Influence of lockdown caused by the COVID-19 pandemic on air pollution and carcinogenic content of particulate matter observed in Croatia

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Abstract
Due to the pandemic of SARS-CoV-2 in Croatia, all unnecessary activities were prohibited during the designated lockdown period (March–May 2020). With reduced human activity, levels of some air pollutants decreased. In this study, mass concentrations of the PM1 particle fraction (particulate matter with an equivalent aerodynamic diameter < 1 μm) and polycyclic aromatic hydrocarbons (PAHs) in PM1 and NO2 were measured and compared with concentrations measured in the same period the year before. Air pollutant concentrations were measured at two measuring sites: urban residential and urban traffic. Our results show a concentration decrease by 35% for NO2 and PM1 particles and by 26% for total PAHs at the traffic measuring site. At the residential measuring site, only concentrations of NO2 decreased slightly, but PM1 particles and PAHs were similar to the year before.

Keywords Air pollution · COVID-19 pandemic · PAHs · PM1 · NO2

Introduction
At the end of 2019, the coronavirus (COVID-19) pandemic emerged in China (Tian et al. 2020; Wang et al. 2020). The first case in Croatia was detected on 25 February 2020 and after that, the number of cases rapidly increased (www.koronavirus.hr). The pandemic caused over 16.8 million global infections and thus far more than half of million deaths (28th July 2020) (https://covid19.who.int/). At the moment of writing this paper, the number of registered infections by COVID-19 in Croatia was 4993 and deaths 141. To prevent the rapid increase of COVID-19 infections, at the beginning of the epidemic, the Civil Protection Directorate of Croatia required people to stay at home. Every school and faculty in the country was closed beginning from 15 March until the end of the school year (20 June). Public transport between and within cities was suspended as well. Only a week later, all human activities were reduced to only the bare minimum, resulting in a significant reduction in the number of vehicles on the streets. This “lockdown” period lasted until 15 May 2020. The last week before the end of “lockdown”, the number of daily infection was minor.

Vehicle exhaust emissions are the most significant source of urban NO2 and a significant source of PM1 particles. PM1 particles are particles with an aerodynamic diameter smaller than 1 μm and are considered to be of importance in the context of adverse health effects induced by particulate pollution (Wenger et al. 2009). However, measurements of PM1 and its content are not part of routine air quality monitoring, although they may contain significant amounts of harmful compounds. Polycyclic aromatic hydrocarbons (PAHs) usually occur as complex mixtures, produced mostly from natural and anthropogenic incomplete combustion processes (ATSDR 1995; Lee 2010). High PAH levels in ambient air of urban cities are usually associated with traffic as well as household heating (Ströher et al. 2007; Ravindra et al. 2008).

In Croatia, this pollution attracts much attention especially during winter, when the meteorological conditions are unfavourable (Jakovljević et al. 2018; Pehnec et al. 2016). McMahon (2020) reported that “lockdown” reduced the level of air pollution in China. The European Space Agency (2020) also reported a significant decrease level of nitrous oxide emissions in Northern Italy during the “lockdown” period. Because of the COVID-19 lockdown, human activities reduced up to 90%, and environmental pollution reduced by
about 30% in Spain, USA, Italy and Wuhan (Muhammad et al. 2020).

Some studies reported increasing pollution levels during the COVID pandemic comparing with the same period last year (Faridi et al. 2020; Mohd Nadzir et al. 2020).

Most of the studies related to the “lockdown’s” influence on air pollution are focused on “classic” pollutants such as NO2, CO, SO2, PM2.5 and PM10 (Anil and Alagha 2020; Shakoor et al. 2020; Bilal Bashir et al. 2020; Pata 2020). This work intends to focus on the levels of some additional pollutants such as particulate-bound carcinogenic PAHs and particle fraction PM1. Air quality changes due to reduced human activity during the COVID-19 pandemic in Croatia were investigated. The levels of PM1 and PAHs in PM1 and NO2 were quantified and compared with the same period in the year before at two different sites in Zagreb, the Croatian capital.

Material and methods

Daily average concentrations of PM1, NO2 and PAHs in PM1 were measured in Zagreb during the COVID-19 lockdown period (March–May) and compared with daily concentrations for the same period in the previous year. Measuring station A was located in the northern, residential part of Zagreb. A road with modest traffic density, which contain two lanes in both directions (north-south and south-north), was about 50 m away. Measuring station B was located near the historical city centre of Zagreb. The nearby street is 15 m wide and 495 m long and the location was surrounded by buildings approximately 27 m high. Zagreb has a population of around 790,000 inhabitants and over 350,000 registered automobiles. The positions of the measuring sites are shown in Fig. 1.

Twenty-four-hour PM1 samples were collected every day during the lockdown period (March–May). PM1 particle fraction was collected on quartz filters with a low-volume Sven Leckel sampler (55 m3). Concentrations of PM1 fractions were determined gravimetrically (Mettler Toledo MX-5 micro balance). For PAH determination, filters were extracted with a solvent mixture of toluene and cyclohexane; the procedure of extraction was described in Jakovljević et al. (2015). PAH analysis was performed using an Agilent Infinity high-performance liquid chromatography (HPLC) with a fluorescence detector. The following PAHs were determined: fluoranthene (Flu), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chry), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), dibenzo(ah)anthracene (DahA), benzo(ghi)perylene (BghiP) and indeno(1,2,3,cd)pyrene (IP).

The same procedure was applied to the samples collected the year before. The measurements were conducted within the internal scientific project “Organic content of PM1 particle fraction” funded by the Institute for Medical Research and Occupational Health (PI: R. Godec). Measurements of NO2 are part of the local air quality monitoring programme funded by the City of Zagreb, City Office for Economy, Energetics and Environment Protection. NO2 concentrations were determined using an automatic device (Horiba APNA-370, Kyoto, Japan) based on chemiluminescence, according to the EN 14211:2012 standard.

Results and discussion

Daily variations of NO2, PM1, total (ΣPAH) and individual PAHs during the COVID-19 lockdown period (March–May) were compared with concentrations of pollutants in the same period the year before (Figs. 2 and 3).

The results show that the lockdown differently affected the air quality at both measuring sites. The effect was more pronounced at traffic site B, where the concentrations of all measuring pollutants were much lower during the COVID-19 lockdown than the year before.

For NO2, the decrease was evident at both locations but more pronounced at traffic site B. At measuring site A, the average NO2 concentration in the 2019 period was 15 μg m⁻³, while in 2020, it was slightly lower (11 μg m⁻³). At site B, the concentrations of NO2 were about 35% lower (Fig. 2) than the same period in the year before, with average values of 22 μg m⁻³ and 34 μg m⁻³, respectively. Similarly, PM1 levels at site A were only slightly lower than the year before (average values 9 μg m⁻³ and 10 μg m⁻³, respectively). At site B, the concentrations of PM1 during the COVID-19 lockdown were significantly lower (by 35%) compared with the year before (average mass concentrations 7 μg m⁻³ and 12 μg m⁻³, respectively).

Concentrations of NO2 in urban areas are primarily linked to road traffic, especially motor vehicle exhaust. However, particulate matter may originate from different sources. One of the sources is regular road cleaning by manual dry sweeping. Also, uncontrolled construction around the city could be a substantial contributor to particulate matter. However, in urban areas, the PM1 fraction is often linked to traffic (Agudelo-Castañeda and Teixeira 2014). During the COVID-19 lockdown in Croatia, traffic and all other activities were reduced to a minimum, which is the reason for lower NO2 and PM1 concentrations at the traffic site.

Recent studies comparing air pollutant levels before and during the lockdown found that lockdown measures had a large influence at NO2 levels (Anil and Alagha 2020; Gautam 2020; Sarfraz et al. 2020). In the study of Anil and Alagha (2020) in Saudi Arabia as well as in the study of Wang et al. (2020) in Beijing, a much higher decrease in NO2 concentrations was observed comparing with this study. In Saudi
Arabia, lockdown measures resulted in NO$_2$ decreased by 12–86%, while PM$_{10}$ concentrations decreased between 21 and 70%.

Significantly reduced environmental pollution was also reported for three cities in central China, comprising 60% lower concentrations of NO$_2$ and 30% lower concentrations of PM$_{2.5}$ particles during the lockdown (Masum and Pal 2020). Another study found 26–49% lower NO$_2$ concentrations in six cities in China and 29–61% in some states and provinces of the USA comparing with the first quarter of 2019 (Shakoor et al. 2020). The same investigation showed a reduction of PM$_{2.5}$ concentrations by 7–27% and 11–31%, as well as reduction of PM$_{10}$ by 23–79% and 20–25% in China and the USA, respectively. Similarly reduced air pollution (NO$_2$) was observed in Italy and Spain (30%) (Muhammad et al. 2020). The European Environment Agency reported a decrease in NO$_2$ concentrations in many European cities where lockdown measures were implemented. Although a decrease in concentrations of fine particulate matter (PM$_{2.5}$) may also be expected, a consistent reduction cannot be seen across European cities, mostly due to the large variety of particulate matter sources (fuel combustion for residential heating, commercial and institutional buildings, industrial activities, secondary aerosol formation) and factors, such as weather conditions (EEA 2020). However, less is known about the smaller particle fractions such as PM$_1$ and especially their content. According to the best of our knowledge, this study is the first to report PM$_1$ and PM$_{1}$-bound PAHs during the lockdown period and analyse the possible influence of measures taken.

In this study, all of the measured PAHs in the PM$_1$ particle fraction at the urban residential site A were similar or slightly higher during the pandemic; however, the difference was not significant (Fig. 3). These results indicated that the concentration of PAHs in PM$_1$ particles originated from sources other than traffic. A previous study at the same locations showed that PAHs in PM$_1$ particles in this part of the year (heating period) originated mostly from house heating and wood burning than from traffic, which is probably the main reason why the pandemic lockdown did not affect the PAH concentrations levels at this measuring sites (Jakovljević et al. 2018). At site B, the PAH concentrations decreased between 17% for Pyr and 40% for IP (Fig. 3). Total PAH mass concentrations for the lockdown period were lower by 26% compared with the same period in 2019.
Contributions of the mass of individual PAH to the total PAH mass and to the PM$_1$ mass were calculated as well (Figs. 4 and 5). Comparing the mass contributions of individual PAHs to the total PAHs (Fig. 4), it is evident that they were almost identical during the lockdown period and during the same period the year before. Changes in individual contributions were within 1.5% for all PAHs at both locations. The mass contributions of individual and total PAHs to the PM$_1$ (Fig. 5)
at traffic site B were almost identical in 2020 and 2019 (ΣPAH/PM$_1$ mass ratio $35.4 \times 10^{-3}$ and $35.6 \times 10^{-3}$, respectively). However, at urban residential site A, the mass contribution of each individual PAH was slightly higher during the lockdown period (between 0.1 and 1.1%), which caused an increase of total PAH contribution during the lockdown from $27.8 \times 10^{-3}$ to $35.2 \times 10^{-3}$ compared with the same period in 2019.

This worldwide lockdown period provided us with a good chance to understand our pressure on nature and the environment and to assess we can recover the quality of our environment. The results of this study will therefore be helpful for environmental strategies giving knowledge of how reduced traffic emissions could influence ambient levels of air pollutants.

**Fig. 3** Average mass concentrations of PAHs from March to May 2019 and 2020 (COVID-19 pandemic) for measuring sites A (urban residential) and B (traffic)

**Fig. 4** Individual PAH contributions to the ΣPAH from March to May 2019 and 2020 (COVID-19 pandemic) for measuring sites A (urban residential) and B (traffic)

**Fig. 5** Individual PAH contributions to the PM$_1$ mass from March to May 2019 and 2020 (COVID-19 pandemic) for measuring sites A (urban residential) and B (traffic)
Conclusions

In this study, the influence of the “lockdown” caused by the COVID-19 pandemic on air quality was analysed at two locations in Zagreb, Croatia. Measures conducted during the lockdown period differently affected levels of air pollutants at the traffic and urban residential site. Reduced traffic and other lockdown measures caused a decrease of 35% for NO₂ and PM₁ at traffic location compared with the same period the year before. At the residential site, the concentrations of NO₂, PM₁ and total PAHs were similar or only slightly lower than the year before.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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