1 What can we learn about urban air quality

with regard to the first outbreak of the COVID-19 pandemic? A case study from Central Europe

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Abstract. Motor vehicle road traffic in central Budapest was reduced by approximately 50 % 10 of its ordinary level for several weeks as a consequence of various limitation measures 11 introduced to mitigate the first outbreak of the COVID-19 pandemic in 2020. The situation was 12 utilised to assess the real potentials of urban traffic on air quality. Concentrations of NO, NO₂, 13 CO, O₃, SO₂ and particulate matter (PM) mass, which are ordinarily monitored in cities for air 14 quality considerations, aerosol particle number size distributions, which are not rarely 15 measured continuously on longer run for research purposes and meteorological properties 16 usually available were collected and jointly evaluated in different pandemic phases. The largest 17 changes occurred over the severest limitations (partial lock-down in the Restriction phase from 18 28 March to 17 May 2020). Concentrations of NO, NO₂, CO, total particle number (N₆₋₁₀₀₀) 19 and particles with a diameter <100 nm declined by 68, 46, 27, 24 and 28 %, respectively in 20 2020 with respect to the average reference year of 2017–2019. Their quantification was based 21 on both relative difference and standardised anomaly. The change rates expressed as relative 22 concentration difference due to relative reduction in traffic intensity for NO, NO₂, N₆₋₁₀₀₀ and 23 CO were 0.63, 0.57, 0.40 and 0.22 (%/%), respectively. Of the pollutants which reacted in a 24 sensitive manner to the change in vehicle circulation, it is the NO₂ that shows the most frequent 25 exceedance of the health limits. Intentional tranquillizing of the vehicle flow has considerable 26 27 potentials in improving the air quality. At the same time, the concentration levels of PM₁₀ mass, 28 which is the most critical pollutant in many European cities including Budapest, did not seem to be largely affected by vehicles. Concentrations of O₃ concurrently showed an increasing 29 tendency with lower traffic, which was explained by its complex reaction mechanism. 30 Modelling calculations indicated that spatial gradients of NO and NO₂ within the city became 31 32 further enhanced by reduced vehicle flow.

33 **1 Introduction**

The coronavirus disease (COVID-19) is caused by the novel, Severe Acute Respiratory 34 Syndrome CoronaVirus 2 (SARS-CoV-2) virus. The outbreak was declared as a pandemic by 35 the WHO on 11 March 2020 (WHO, 2020). National governments, international agencies and 36 organisations enacted widespread emergency actions for individuals, some professionals, 37 communities and the public to reduce the risk of infection and to combat the plague. As a 38 consequence of the implemented measures, road traffic in many cities worldwide was reduced 39 40 in a substantial manner and for a considerable time interval. In parallel, lower concentrations of several air pollutants were reported from both satellite observations and in situ 41 42 measurements (Keller et al., 2020; Lal et al., 2020; Le et al., 2020; Lee et al. 2020; Mahato et al., 2020; Nakada and Urban, 2020; Petetin et al., 2020; Tobías et al., 2020; Wang et al., 2020). 43 44

This situation offers a unique possibility for atmospheric scientists to investigate experimentally some important atmospheric chemical and physical issues including urban air quality and climate change under extraordinary conditions of lower traffic and industrial productivity (Sussmann and Rettinger, 2020). The results and consequences of this real "ambient experiment" can be utilised to determine the true potentials of action plans on tranquillizing urban road circulation for handling air quality, overcrowding, traffic congestions, noise contamination and other environmental, health and climate impacts in large cities.

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The task is, however, somewhat complicated. Actual concentrations of atmospheric 53 constituents can depend on 1) their emissions from several sectors, 2) their physical removal 54 processes, 3) local meteorological conditions mainly precipitation (P), wind speed (WS), 55 planetary boundary layer height (PBLH) and atmospheric stability, 4) their (long-range) 56 transport and 5) possible photochemical reactions, which are largely influenced by other 57 meteorological properties such as global solar radiation (GRad), relative humidity (RH) and 58 air temperature (T), and by availability of and interactions with other chemical species present 59 in the air. Many of the phenomena or properties listed are, in addition, interconnected and 60 61 confound, which further obscures the situation since they create an internally interacting 62 environmental system.

63

Tropospheric residence time of constituents can also play a role under non-steady-state
 conditions (Harrison, 2018). As a result, atmospheric concentrations at a fixed site change both

66 periodically and randomly (fluctuate) on daily, seasonal or annual scales. The variations are

- also linked to the geographical location and features of urban sites (de Jesus et al., 2019).
- 68

Source-specific markers generated by internal combustion engines or added on purpose into 69 their fuel (e.g. Horvath et al., 1988; Gentner et al., 2017) or multivariate statistical methods 70 71 (Hopke, 2016) can be applied to estimate the importance of vehicle traffic for air quality. These 72 methods usually require advanced analytical methods to obtain data for specific species, which may not be available with a required time resolution, or need a larger number of data, which 73 74 can be constrained by duration of the time intervals of interest. Another possibility is to examine jointly the time series of multicomponent atmospheric data sets. This approach 75 (described later in more detail) can be utilised retrospectively and it is generally applicable in 76 different cities in the world, which were affected by road traffic restrictions. 77

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79 In Hungary, state of emergency was introduced on 11 March 2020. It involved sequential closure of education institutes, beginning of work-from-home and social distancing. It was 80 81 followed by restrictions on movement. During this, residences could only be left with specified basic purposes, administrative centres, restaurants and touristic places were closed, distant 82 83 travels were ceased, public parks were closed for long weekends and there were various time limitations on shopping. The mitigating measures resulted in perceivable changes in vehicular 84 85 road traffic and atmospheric concentrations. The main objectives of the present paper are 1) to introduce and demonstrate a general method for quantifying concentration changes, 2) to 86 evaluate whether the changes observed were related to motor vehicle road traffic, 3) to assess 87 the effect of traffic on these alterations, and 4) to estimate and debate the potentials of 88 tranquillized urban vehicle flow on the air quality. 89

90 **2 Methods**

91 Criteria air pollutants, namely NO, NO₂=NO_x-NO, CO, O₃, SO₂ and particulate matter (PM) 92 mass in various size fractions were involved in the study. The species originate from different 93 sources. Vehicular road traffic is usually associated with NO and CO, while NO₂ and O₃ are 94 formed by chemical reactions in the air. Contributions of residential heating, cooking, industrial 95 activities, regional traffic in winter and secondary processes to PM_{2.5} mass are of large 96 importance in many cities, including Budapest. At the same time, PM₁₀ mass represents 97 disintegration sources, e.g. windblown soil, crustal rock, mineral and roadside dust, resuspended dust by car movement, agricultural activities in the region, construction work and
material wear such as tire abrasion of cars at kerbside sites (Salma and Maenhaut, 2006; Putaud
et al., 2010; Harrison et al., 2012; Salma et al., 2020a). They all can be important particularly
under dry weather conditions.

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103 Aerosol particle number concentrations in the diameter ranges from 6 to 1000 nm (N_{6-1000}) and from 6 to 100 nm (N_{6-100}) are mainly assigned to high-temperature emission sources (such as 104 vehicle road traffic or incomplete burning) and atmospheric new particle formation and growth 105 106 (NPF) events (Paasonen et al., 2016; Rönkkö et al., 2017; Salma et al., 2017). The latter process occurs as a daily phenomenon with a typical shape of its monthly occurrence frequency (Salma 107 and Németh, 2019). This distribution changes in Budapest from year to year without any 108 tendentious character (Salma et al., 2020b). Particles with a diameter from 25 to 100 nm (N_{25-} 109 100) in cities are mainly emitted by incomplete combustion or consist of grown new particles 110 by condensation, while the size fraction with a diameter from 100 to 1000 nm ($N_{100-1000}$) 111 expresses physically and chemically aged particles, thus, they represent larger spatial extents 112 113 (Salma et al., 2014; Mikkonen et al., 2020).

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Approximate tropospheric residence time of NO_x, CO, O₃, SO₂ and PM are estimated to 1–2
days, 2 months, 1–2 months, 4–12 days and from several hours up to 1 week depending largely
on particle size and chemical composition, respectively (Warneck and Williams, 2012;
Harrison, 2018).

119 2.1 Experimental data

120 The concentrations of NO/NO_x, CO, O₃, SO₂, PM₁₀ mass and PM_{2.5} mass were measured by chemiluminescence (Thermo 42C), IR absorption (Thermo 48i), UV fluorescence (Ysselbach 121 122 43C), UV absorption (Ysselbach 49C) and beta-ray attenuation (two Environment MP101M instruments with PM₁₀ and PM_{2.5} inlets) methods, respectively with a time resolution of 1 h. 123 The concentrations of gases were expressed at a temperature of 293 K and pressure of 101.3 124 kPa. The particle number concentrations were determined by a flow-switching type differential 125 126 mobility particle sizer (DMPS; Salma et al., 2016b) with a time resolution of 8 min. The latter measurements were performed in a diameter range from 6 to 1000 nm in 30 size channels with 127 equal width in the dry state of particles. The meteorological data of T, RH and WS and of GRad 128

were measured by standardised sensors (HD52.3D17, Delta OHM, Italy, and SMP3 pyranometer, Kipp and Zonen, the Netherlands, respectively) with a time resolution of 1 min.

The DMPS and meteorological measurements were accomplished at the Budapest platform for Aerosol Research and Training (BpART) Laboratory (N 47° 28' 29.9", E 19° 3' 44.6", 115 m above mean sea level) of the Eötvös University (Fig. 1). The location represents a well-mixed, average atmospheric environment for the city centre due to its geographical and meteorological conditions (Salma et al., 2016a). The local emissions include diffuse urban traffic exhaust, household/residential emissions and limited industrial sources together with some off-road transport (Salma et al., 2020a). In some time intervals, long-range transport of air masses can also play a role.

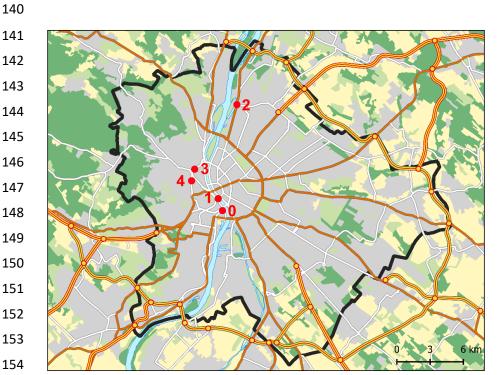


Figure 1. Location of the measurement sites in Budapest. 0: BpART Laboratory, 1: Szabadság Bridge, 2: Váci Road, 3: Széna Square and 4: Alkotás Road. The border of the city (in black colour), Danube River and the major routes are also indicated.

The data of the criteria air pollutants were acquired from a measurement station of the National Air Quality Network at Széna Square (Fig. 1) located in 4.5 km from the BpART Laboratory in the upwind-prevailing direction (Salma and Németh, 2019). This station serves as a reference for our long-term air quality-related research activities in several aspects and proved to be acceptable for this purpose.

Atmospheric transport of chemical species was assessed through large-scale weather types. We 165 utilised macrocirculation patterns (MCPs), which were invented specifically for the Carpathian 166 Basin (Péczely, 1957; Maheras et al., 2018). The classification of the MCPs is based on the 167 position, extension and development of cyclones and anticyclones relative to the Carpathian 168 169 Basin considering the sea-level pressure maps constructed for 00:00 UTC in the North-Atlantic-European region on a daily basis. A brief survey on the MCPs and the actual codes 170 for year 2020 utilised in the interpretations are given in Table S1 and Fig. S1, respectively in 171 172 the Supplement. The relative occurrences of the weather types in year 2020 were roughly in line with multiple-year frequencies. Extended anticyclonic weather types usually indicate that 173 the air masses are stagnant, and that the importance of local or regional sources prevail over 174 the air transport from distant sources. Under cyclonic weather conditions and frontal systems, 175 the transported air masses can yield more pronounced effects and contributions. 176

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Census of motor road vehicles was performed on three major routes and on a bridge over the 178 179 Danube River by the Budapest Public Roads Ltd. The measurement sites were on Szabadság Bridge, Váci Road, around Széna Square and Alkotás Road (Fig. 1), which are described in 180 181 more detail in the Supplement. The counting was based on permanent electronic devices with inductive loops and passenger cars, high- and heavy-duty vehicles and buses were recorded in 182 both directions. The time resolution of the data was 1 h and their coverage was >90 % of all 183 possible item in a year. The sites cover a wide range of maximum hourly mean vehicle flow 184 185 from about 1200 to 4600 h⁻¹. Szabadság Bridge has the smallest traffic intensity of the sites, but it proved to be a very valuable microenvironment for the study since it is part of the internal 186 boulevard. The routes showed coherent and common aggregate time properties and, therefore, 187 their data are to be proportional to general vehicular traffic flow in the city centre. 188

189 **2.2 Time intervals of interest**

Time intervals from 1 January to 31 July in 2017, 2018, 2019 and 2020 were studied. This included all major measures related to the first outbreak of the Covid-19 pandemic in Budapest in 2020. Within these seven months, five consecutive time intervals were selected for comparative purposes: 1) from 1 January till the beginning of the state of emergency at 15:00 on 11 March, which is referred as Pre-emergency phase, 2) from the beginning of the state of emergency to 27 March (till the beginning of the restriction on movement), which is called here Pre-restriction phase, 3) from the beginning of the restriction on movement till its end in
Budapest on 17 May, which is denoted as Restriction phase, 4) from the end of the restriction
on movement in Budapest till the end of the state of emergency on 17 June, which is referred
as Post-restriction phase and 5) from the end of the state of emergency till 31 July, which is
called Post-emergency phase. An overview on the pandemic phases with further details of
possible relevance for air quality issues is summarised in Fig. S1. Equivalent time intervals in
years 2017–2019, which correspond to these phases were considered for comparative purposes.

Local daylight saving time (LDST=UTC+1 or UTC+2) was chosen as the time base for the atmospheric concentrations and road traffic data because it was observed that the daily activity time patterns of inhabitants largely influences these variables in cities (Salma et al., 2014). The meteorological data were expressed in UTC+1 since their diurnal and seasonal behaviours are primarily controlled by sun path and other natural processes.

209 **2.3 Data treatment and modelling**

Medical studies with the influenza virus indicated that absolute humidity (AH) constrains both transmission efficiency and virus survival more than RH (Shaman and Kohn, 2009). In order to facilitate the future comparison with other locations or cities in the world mainly for possible virology purposes, the hourly mean RH values (%) were converted to AH (g m⁻³) using a calculation recommended by WMO (2008):

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$$AH = \frac{e(T_0) \times \exp(A \times \frac{T}{T+B}) \times RH \times C}{T+273.15},$$
 (1)

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where *T* is expressed in °C, $e(T_0)=6.112$ hPa is the saturation vapour pressure at $T_0=0$ °C, A=17.67, B=243.5 °C and C=2.167. For air temperatures <0 °C, we used an approximation for sub-cooled liquid water and adopted identical coefficients. This seems to be a plausible approach since the saturation vapour pressure curves for liquid water and ice surface follow each other closely near the freezing point. The AH values are summarised in Table S2, while we keep evaluating the RH because it seems to be more relevant for the purpose of this atmospheric study than the former property.

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Vertical transfer of gases and aerosol particles emitted or generated at the Earth surface canlargely be affected by the dynamics of the PBLH. It is realised by the dilution of pollutants

with mixing. The PBLH data were obtained from the 5th generation of the European Center of
Medium-Range Weather Forecasts (ECMWF) atmospheric reanalysis (ERA5) database using
Copernicus Climate Change Service (C3S, 2017). The ERA5 combines the modelled data of
the ECMWF's Integrated Forecast System, version CY41R2 on 137 hybrid sigma vertical
levels with newly available observations assimilated at every hour. In the present study, the

 $\label{eq:233} \mbox{ daily maximum PBLH values (PBLH_{max}) were considered to be proportional to the volume of }$

the mixed air parcel.

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236 The data with a time resolution of smaller than 1 h were averaged for 1 h. The coverage of the hourly data was typically above 90 % of all items in each year. Descriptive statistics, thus 237 count, minimum, median, maximum, geometric mean with standard deviation (SD) of all 238 variables were derived for the time interval studied and its each pandemic phase in year 2020 239 (Y2020). The characteristics were compared to the corresponding data in an average reference 240 year (Y3Ref). This contains averages of the parallel hourly mean data of the years 2017–2019. 241 Longer time span than three years would not necessarily be advantageous since some chemical 242 species in Budapest show tendentious change on a scale of ten years (Mikkonen et al., 2020) 243 and the urban traffic could also change substantially. 244

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Comparative evaluations are often performed via the relative change (RDiff) of medians (*m*)derived for a selected time span, which can be described as

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249 RDiff =
$$\frac{m(Y2020) - m(Y3Ref)}{m(Y3Ref)}$$
. (2)

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In our case, the time spans considered were the intervals of the five pandemic phases in both Y2020 and Y3Ref. The quantity RDiff essentially expresses the ratio of medians. It is very important to stress immediately that the ratios are largely influenced by the absolute magnitude of variables and could be misleading if interpreted alone. In addition, different variables can have very different ranges of variability. A further metric that could, therefore, be involved is the standardised anomaly (SAly), which is described as

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$$SAly = \frac{m(Y2020) - m(Y3Ref)}{SD}$$
. (3)
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This quantity expresses the observed differences in units of SD, so it brings out the relative asset of the actual difference. For GRad, which evolves daily from their very low values overnight in a large number, which were not considered, the anomaly was not standardised to its (expanded) SD, but instead, it was calculated simply as a difference m(Y2020)-m(Y3Ref)in its absolute unit.

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A difference in the fluctuating and periodically varying data sets (see Sect. 1) over a pandemic phase in Y2020 was quantified to be significant with respect to the equivalent interval in Y3Ref if both their RDiff and SAly metrics were significant. The actual criteria adopted are specified and discussed in Sect. 3.5.

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Average diurnal variations of all variables for workdays and holidays over each pandemic phase in the average reference year 2017–2019 and year 2020 were calculated by selecting all individual data for a particular hour of day on workdays or on holidays over the time interval under evaluation and by averaging them.

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The spatial distributions of the chemical species of interest over the city during each pandemic 276 277 phase were modelled via the surface concentrations derived from the Copernicus Atmosphere 278 Monitoring Service (CAMS) with a grid resolution of 0.1°×0.1° in order to study their potential differences (CAMS, 2019). The reanalysed concentrations are based on the following state-of-279 the-art European models CHIMERE, EMEP, EURAD-IM, LOTOS-EUROS, MATCH, 280 MOCAGE and SILAM (Marécal et al., 2015). The modelled concentrations are represented by 281 282 the CAMS ensemble, which is the median of the available model results at each grid-point. The CAMS modelling shares the meteorological driver of the ECMWF's Integrated Forecast 283 284 System and the Monitoring Atmospheric Composition and Climate emission inventory of the 285 Netherlands Organization for Applied Scientific Research. The system provides daily 96-h 286 estimates with hourly outputs of several chemical species. The hourly analysis at the Earth surface is done a posteriori for the past day using a selection of air quality data from the 287 corresponding European monitoring stations. 288

289 3 Results and discussion

The changes in atmospheric concentrations are presented and interpreted after the effects of the confound variability in local meteorological conditions and in (long-range) transport of atmospheric air masses are evaluated and quantified.

293 **3.1 Meteorological conditions**

The hourly average meteorological data over the time interval considered were in line with 294 ordinary characteristics measured at the BpART Laboratory (Salma and Németh, 2019; 295 Mikkonen et al., 2020). The T in 2020 was colder by 0.4 °C than in the average reference year, 296 297 and the relative differences for median RH, WS, GRad and PBLH_{max} were -3, -8, +3 and +15 %, respectively. These alterations, except for the PBLH_{max}, are not significant (remained within 298 ± 10 %). There were, however, two important alterations from the multiple-years' weather 299 situations. First, spring 2020 was extraordinary dry; it was the third driest season since 1901. 300 301 This can likely be related to multifactorial meteorological reasons. Between 14 March and 24 302 April, anti-cyclonic weather types prevailed in the Carpathian Basin almost continuously for 41 days (Fig. S1). After this interval, the weather type was mostly cyclonic but with northerly 303 304 wind, which ordinarily brings dry and cold air masses to the Budapest area. These factors together resulted in the long and severe drought. Finally, it was followed by frequent, continued 305 306 and spatially extended rains in June (Fig. S1). Secondly, the number of foggy hours (160) in January 2020 was more than four times larger than in the average reference year. This 307 308 conclusion is based on measurements at the Budapest Liszt Ferenc International Airport.

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310 An overview on the major meteorological data during the whole state of emergency interval (98 days) is summarised in Table S2. The drought did not seem to influence substantially the 311 WS and GRad but affected considerably the RH and indirectly the PBLH_{max}. The alterations in 312 the PBLH_{max} in the average reference year and year 2020 over the pandemic phases are, 313 therefore, quantified separately in Table 1 and are also displayed in Fig. S2. The time series for 314 WS and T are also given in Figs. S3 and S4, respectively. It is seen in Fig. S2 that the Restriction 315 phase – which is of particular interest for this study – was influenced by the PBLH_{max} in a 316 more-or-less persistent manner without larger oscillations or fluctuations. The RDiff properties 317 are taken into consideration when quantifying the concentration changes (Sect. 3.5). Lastly, it 318 should also be mentioned that some of the differences in the meteorological data become small 319

or insignificant when comparing them to their uncertainty intervals (in particular, for the modelled $PBLH_{max}$).

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Table 1. Medians of the daily maximum planetary boundary layer height (km) in the average reference

year of 2017–2019 (Y3Ref) and year 2020 (Y2020) together with their relative difference (RDiff) in %
and their anomaly standardised to SD (SAly) over the five consecutive phases of the first COVID-19

- 326 outbreak.
- 327

	Pandemic phase	SAly	RDiff
Pre-emergency 0.66 0.88 $+32$ $+0.4$ Pre-restriction 1.4 1.4 $+1$ $+0.0$ Restriction 1.5 1.8 $+18$ $+0.5$ Post-restriction 1.6 1.3 -21 -0.7 Post-emergency 1.8 1.7 -8 -0.3	Pre-restriction	+0.0	+1
	Restriction	+0.5	+18
	Post-restriction	-0.7	-21

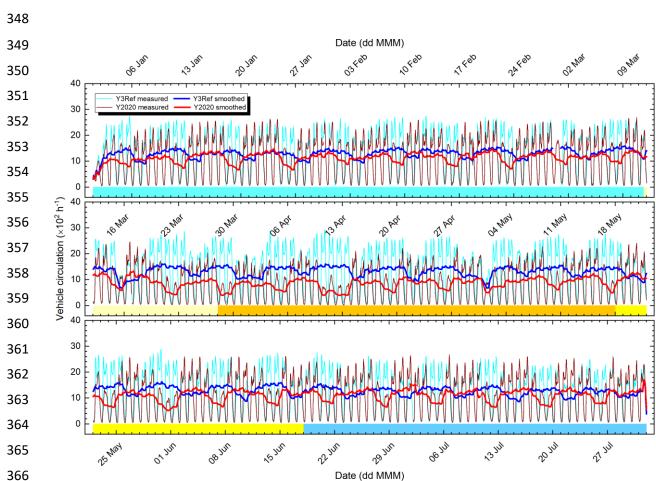
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329 **3.2 Motor vehicle road traffic**

Time series of vehicle flow on a major route (Váci Road, site no. 2 in Fig. 1) over the time
interval studied in the average reference year and year 2020 are shown in Fig. 2 as examples.
The other urban sites exhibited very similar time behaviour and tendencies.

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The time series for vehicle flow showed a clear periodicity. On each workday, two peaks -334 corresponding to the early morning and late afternoon rush hours – can be identified. In addition 335 to this periodicity, the smoothed curves also revealed an obvious cycling due to repeated 336 workdays and holidays sequence. More importantly, the time series implied that in the Pre-337 emergency pandemic phase, the road traffic in the city centre in Y2020 was very similar to that 338 in Y3Ref. The difference only appeared as a horizontal shift in time, which was caused by the 339 occurrence of holidays in the average reference year and year 2020. Two weeks before the 340 introduction of the restriction on movement, the vehicle circulation already started declining, 341 342 and in the last week of the Pre-restriction phase, it already reached the level observed later in 343 the Restriction phase. During these eight or nine weeks, the vehicular circulation on workdays was around the ordinary levels on holidays in 2017-2019. The circulation approached its 344 345 ordinary values within or after the first week of the Post-restriction phase step wisely. After



that, the curves for the two years were at almost identical levels again. The changes in thevehicle flow are quantified in Sect. 3.5 together with the pollutant concentrations.

Figure 2. Time series of motor vehicle circulation on a major route (Váci Road) in Budapest in both directions in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their 24-h smoothed curves over the five consecutive phases of the first COVID-19 outbreak. The phases are marked by the following colour codes: Pre-emergency phase lighter blue, Pre-restriction phase lighter yellow, Restriction phase orange, Post-restriction phase darker yellow and Post-emergency phase darker blue. The tick labels of the abscissa indicate the Mondays in 2020.

The shapes of the diurnal patterns (Fig. S5) for the average reference year and year 2020 were 374 375 similar to each other with some modifications. Differences could be identified in Pre-restriction and Restriction pandemic phases between 16:00 and 19:00, when the traffic flow on weekends 376 377 seemed to be systematically and in excess lower in 2020 than in the average reference year. This could be due to the limitations on shopping and to modified going out routines of 378 inhabitants under the restrictions. Similarly, the early morning peak on workdays in the Post-379 emergency (and partly in the Post-restriction) phases was smaller in excess in Y2020 than in 380 Y3Ref, which can likely be linked to less people going physically to work due to propagated 381

home-office jobs. To facilitate the comparison of diurnal patterns of vehicle circulation and of
atmospheric concentrations, the plot showing the diurnal variation of concentrations in the
Restriction phase was extended by the vehicle flow in the same pandemic phase (Fig. 7).

385 **3.3 Time series of concentrations**

Time series of NO, O₃, PM_{2.5} mass and N_{6-1000} atmospheric concentrations over the time interval studied are shown in Figs. 3–6, respectively. The chemical species selected represent primary pollutant gases, secondary pollutant gases and two different aerosol properties, respectively. The corresponding curves for NO₂, CO, SO₂, PM₁₀ mass and $N_{100-1000}$ are displayed in Figs. S6–S10, respectively.

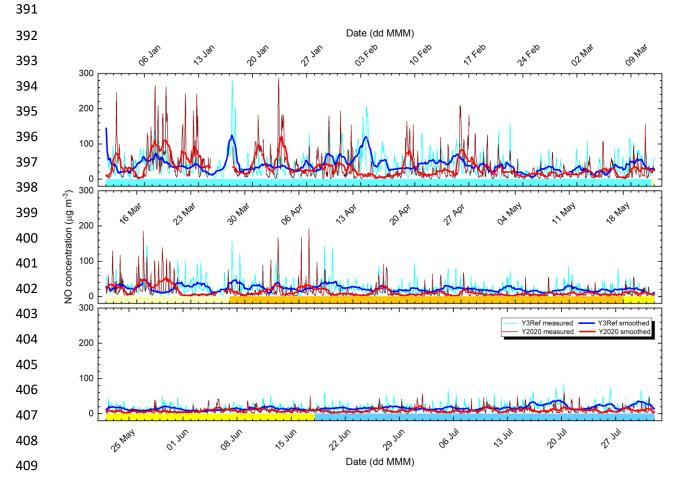


Figure 3. Time series of NO concentration in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their 24-h smoothed curves over the five consecutive phases of the first COVID-19 outbreak. The phases are marked by the following colour codes: Pre-emergency phase lighter blue, Pre-restriction phase lighter yellow, Restriction phase orange, Post-restriction phase darker yellow and Post-emergency phase darker blue. The tick labels of the abscissa indicate the Mondays in 2020.

The curves for both measured and smoothed data demonstrated that the concentrations varied substantially in time. The changes on the smoothed curves seemed to be fluctuations on a daily scale and for some pollutants, they appeared to exhibit some tendencies on a monthly scale, while the data series possessed diurnal periodicity as well. The trends, i.e. the smoothed curves over the seven months are in line with the distributions of the monthly median concentrations of the species at identical locations determined for several years (Salma et al., 2020b).



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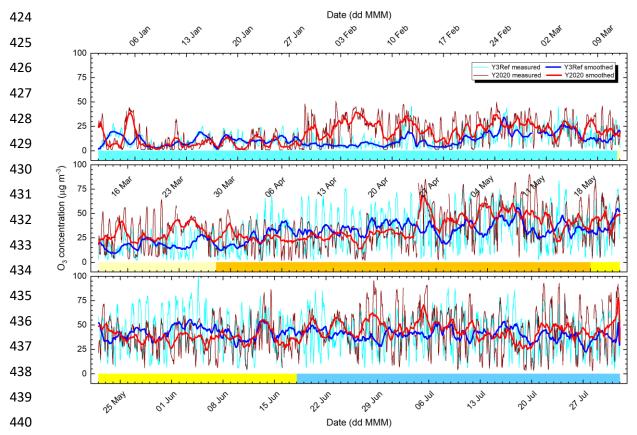


Figure 4. Time series of O₃ concentration in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their 24-h smoothed curves over the five consecutive phases of the first COVID-19 outbreak. The phases are marked by the following colour codes: Pre-emergency phase lighter blue, Pre-restriction phase lighter yellow, Restriction phase orange, Post-restriction phase darker yellow and Post-emergency phase darker blue. The tick labels of the abscissa indicate the Mondays in 2020.

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447The annual relative SDs (RSDs) for NO, NO2, CO, O3, SO2, PM10 mass, PM2.5 mass, N_{6-1000} ,448 N_{6-100} , N_{25-100} and $N_{100-1000}$ in years 2017–2019 were 115, 56, 43, 91, 37, 56, 74, 63, 69, 68 and44968 %, respectively (cf. Sect. 1). Their time distributions were complex. For species, which do

450 not normally show seasonal tendency such as particle number concentrations and perhaps PM_{10}

mass, the distributions of monthly RSDs were also featureless. For SO₂, which tends to exhibit 451 smaller concentration levels in summer than in winter, the distribution of its monthly RSDs 452 seemed to have an opposite behaviour. For O₃, which exhibits larger concentrations in summer 453 than in winter, the distribution of monthly RSDs showed again an opposite behaviour. These 454 relationships are in accordance with general metrological expectations. Excitingly, for NO, 455 NO₂, CO and perhaps PM_{2.5} mass, the distributions of monthly RSDs appeared to roughly 456 follow in parallel the concentration trends within the concentration ranges actually obtained. 457 The largest decrease in the RSDs from winter to summer was observed for NO, which was 458 459 approximately 20 % (of its annual mean RSD). The latter association could likely be linked to 460 meteorological conditions and source/sink intensities of these pollutants.

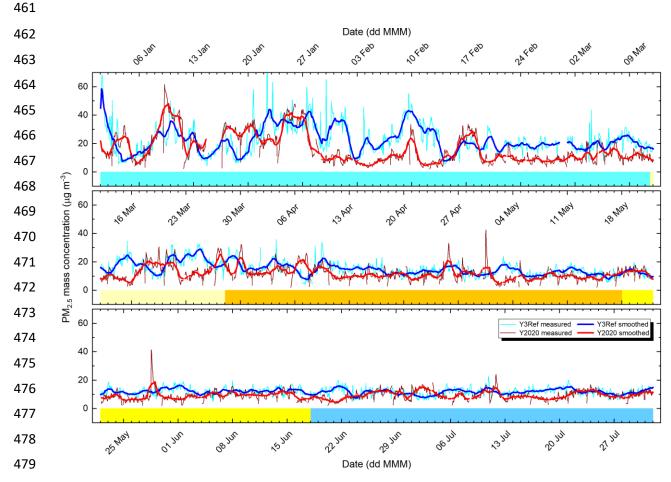


Figure 5. Time series of PM_{2.5} mass concentration in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their 24-h smoothed curves over the five consecutive phases of the first COVID-19 outbreak. The phases are marked by the following colour codes: Pre-emergency phase lighter blue, Pre-restriction phase lighter yellow, Restriction phase orange, Post-restriction phase darker yellow and Post-emergency phase darker blue. The tick labels of the abscissa indicate the Mondays in 2020.

Many chemical species investigated originate from rather different sources. Nevertheless, their 487 atmospheric concentrations often changed coherently, particularly in winter and early spring. 488 A nice example is the interval of approximately 14–28 March 2020 when most species varied 489 consistently. The MCPs for these days indicate strong anticyclonic weather types over the 490 Carpathian Basin, stagnant and relatively calm meteorological conditions without precipitation 491 in the area (Fig. S1). It is a confirmation of the common effects of regional meteorology on 492 atmospheric concentrations, and that the daily evolution of meteorology can have higher 493 influence on atmospheric concentrations than the source intensities under such specific 494 495 conditions (Salma et al., 2020a). Its consequences on the air quality are discussed in Sect. 3.8.

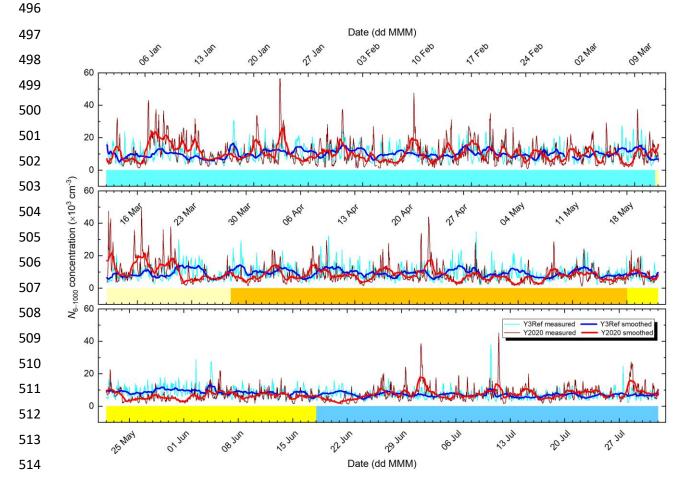


Figure 6. Time series of N_{6-1000} concentration in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their 24-h smoothed curves over the five consecutive phases of the first COVID-19 outbreak. The phases are marked by the following colour codes: Pre-emergency phase lighter blue, Pre-restriction phase lighter yellow, Restriction phase orange, Post-restriction phase darker yellow and Post-emergency phase darker blue. The tick labels of the abscissa indicate the Mondays in 2020.

The curves for PM_{2.5} mass and N_{6-1000} approved that there is week association between these two types of aerosol metrics (de Jesus et al., 2019). They are connected mainly via meteorological properties, which is anyway active for all pollutants. It was sensible, therefore, that both types of aerosol concentrations were included into the study as separate variables.

526 **3.4 Diurnal variations**

Average diurnal variations of NO, O₃, SO₂, PM_{2.5} mass and N_{6-100} together with the vehicle circulation separately for workdays and holidays over the Restriction pandemic phase, for which the differences in the shapes are expected to be the largest, are shown in Fig. 7 as examples.

531

The curves of NO and N_{6-100} (together with NO₂, CO and N_{6-1000} , which are not shown) 532 followed the typical pattern of road traffic. They can largely be associated with vehicular 533 sources (tailpipe emissions, primary and secondary particles), and can advantageously be 534 applied for assigning potential concentration changes to traffic reduction. It is seen that their 535 morning peak coincided with the peak of the morning rush hour, while their evening peak 536 appeared later than the peak of the afternoon rush hour. This shift could likely be related to the 537 daily evolution and cycling of the PBLH and to mixing intensity. The curves of N₆₋₁₀₀ contained 538 539 in addition the characteristic midday peak, which is caused by atmospheric NPF events. It is worth realising that its position was shifted to later time. There were only nine quantifiable 540 541 NPF events during the Restriction phase in year 2020, which might not result in a representative 542 shape. An alternative explanation could be that this peak was caused by the overlapping effects of direct traffic emissions and NPF events superimposed on each other. This experimental 543 544 observation should definitely be investigated and clarified when the necessary data sets become 545 available.

546

The curves for O₃ seemed to be opposite to NO as far as both their daily variations and the orders of concentration magnitudes on workdays and holidays are concerned. These are in line with the understanding of their atmospheric processes and coupled reaction mechanisms (Jacob, 1999). In addition, the shapes in Y2020 seemed to be flattened with respect to Y3Ref. The O₃ curves were also affected by the clock change, since the concentration of O₃ are substantially influenced by solar radiation.

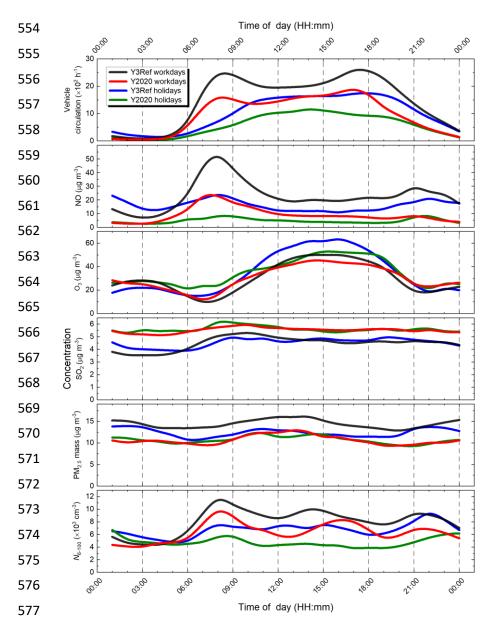


Figure 7. Average diurnal variations of motor vehicle road traffic in both directions on a major route (Váci Road) in Budapest and of NO, O₃, SO₂, PM_{2.5} mass and N_{6-100} concentrations separately for workdays and holidays in the average reference year of 2017–2019 (Y3Ref) and year 2020 during the Restriction phase of the first COVID-19 outbreak.

The curves for SO₂ (together with PM₁₀ mass and $N_{100-1000}$, which are not shown) tracked the traffic pattern very loosely if at all. They could partially be related to traffic through diesel fuel, resuspension of urban dust by moving vehicles, dispersion of road surfaces, (non-exhaust) emissions from material wear of moving parts of vehicles and growth/ageing of exhausted particles from vehicles (Salma and Maenhaut, 2006). Additional changes in the shape of the time variation of SO₂ could be caused by altered heating of and cooking at homes due to spreading practice of the work-from-home.

591 There was no obvious connection between the traffic and $PM_{2.5}$ mass, which confirms our 592 earlier conclusion that the fine particles in Budapest mainly originate from non-vehicular 593 sources (Salma et al., 2020a).

594 **3.5 Quantification of concentration changes**

595 There are several mathematical statistical tests to determine whether atmospheric 596 concentrations over some time intervals in different years belong to the same distribution or 597 not. These methods, however, quantify the joint influence of all environmental effects (Sect. 598 1) and do not provide information on their causal relationships. The method described and 599 applied below allows to unfold some potential confounding influence of environmental 600 variables (e.g. PBLH) from concentration changes in order to gain a closer insight into the 601 source intensities of motor vehicles.

602

603 Median concentrations of pollutant gases and aerosol particles, median traffic circulation data 604 together with their relative differences and standardised anomaly values for the five pandemic 605 phases in the average reference year and year 2020 are summarised in Tables 2–6. It should be 606 noted that the standardised anomalies are rather small when recalling, for instance, the rigorous 607 concept of the limits of detection ($3 \times$ SD) and determination ($10 \times$ SD) in analytical chemistry. 608 This is largely caused by the strong dynamic features of related atmospheric properties and 609 processes (Sects. 1. and 3.3).

610

We showed in Sect. 3.1 that it is the PBLH_{max} of the meteorological conditions that likely 611 612 caused the largest side effects on the concentrations, and, therefore, its influence was taken into account. A change in median concentrations for a pandemic phase was quantified to be 613 614 significant if both its relative difference fell outside the band of $[\pm 10 - f_{mix} \times RDiff(PBLH_{max})]$ % and its SAly was outside the range of ± 0.3 . The multiplication factor f_{mix} accounts for non-615 homogeneous mixing of pollutants within the boundary layer and for the effects of the daily 616 PBLH evolution. It was roughly estimated to be approximately 0.5. Its negative sign expresses 617 618 that atmospheric concentrations vary in a reciprocal manner with PBLH. The selected criteria were based upon exercises with the data in the individual years 2017, 2018 and 2019. The 619 620 procedure represents a sensible and consequent approach, though alternative limits could also be set. 621

Table 2. Median atmospheric concentrations of NO, NO₂ (both in units of μ g m⁻³) CO (mg m⁻³), O₃, SO₂, PM₁₀ mass, PM_{2.5} mass (all in μ g m⁻³), N_{6-1000} , N_{25-100} , $N_{100-1000}$ (all in 10³ cm⁻³) and median vehicle road traffic (h⁻¹) on Szabadság Bridge, Váci Road, Széna Square and Alkotás Road in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their relative difference (RDiff, %) and their anomaly standardised to SD (SAly) for Pre-emergency phase of the first COVID-19 outbreak. Chemical species with significant change are shown in bold.

628

Variable	Y3Ref	Y2020	RDiff	SAly
NO	31	18	-43	-0.5
NO_2	51	40	-22	-0.7
CO	0.74	0.58	-21	-0.8
O ₃	9.4	16	+68	+0.3
SO_2	5.5	5.4	-1	-0.0
PM_{10}	45	29	-36	-1.2
PM _{2.5}	21	12	-42	-1.2
N_{6-1000}	9.5	8.8	-7	-0.2
N_{6-100}	7.2	6.8	-6	-0.1
N_{25-100}	3.5	3.1	-10	-0.2
$N_{100-1000}$	2.2	1.7	-21	-0.5
Szabadság B.	676	640	-5	-0.1
Váci R.	1589	1299	-18	-0.4
Széna S.	1374	1437	+5	+0.1
Alkotás R.	2517	2425	-4	-0.1

629

630 The Pre-emergency phase (Table 2) fitted completely into the heating season. The traffic flows in city centre were identical, except for Váci Road, where it was somewhat lower in Y2020 631 632 than in Y3Ref. This could be cause by some local traffic arrangements. The PBLH_{max} increased by 32 % (Table 1), which is substantial and affected the concentrations. Most concentration 633 634 changes were not significant. The exceptions were NO, O₃, PM₁₀ mass and PM_{2.5} mass, and the latter two exhibited the largest anomalies. These two species have multiple sources. Organic 635 636 matter and elemental carbon, for instance, make up approximately 35 % of the PM_{2.5} mass in winter (Salma et al., 2020a), and biomass burning is the major source of carbonaceous aerosol 637 in this season with an approximate relative contribution to the total carbon of 67 %. The share 638 of fossil-fuel combustion is around 25 %. This all implies that PM_{2.5} mass concentrations can 639 fluctuate extensively and irregularly in the heating season due to the source intensities. The 640 reductions could also be related with changes in further meteorological properties such as T641

(mild February 2020, Fig. S4) or larger WS that acted on a shorter time scale than the pandemicphase (Fig. S3).

644

The higher O₃ concentration could partly be associated with the lower concentrations of NO. 645 Ozone exhibits a strong seasonal dependency (Salma et al., 2020b). Lower concentrations in 646 winter and early spring can be easily disturbed by its non-linear chemistry and by high WS. 647 The modest SAly for O₃ suggests that this considerable relative concentration increase was 648 mostly a consequence of low levels of O₃ in winter. The case nicely demonstrates the strength 649 650 of and requirement for the coupled utilisation of RDiff and SAly criteria. Furthermore, the main differences in the concentrations appeared sporadically in an isolated manner. In addition, there 651 was no coherence among the traffic-related variables. Therefore, all significant variations were 652 interested as results of inter-annual variability in local meteorology, emissions and formation 653 654 processes.

655

Table 3. Median atmospheric concentrations of NO, NO₂ (both in units of μ g m⁻³) CO (mg m⁻³), O₃, SO₂, PM₁₀ mass, PM_{2.5} mass (all in μ g m⁻³), N₆₋₁₀₀₀, N₆₋₁₀₀, N₂₅₋₁₀₀, N₁₀₀₋₁₀₀₀ (all in 10³ cm⁻³) and median vehicle road traffic (h⁻¹) on Szabadság Bridge, Váci Road, Széna Square and Alkotás Road in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their relative difference (RDiff) in % and their anomaly standardised to SD (SAly) for Pre-restriction phase of the first COVID-19 outbreak. Chemical species with significant change are shown in bold.

Variable	Y3Ref	Y2020	RDiff	SAly
NO	20	12	-39	-0.3
NO ₂	46	38	-18	-0.5
CO	0.60	0.56	-8	-0.2
O ₃	18	33	+80	+0.7
SO_2	5.1	5.4	+7	+0.3
PM_{10}	34	30	-12	-0.3
PM _{2.5}	19	13	-32	-0.8
N_{6-1000}	8.1	8.4	+4	+0.1
N_{6-100}	6.7	6.9	+4	+0.1
N_{25-100}	2.9	3.2	+9	+0.1
$N_{100-1000}$	1.4	1.6	+11	+0.2
Szabadság B.	652	417	-36	-0.8
Váci R.	1522	939	-38	-0.7
Széna S.	1371	1001	-27	-0.5
Alkotás R.	2792	1925	-31	-0.7

The Pre-restriction phase (Table 3) was rather short (16 days), and, therefore, its interpretation should be approached with a special caution due to some issues in representativity. It was also completely part of the heating season, and the extreme drought in the Carpathian Basin in 2020 could also play a role. The PBLH_{max} was almost identical in both years (Table 1). The concentrations of NO₂, PM_{2.5} mass and perhaps NO declined, while O₃ was enhanced. Excitingly, CO did not show substantial decrease. The changes could be affected by lower traffic during its last half/week (Fig. 2) and increased GRad.

670

Table 4. Median atmospheric concentrations of NO, NO₂ (both in units of μ g m⁻³) CO (mg m⁻³), O₃, SO₂, PM₁₀ mass, PM_{2.5} mass (all in μ g m⁻³), N₆₋₁₀₀₀, N₆₋₁₀₀, N₂₅₋₁₀₀, N₁₀₀₋₁₀₀₀ (all in 10³ cm⁻³) and median vehicle road traffic (h⁻¹) on Szabadság Bridge, Váci Road, Széna Square and Alkotás Road in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their relative difference (RDiff) in % and their anomaly standardised to SD (SAly) for Restriction phase of the first COVID-19 outbreak. Chemical species with significant change are shown in bold.

Variable	Y3Ref	Y2020	RDiff	SAly
NO	19	6.0	-68	-0.5
NO_2	44	26	-39	-1.1
СО	0.58	0.43	-27	-0.8
O ₃	31	35	+13	+0.2
SO_2	5.4	5.5	+3	+0.1
PM_{10}	32	28	-13	-0.3
PM _{2.5}	14	11	-22	-0.4
N_{6-1000}	8.8	6.7	-24	-0.5
N_{6-100}	7.4	5.3	-28	-0.6
N_{25-100}	3.2	2.8	-12	-0.2
$N_{100-1000}$	1.3	1.2	-5	+0.1
Szabadság B.	689	318	-54	-1.2
Váci R.	1626	803	-51	-1.0
Széna S.	1537	844	-45	-1.0
Alkotás R.	3031	1516	-50	-1.1

The beginning one-third part of the Restriction phase (Table 4) fell into the heating season, and it was fully incorporated into the extremely dry weather season. The vehicle flows were reduced by approximately half uniformly at all urban locations. Concentrations of NO, NO₂, CO, PM_{2.5} mass, N_{6-1000} and N_{6-100} changed significantly, and they all declined. The alterations happened in a systematic or continuous manner in time (Figs. 2–6, S6 and S7). These species

can be associated with vehicular road traffic. Except for PM2.5 mass, which is linked more to 684 household and residential sources. At the same time, some other important pollutants such as 685 $N_{100-1000}$ or SO₂ – which are typically related to larger spatial extent or region and which could, 686 therefore, be influenced by meteorology – did not change significantly. Similar reductions were 687 reported for other urban locations in the world (Keller et al., 2020; Lal et al., 2020; Le et al., 688 2020; Lee et al., 2020; Tobías et al., 2020). This all can be interpreted that the alterations in 689 NO, NO₂, CO, N_{6-1000} and N_{6-100} concentrations were primarily caused by the lower vehicular 690 traffic intensity in the city, and that the PBLH could also contribute by approximately 9 % in 691 692 an absolute sense (Table 1). The increased O_3 can be explained by its production primarily from volatile organic compounds (VOCs) and NO_x in the VOC-limited chemical regime even 693 under decreasing NO_x conditions (Jacob, 1999; Lelieveld and Dentener, 2000). This regime is 694 typical for many large cities, where the VOCs can involve, for instance, aromatics such as 695 benzene and toluene, which largely originate from traffic sources. 696

697

Table 5. Median atmospheric concentrations of NO, NO₂ (both in units of μ g m⁻³) CO (mg m⁻³), O₃, SO₂, PM₁₀ mass, PM_{2.5} mass (all in μ g m⁻³), N₆₋₁₀₀₀, N₆₋₁₀₀, N₂₅₋₁₀₀, N₁₀₀₋₁₀₀₀ (all in 10³ cm⁻³) and median vehicle road traffic (h⁻¹) on Szabadság Bridge, Váci Road, Széna Square and Alkotás Road in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their relative difference (RDiff) in % and their anomaly standardised to SD (SAly) for Post-restriction phase of the first COVID-19 outbreak. Chemical species with significant change are shown in bold.

Variable	Y3Ref	Y2020	RDiff	SAly
NO	12	6.4	-44	-0.2
NO ₂	40	26	-35	-0.9
CO	0.48	0.42	-13	-0.3
O ₃	42	37	+11	-0.2
SO_2	4.7	5.9	+26	+1.0
PM_{10}	29	21	-28	-0.6
PM _{2.5}	12	9.3	-24	-0.4
N_{6-1000}	8.2	6.0	-27	-0.5
N_{6-100}	6.8	4.9	-27	-0.5
N_{25-100}	3.3	2.4	-27	-0.5
$N_{100-1000}$	1.3	1.0	-22	-0.3
Szabadság B.	670	575	-14	-0.3
Váci R.	1536	1137	-26	-0.5
Széna S.	1540	1387	-10	-0.2
Alkotás R.	2597	2281	-12	-0.2

In the Post-restriction phase (Table 5), the vehicle flow recovered step wisely. The PBLH_{max} 706 in Y2020 decreased substantially relative to Y3Ref (Table 1). Most chemical species such as 707 NO₂, SO₂, PM₁₀ mass, PM_{2.5} mass, N₆₋₁₀₀₀, N₆₋₁₀₀, N₂₅₋₁₀₀ and N₁₀₀₋₁₀₀₀ exhibited significant 708 changes. The list also included variables which characterize the region. At the same time, some 709 710 typical vehicular-related species such as NO and CO - which are not really water soluble were not among them. Most significant changes showed decreasing tendency, except for SO₂ 711 which increased. The latter was caused by a continuously increasing SO₂ concentration (Fig. 712 713 S8), recorded at the other air quality monitoring stations as well. The increase was likely caused as a perturbance by some local sources in the upwind direction from the city. This all suggests 714 that the alterations were mainly produced by arrival of continued and spatially extended rains 715 in its second half of the pandemic phase (Fig. S1). The precipitation washed out many chemical 716 species from the urban and regional atmospheres. This time interval unambiguously 717 demonstrated that the regional weather can cause similar modifications in atmospheric 718 719 concentrations as a substantially reduced (by 50 %) urban traffic.

720

In the Post-emergency phase (Table 6), the traffic was at its ordinary level and there were no 721 722 larger weather alternations. Most concentrations - including some major vehicle-related pollutants such as CO and N_{6-100} – did not change significantly. The exceptions were NO, NO₂, 723 724 O₃ and PM_{2.5} mass. The first three variables are connected to each other through atmospheric chemistry. The changes can likely be linked to inter-annual variability in sources, sinks, 725 726 meteorological properties that act on a shorter time scale than the pandemic phase and 727 atmospheric transformation and transport – similarly to that observed in the Pre-emergency 728 phase.

Table 6. Median atmospheric concentrations of NO, NO₂ (both in units of μ g m⁻³) CO (mg m⁻³), O₃, SO₂, PM₁₀ mass, PM_{2.5} mass (all in μ g m⁻³), N₆₋₁₀₀₀, N₆₋₁₀₀, N₂₅₋₁₀₀, N₁₀₀₋₁₀₀₀ (all in 10³ cm⁻³) and median vehicle road traffic (h⁻¹) on Szabadság Bridge, Váci Road, Széna Square and Alkotás Road in the average reference year of 2017–2019 (Y3Ref) and year 2020 together with their relative difference (RDiff) in % and their anomaly standardised to SD (SAly) for Post-emergency phase of the first COVID-19 outbreak. Chemical species with significant change are shown in bold.

Variable	Y3Ref	Y2020	RDiff	SAly
NO	16	7.4	-54	-0.3
NO ₂	39	27	-31	-0.7
CO	0.43	0.42	-2	-0.1
O ₃	39	46	+17	+0.3
SO_2	4.0	4.3	+9	+0.3
PM_{10}	26	22	-15	-0.3
PM _{2.5}	12	9.1	-22	-0.3
N_{6-1000}	6.7	6.7	+0	+0.0
N_{6-100}	5.5	5.4	-2	-0.0
N_{25-100}	2.7	2.8	+5	+0.1
$N_{100-1000}$	1.1	1.2	+9	+0.1
Szabadság B.	690	663	_4	-0.1
Váci R.	1471	1218	-17	-0.3
Széna S.	1594	1511	-5	-0.1
Alkotás R.	2507	2531	+1	+0.0

737 **3.6 Change rates**

Linear regression analysis between the median RDiff for vehicle traffic on one side and RDiff for pollutants corrected for the RDiff(PBLH_{max}) on the other side for all pandemic phases yielded change rates and SDs for NO, NO₂, N_{6-1000} and CO were 0.63 ± 0.23 , 0.57 ± 0.14 , 0.40 ± 0.17 and 0.22 ± 0.08 , respectively. For PM₁₀ mass and PM_{2.5} mass, the rates were slightly negative and insignificant. The data points for the Post-restriction phase – which were substantially affected by precipitation and frontal weather systems – were excluded from this analysis.

745

The change rates suggest that nitrogen-oxides vary sensitively with traffic, total particle number concentration shows considerable dependency, while variation of CO is modest. This is linked to their residence times as well. The PM mass concentrations do not appear to be closely related to traffic intensity in central Budapest.

750 **3.7 Spatial gradients**

Spatial distributions of NO and O₃ derived by CAMS ensemble reanalysis in 2018–2019 and 751 2020 during the Restriction pandemic phase are shown in Figs. 8 and 9 as examples. The 752 absolute concentrations can be different from the measured values due to the specialities in the 753 applied models, while the relative tendencies are expected to be expressed correctly. Figure 8 754 indicates that the differences from the corresponding median (spatial gradients) in 2020 were 755 larger than in 2018–2019. This can be explained if the relative concentration changes at the 756 757 outer parts of the city or near-city background were even larger than in the centre. The spatial distribution of NO₂ was similar to NO, although its gradients were smaller than for NO. Spatial 758 759 distributions of CO and PM_{2.5} mass were featureless and similar to each other in 2018–2019 and 2020. 760

761

Spatial distributions of O₃ (Fig. 9) and, perhaps SO₂ (which is not shown), exhibited relative decrease in the centre, which gradients were relatively small and similar to each other for both time intervals. This all is in line with the tendencies observed in their measured concentrations (Sects. 3.3 and 3.4). We are aware that several pollutants originate from diffusive line sources, which can be enriched along roads and, therefore, much larger concentration gradients can occur on smaller spatial scales.

768 **3.8 Potentials for improving air quality**

In order to assess the importance of concentration changes that could be achieved by tranquilizing the vehicle road traffic in Budapest, the atmospheric concentrations and their possible decrements were compared to various limit values (EU Directives, 2008; VM 4, 2011).

NO₂ exhibited the most frequent exceedances of standards for the protection of health. Its 773 concentrations in 2017, 2018 and 2019 were larger in 172, 155 and 61 cases than the 1-h 774 national health limit of 100 μ g m⁻³ (Fig. S6). The permitted number of exceedances is 18 a 775 year. It is mentioned that this concentration limit is 200 μ g m⁻³ for the EU, which would be 776 777 fulfilled completely. The NO₂ excess usually remained modest – particularly when contrasted to the smog alert thresholds of 350 (for the warning state) and 400 μ g m⁻³ (for the alarm state). 778 The daily health limit of 85 μ g m⁻³ was also exceeded in 6, 2 and 0 days in the three years, 779 respectively. A reduction of 10 % in vehicle circulation (that corresponds to a 6 %-decline in 780

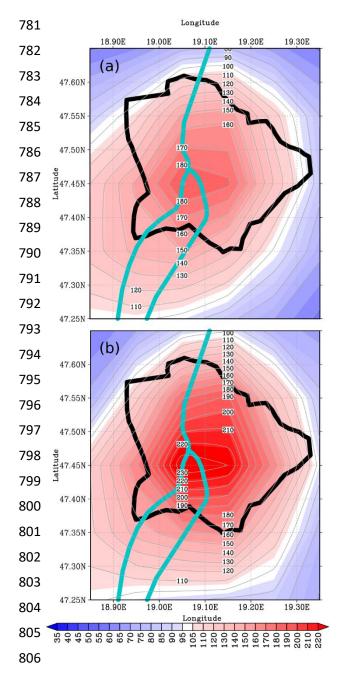


Figure 8. Spatial distribution of median NO 807 808 concentration in Budapest in 2018-2019 (a) 809 and 2020 (b) during the Restriction phase of 810 the first COVID-19 outbreak obtained from 811 CAMS ensemble reanalysis. The concentrations were normalised to the overall 812 spatial median concentrations of 0.93 and 813 $0.59 \ \mu g \ m^{-3}$, respectively. The border of the 814 city and the Danube River are indicated with 815 816 curves in black and blue colours, respectively 817 for better orientation.

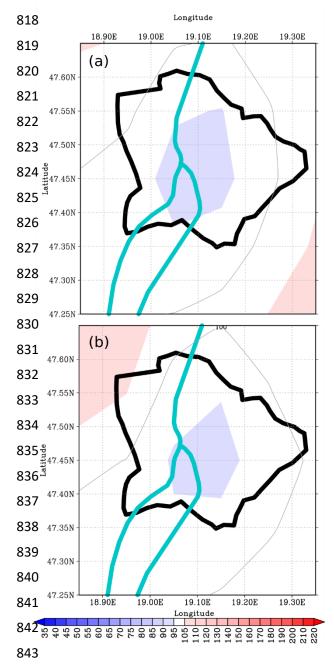


Figure 9. Spatial distribution of median O₃ 844 845 concentration in Budapest in 2018–2019 (a) 846 and 2020 (b) during the Restriction phase of 847 the first COVID-19 outbreak obtained from 848 CAMS ensemble reanalysis. The 849 concentrations were normalised to the overall 850 spatial median concentrations of 60 and 67 µg 851 m⁻³, respectively. The border of the city and 852 the Danube River are indicated with curves in 853 black and blue colours, respectively for better 854 orientation.

in NO₂ concentration) would decrease the number of exceedances of the 1-h national health
limit to 114, 98 and 42, respectively (thus, typically by ca. 30 %), while the number of days
above the daily health limit would be lowered to 2, 0 and 0, respectively.

858

Less frequent though more severe exceedances than for NO₂ happened for PM₁₀ mass (Fig. 859 S9). The daily mean PM₁₀ mass concentrations in 2017, 2018 and 2019 exceeded the daily 860 health limit of 50 μ g m⁻³ in 36, 93 and 57 days, respectively at this actual air quality monitoring 861 station. Most exceedances occurred in the heating season. Their permitted number is 35 a year. 862 The smog alert thresholds for the warning and alarm states are 75 and 100 μ g m⁻³, respectively. 863 The number of exceedances for the warning stage were 12, 17 and 9, respectively. It should be 864 added that smog alerts are announced on the basis of a complex set of conditions which include 865 larger numbers of monitoring stations and days. As a matter of fact, the warning state was 866 announced 3 times for 11 days in total and once for 2 days in 2017 and 2018, respectively. 867 868 There was no smog alert in 2019. It is stressed that all alarm states since 2007 were announced exclusively because of high PM₁₀ mass concentrations, and all alert intervals were confined to 869 870 winter. This points to the role of local and regional meteorology and other sources than vehicle traffic (Salma et al., 2020a). There is cold air pool that develops from time to time above the 871 Carpathian Basin in winter, which generates a lasting T inversion and a shallow planetary 872 boundary layer, restricts the vertical mixing and results in poor air quality over extended areas 873 874 of the basin in larger and smaller cities as well as in rural areas.

875

All O₃ concentrations were below the maximum daily 8-h health limit of 120 μ g m⁻³ (Fig. 4). The concentrations of CO were far away from both the 1-h and maximum daily 8-h health limits of 10 and 5 mg m⁻³, respectively (Fig. S7), and the situation was similar for SO₂, for which the 1-h and daily limits are 250 and 125 μ g m⁻³, respectively (Fig. S8).

880 4 Conclusions

The relationships between urban air quality and motor vehicle road traffic are not straightforward since the contributions of traffic flow to pollutants concentrations are superimposed in the variability in local meteorological conditions, long-range transport of air masses and other sources/sinks. We introduced here an approach based on both relative difference and standardised anomaly, which helps unfolding some important confounding environmental factors. It can support creating a generalised picture on urban atmospheres.

The method was deployed on the Budapest data during the different phases of the first COVID-888 19 outbreak. Various restriction measures introduced due to the pandemic resulted in a decline 889 of vehicle road traffic down to approximately 50 % during the severest limitations. In parallel, 890 concentrations of NO, NO₂, CO, N_{6-1000} and N_{6-100} decreased substantially, some other species 891 892 such as PM_{2.5} mass, PM₁₀ mass and N₁₀₀₋₁₀₀₀ changed modestly and inconclusively, while O₃ showed an increasing tendency. Change rates of NO and NO₂ with relative change of traffic 893 intensity were the largest (approximately 0.6), total particle number concentration showed 894 895 considerable dependency (0.4), while variation of CO was modest (0.2). It was demonstrated that a similar decrease in concentrations as observed in the strictest pandemic phase can also 896 be caused by other (natural/meteorological) effects than traffic. The rainy weather in June 2020 897 (the so-called St. Medard's forty days of rain in Central European folklore) yielded, for 898 899 instance, very similar low pollution levels.

900

The study revealed that intentional reduction of traffic intensity can have unambiguous potentials in improving urban air quality as far as NO, NO₂, CO and particle number concentrations are concerned. It should be added that the most critical pollutant in many European cities including Budapest, namely the PM₁₀ mass, however, did not seem to be considerably affected by vehicle flow. Nevertheless, measures for tranquillizing urban traffic can contribute to improved air quality through a new strategy for lowering the population exposure of inhabitants instead of high-risk management of individuals.

908

909 The method could be expanded by other important ordinary chemical species such as soot and 910 by other location types such as near-city or regional background sites jointly with central 911 locations in order to obtain more exact meteorology-normalized changes. The results also point 912 to the importance of non-linear relationships among precursors and secondary pollutants, 913 which are to be further studied to gain better insights into urban atmospheric chemistry and air 914 quality issues.

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919 *Supplement.* The supplement related to this article is available online.

⁹¹⁶ *Data availability.* The observational data are accessible at http://www.levegominoseg.hu/ or are
917 available from the corresponding author – except for the vehicle road traffic – upon request.

- 921 Author contributions. IS conceived the study. AZGy, WT and IS performed most aerosol and 922 meteorological measurements. All co-authors participated in the data processing, calculations and 923 interpreting the results. The figures were created by MV and AZGy. IS wrote the manuscript with 924 comments from all coauthors.
- 925
- 926 *Competing interests.* The authors declare that they have no conflict of interest.

927
928 Acknowledgements. The authors thank the leaders of the Budapest Public Roads Ltd. (Budapest Közút
929 Zrt.) for providing the vehicle road traffic data and its coworker Dezső Huszár for valuable discussions.
930 The map in Fig. 1 was created by Márton Pál, Ph. D. student of the Department of Cartography and
931 Geoinformatics, Eötvös University. The authors are grateful to Attila Machon (Hungarian

- 932 Meteorological Service) for his help with the criteria air pollutant data.
- 933

Financial support. This research was supported by the Hungarian Research, Development and
 Innovation Office (grant nos. K116788 and K132254) and by the European Regional Development
 European data the Hungarian Comment (CINOP 2.2.2.15, 2016, 00055)

Fund and the Hungarian Government (GINOP-2.3.2-15-2016-00055).

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