour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

101 3.1. Average hour concentrations

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

105

106



107

108

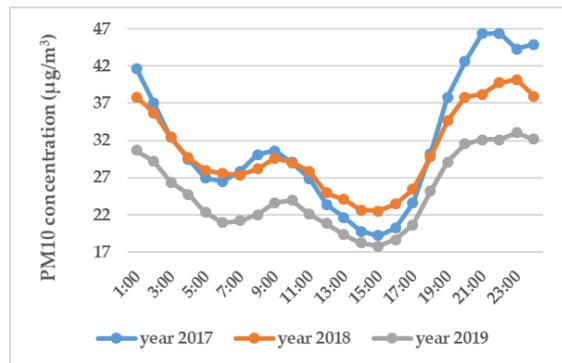
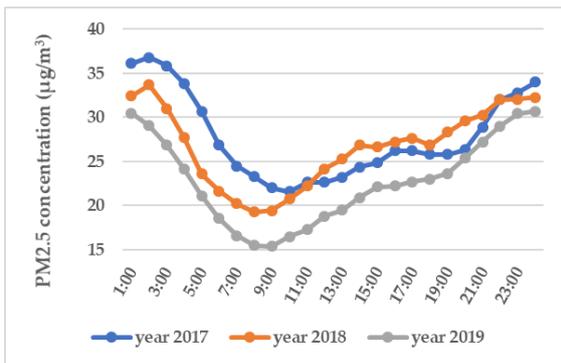
109

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

110

111

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.



112

113

114

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.

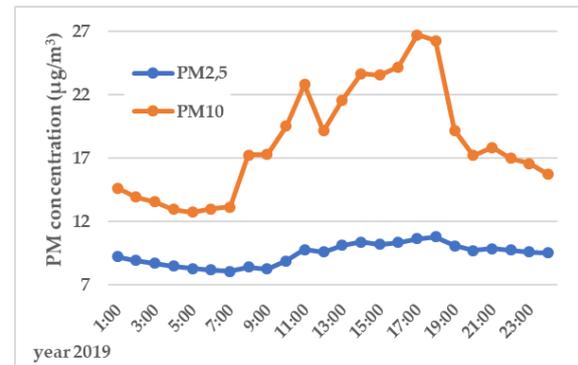
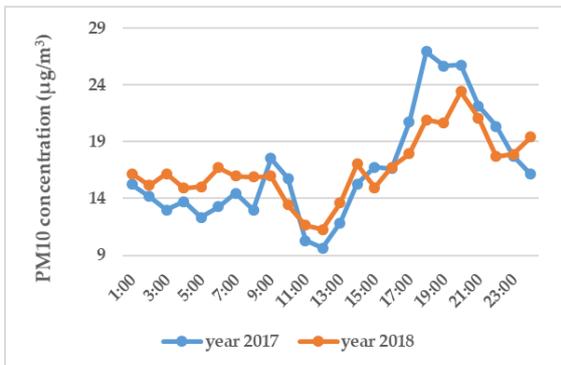
115

116

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.

117

118



119

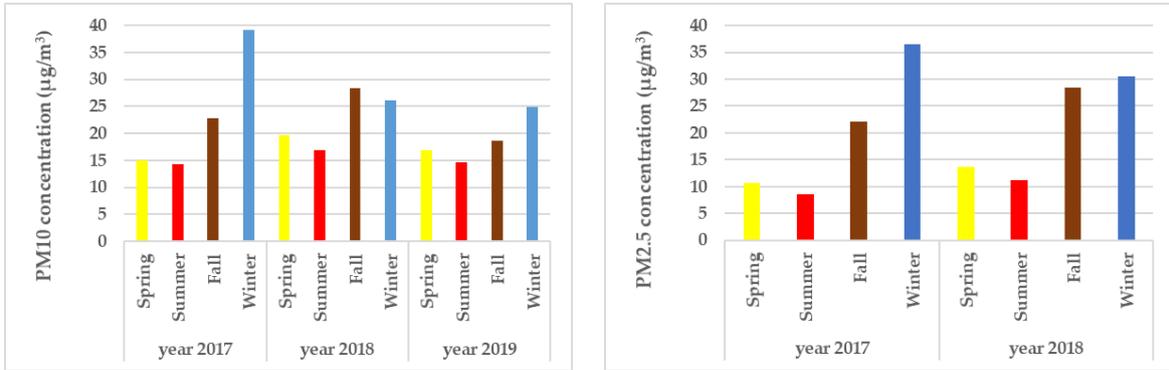
120

121

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.

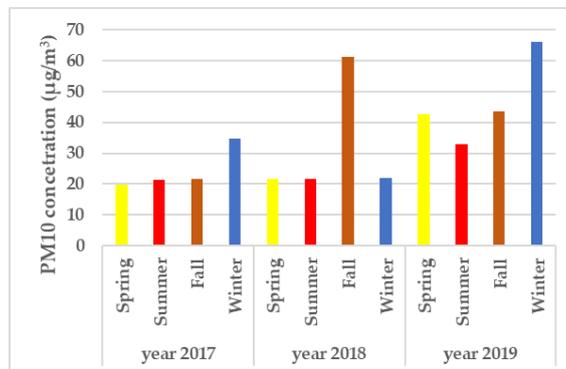
122 3.2. Average seasonal concentrations

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



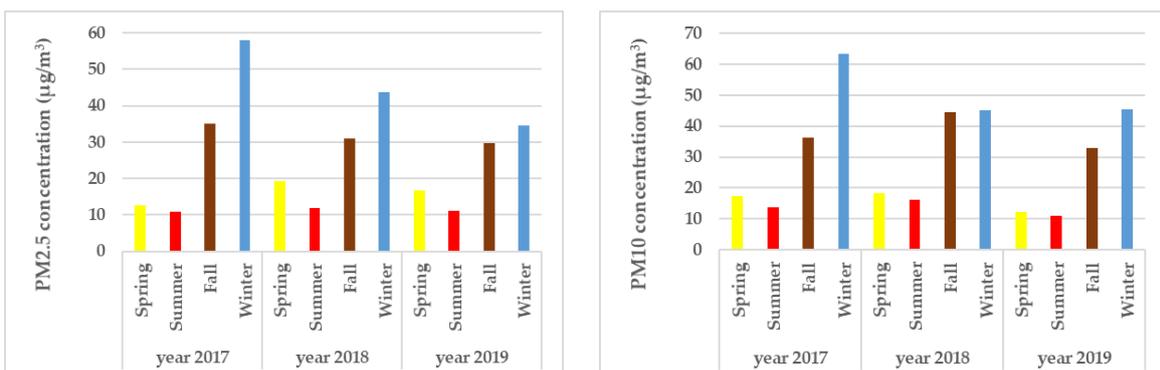
127
 128 **Figure 5.** Average seasonal PM10 concentrations in Zagreb-1 (left) and PM2.5 concentrations in Zagreb
 129 PPI (right) in a period from 2017 to 2019 and 2017 to 2018, respectively.

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



132
 133 **Figure 6.** Average seasonal PM10 concentrations in Osijek-1 in a period from 2017 to 2018.
 134

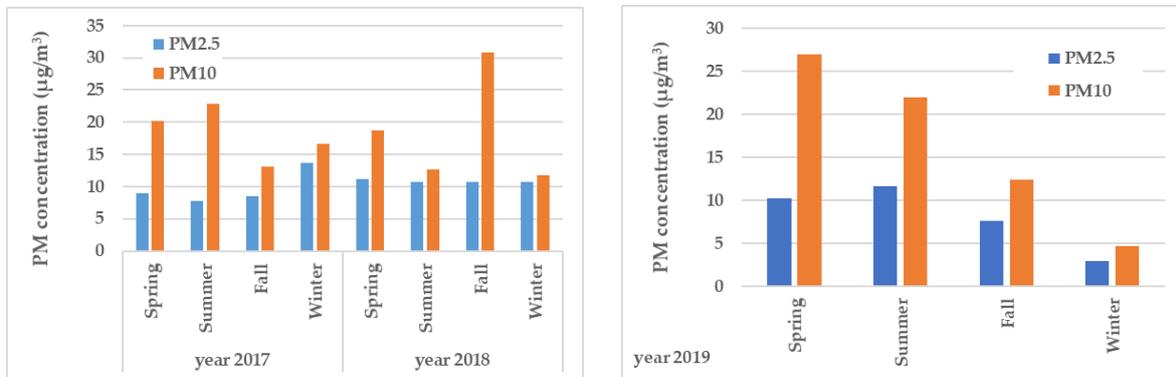
135 The average seasonal PM2.5 concentrations in Slavonski Brod-1 and PM10 concentrations in
 136 Slavonski Brod-2 in a period from 2017 to 2018 are presented in Figure 7.



137

138 **Figure 7.** Average seasonal PM_{2.5} concentrations in Slavonski Brod-1 (left) and PM₁₀ concentrations in
139 Slavonski Brod-2 (right) in a period from 2017 to 2019.

140 The average seasonal PM₁₀ and PM_{2.5} concentrations in Rijeka-2 and Dubrovnik airport in a
141 period from 2017 to 2018 and during 2019 are presented in Figure 7.



142 **Figure 8.** Average seasonal PM₁₀ and PM_{2.5} concentrations in Rijeka-2 (left) and Dubrovnik airport
143 (right) in a period from 2017 to 2018 and during 2019, respectively.
144

145 4. Discussion

146 4.1. Average hour concentrations

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

149 When observing the average 24-hour values for the capital of Croatia, the city of Zagreb, at
150 measuring station Zagreb-1 (Figure 2, left) it can be seen then there is a slight sinusoidal shape of the
151 average PM₁₀ values for 1-hour interval within 24 hours. The PM₁₀ values started to raise from 5
152 a.m., achieving the maximum between 9 and 10 a.m. (28 µg/m³ in 2017, 27 µg/m³ in 2018, and 24
153 µg/m³ in 2019), and then slowly decreasing until 3 p.m. After 3 p.m. the values started to increase
154 achieving maxima values at 8 p.m. (but still lower than daytime maxima). For all three observed
155 years, the values were similar.

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

161 When observing the average 24-hour PM₁₀ and PM_{2.5} values for the city of Slavonski Brod, the
162 PM value trends look different than in previous cities. PM₁₀ values (Figure 3, right) were starting to
163 raise from 6 a.m. to 9 a.m. Then the values were slowly decreasing until 4 p.m. After 5 p.m. values
164 start to rapidly increase reaching maxima at 9 p.m. (32 to 47 µg/m³, from 2019 to 2017). The city of
165 Slavonski Brod is located at the border with Bosnia and Herzegovina, which is a hard transit border
166 and a border between EU and outer Balkan countries. Heavy traffic, commuting and a petrol plant
167 near Slavonski Brod are the possible cause for high PM₁₀ values in the late evenings, and during the
168 night. PM_{2.5} values have a similar trend (Figure 3, left), with the maxima at 2 a.m. (28 to 37 µg/m³,
169 from 2019 to 2017), following the minimum at 8 a.m. and the slowly increasing up to late in the night.
170 It can be noted that initial values of both PM₁₀ and PM_{2.5} in 2019 are lower than in previous years.

171 When observing the average 24-hour PM₁₀ values for two cities at the Adriatic coast, cities of
172 Rijeka and Dubrovnik airport, it can be seen that 24-hours PM values were lower than in the
173 continent. In the city of Rijeka (Figure 4, left) the PM values started to raise from 1 p.m. reaching
174 maxima values in the evening, at 6 p.m. (27 $\mu\text{g}/\text{m}^3$) and 8 p.m. (23 $\mu\text{g}/\text{m}^3$). During the night and early
175 morning, the values were constant and low (15 $\mu\text{g}/\text{m}^3$). At the Dubrovnik airport, the PM₁₀ values
176 started to raise from 7 a.m., reaching maxima values at 5 a.m. (26 $\mu\text{g}/\text{m}^3$). PM_{2.5} values have similar
177 trends but at much lower values (maxima at 10 $\mu\text{g}/\text{m}^3$).

178

179 4.2. Average seasonal concentrations

180 When observing the average seasonal PM₁₀ values for the Zagreb-1 (Figure 5, left) measuring
181 station there is a trend in PM₁₀ values behaviour, during summers have the lowest PM₁₀ values
182 (approx. 15 $\mu\text{g}/\text{m}^3$), while during winters have the highest PM₁₀ values (maximum 39 $\mu\text{g}/\text{m}^3$ in
183 winter 2017). The same trend can be observed for PM_{2.5} at Zagreb PPI (Figure 5, right) measuring
184 station, with minimal value during summer (approx. 10 $\mu\text{g}/\text{m}^3$) and maximal during winter (36
185 $\mu\text{g}/\text{m}^3$).

186 When observing the average seasonal PM₁₀ values for the Osijek-1 measuring station (Figure 6)
187 the values during springs and summers in 2017 and 2018 were similar (approx. 20 $\mu\text{g}/\text{m}^3$), but there
188 was a great raise in the PM₁₀ values in fall 2018 (61 $\mu\text{g}/\text{m}^3$). The PM₁₀ values in 2019 were much
189 higher than in previous years, with the trend similar to city of Zagreb, low value during summer (33
190 $\mu\text{g}/\text{m}^3$) and high values during winter (77 $\mu\text{g}/\text{m}^3$), but the values were much higher compared to the
191 city of Zagreb.

192 When observing the average seasonal PM₁₀ and PM_{2.5} values for the Slavonski Brod (Figure 7),
193 it can be observed that the lowest values appeared during summer (approx. 20 $\mu\text{g}/\text{m}^3$ for PM₁₀ and
194 approx. 12 $\mu\text{g}/\text{m}^3$ for PM_{2.5}) and the highest values during winter (58 $\mu\text{g}/\text{m}^3$ for PM₁₀ in 2017 and 62
195 $\mu\text{g}/\text{m}^3$ for PM_{2.5} in 2017). This trend is similar to the PM trend present at the city of Zagreb.

196 When observing the average seasonal PM₁₀ and PM_{2.5} values for the Rijeka-2 measuring station
197 (Figure 8, left) it can be seen that the highest PM₁₀ values were in spring (20 $\mu\text{g}/\text{m}^3$) and summer (23
198 $\mu\text{g}/\text{m}^3$) in 2017 and spring (19 $\mu\text{g}/\text{m}^3$) and fall (31 $\mu\text{g}/\text{m}^3$) in 2018. During 2017 PM_{2.5} values were the
199 lowest during summer (8 $\mu\text{g}/\text{m}^3$) and the highest during winter (14 $\mu\text{g}/\text{m}^3$), while in 2018 the PM_{2.5}
200 values were approximately the same, at 11 $\mu\text{g}/\text{m}^3$.

201 When observing the average seasonal PM₁₀ and PM_{2.5} values during 2019 for the Dubrovnik
202 airport (Figure 8, right) measuring station it can be seen that the highest PM₁₀ values were during
203 spring (27 $\mu\text{g}/\text{m}^3$) with decreasing tendency up to the winter (4 $\mu\text{g}/\text{m}^3$). The highest PM_{2.5} values
204 were obtained during summer (12 $\mu\text{g}/\text{m}^3$) while the lowest values were during winter (3 $\mu\text{g}/\text{m}^3$).
205 Spring and summer obtained higher PM₁₀ and PM_{2.5} values caused by the intensive touristic arrivals
206 and heavier air traffic during these months.

207

208 5. Conclusions

209 Average hour concentrations of PM emissions analysis showed that there is a difference between
210 PM emissions in the capital city of Zagreb and other urban areas in Croatia, with the highest PM₁₀
211 emissions during early morning and later evening.

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

213 Heavy traffic, commuting and border crossing at international border crossing in Slavonski Brod
214 between EU and other Balkan countries seems to influence the average hour concentrations of PM
215 emission, with highest values during late night hours. At Slavonski Brod the PM_{2.5} and PM₁₀
216 emissions are correlated and have the same daily tendency.

217 Coastal cities, Rijeka and Dubrovnik have the lowest emission of PMs. As expected, at the
218 Dubrovnik airport the heavier air traffic raises the PM₁₀ during working hours, from 7 a.m. to 7 p.m.
219 PM_{2.5} has the same tendency, but with much lower emission values.

220 Cities in the continent obtained higher seasonal PM emission vales during fall and winter
221 months, compared to the coastal cities. The lower PM₁₀ and PM_{2.5} vales during fall and winter
222 months for the coastal cities are due to milder (sub)mediterranean climate and reduced amount of
223 fossil fuel consumption during these months since electricity is the primary source for heating.

224 It can be noted that at the coastal cities, the average PM_{2.5} emissions were always much lower
225 than average PM₁₀ emissions, regarding the 24-period or seasonal period.

226

227 **Author Contributions:** Conceptualization, N.S. and B.Đ.; methodology, M.H. and N.S.; formal analysis, A.P.S.
228 and M.H.; writing—original draft preparation, N.S. and B.Đ.; writing—review and editing, N.S. and B.Đ.;
229 visualization, A.P.S. and M.H.; supervision, N.S. All authors have read and agreed to the published version of
230 the manuscript.

231 **Acknowledgments:** We thank the Croatian Meteorological and Hydrological Service for providing us the useful
232 data and professional help.

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

234 References

- 235 1. WHO, "7 million premature deaths annually linked to air pollution," Geneva, 2014.
- 236 2. Cohen, A.J.; Brauer, M.; Burnett, R.; Anderson, H.R.; Frostad, J.; Estep, K.; Balakrishnan, K.; Brunekreef,
237 B.; Dandona, L.; Dandona, R.; Feigin, V.; Freedman, G.; Hubbell, B.; Jobling, A.; Kan, H.; Knibbs, L.; Liu,
238 Y.; Martin, R.; Morawska, L.; Pope, C.A.; Shin, H.; Straif, K.; Shaddick, G.; Thomas, M.; van Dingenen,
239 R.; van Donkelaar, A.; Vos, T.; Murray, C.J.L.; Forouzanfar, M.H. Estimates and 25-year trends of the
240 global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden
241 of Diseases Study 2015. *Lancet* **2017**, *389*, 1907–1918. DOI: 10.1016/S0140-6736(17)30505-6
- 242 3. Garbariene, I.; Kvietkus, K.; Šakalys, J.; Ovadnevaite, J.; Čeburnis, D. Biogenic and anthropogenic
243 organic matter in aerosol over continental Europe: Source characterization in the east Baltic region *J.*
244 *Atmos. Chem.* **2012**, *69*, 159–174. DOI: 10.1007/s10874-012-9232-7
- 245 4. Janssen, N.A.H.; Fischer, P.; Marra, M.; Ameling, C.; Cassee, R. Short-term effects of PM_{2.5}, PM₁₀ and
246 PM_{2.5-10} on daily mortality in the Netherlands *Sci. Total Environ.* **2013**, *463*, 20–26. DOI:
247 10.1016/j.scitotenv.2013.05.062
- 248 5. Brunekreef, B. Epidemiological evidence of effects of coarse airborne particles on health *Eur. Respir. J.*
249 **2005**, *26*, 309–318. DOI: 10.1183/09031936.05.00001805
- 250 6. Zanobetti, A.; Schwartz, J. The Effect of Fine and Coarse Particulate Air Pollution on Mortality: A
251 National Analysis *Environ. Health Perspect.* **2009**, *117*, 898–903. DOI: 10.1289/ehp.0800108

- 252 7. Sarkodie, S.A.; Strezov, V.; Jiang, Y.; Evans, T. Proximate determinants of particulate matter (PM2.5)
253 emission, mortality and life expectancy in Europe, Central Asia, Australia, Canada and the US *Sci. Total*
254 *Environ.* **2019**, *683*, 489–497.
- 255 8. WHO, “Global Health Observatory (GHO) data,” 2016.
- 256 9. Amaral, S.; de Carvalho, J.; Costa, M.; Pinheiro, C. An Overview of Particulate Matter Measurement
257 Instruments *Atmosphere (Basel)*. **2015**, *6*, 1327–1345.
- 258 10. WikimediaCommons, “EU-Croatia.” [Online]. Available: [https://commons.wikimedia.org/wiki/File:EU-](https://commons.wikimedia.org/wiki/File:EU-Croatia.svg)
259 [Croatia.svg](https://commons.wikimedia.org/wiki/File:EU-Croatia.svg). [Accessed: 05-Oct-2020].
- 260 11. GoogleMaps, “GoogleMaps.” [Online]. Available:
261 [https://www.google.com/maps/place/Croatia/@44.4247999,14.1637907,7z/data=!3m1!4b1!4m5!3m4!](https://www.google.com/maps/place/Croatia/@44.4247999,14.1637907,7z/data=!3m1!4b1!4m5!3m4!1s0x133441080add95ed:0xa0f3c024e1661b7f!8m2!3d45.1!4d15.2000001)
262 [1s0x1334410](https://www.google.com/maps/place/Croatia/@44.4247999,14.1637907,7z/data=!3m1!4b1!4m5!3m4!1s0x133441080add95ed:0xa0f3c024e1661b7f!8m2!3d45.1!4d15.2000001)
263 [80add95ed:0xa0f3c024e1661b7f!8m2!3d45.1!4d15.2000001](https://www.google.com/maps/place/Croatia/@44.4247999,14.1637907,7z/data=!3m1!4b1!4m5!3m4!1s0x133441080add95ed:0xa0f3c024e1661b7f!8m2!3d45.1!4d15.2000001). [Accessed: 05-Oct-2020].
- 264



© 2020 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).