



# Short-term effect of COVID-19 lockdowns on atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations in urban environment

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## Abstract

The COVID-19 pandemic has changed all areas of human activity as it forced the authorities around the world to enact unprecedented restrictions such as “lockdowns”. The low economic activity reduced the anthropogenic impact on the environment, in particular, greenhouse gases and aerosols emissions were decreased. However, the associated change in air quality is difficult to directly observe and quantify, since concentrations of these components in urban areas are affected by many other factors. In this work statistical analysis of atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub>, measured in 2017–2020 in the city of Ekaterinburg, Russia, are presented. A detailed focus was made on the lockdown period from March 28 to April 30, 2020. A significant decrease in concentrations and inter-hourly variations of all studied components were observed only in the short “self-isolation” period from April 6 to April 8. The anthropogenic origin of this effect, primarily associated with the reduction in vehicular traffic, was concluded from mean diurnal cycles and air temperature correlations of all components. A decrease in the difference between measured and background CO<sub>2</sub> and CH<sub>4</sub> mole fractions was also found during this period. The difference was  $1.3 \pm 0.2$  ppm for CO<sub>2</sub> and  $8 \pm 4$  ppb for CH<sub>4</sub>, which was many times lower than during any other observed periods, suggesting a short-term effect of lockdown restrictions. Overall, a negative impact on the atmosphere quickly resumed after the recovery of economic activity. The approaches in this study can be used to detect weak fluctuations of atmospheric components in other urban territories.

**Keywords** Aerosols · Background · Diurnal variations · Greenhouse gases · Pandemic

## Introduction

Greenhouse gases (GHG) are the main optically active components of the atmosphere which determine the global climate. The main GHGs are carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) (IPCC 2014). A growth in GHG concentration is known to lead to an increase in the absorption of infrared radiation and, accordingly, to an increase in air temperature. The CH<sub>4</sub> molecule absorbs infrared radiation ten times more efficiently than the CO<sub>2</sub> molecule. But the number of CO<sub>2</sub> molecules in the atmosphere is about 200 times greater than those of methane (Karol and Kiselev 2004). Hence,

the CO<sub>2</sub> contributes to the total radiative forcing more than 60%, and CH<sub>4</sub> accounts for about 20% (IPCC 2013). Over the past 10 years, the mean annual absolute increase in CO<sub>2</sub> and CH<sub>4</sub> mole fractions was 2.26 ppm and 7.0 ppb, respectively (WMO 2019). Therefore, the study of anthropogenic and natural influxes of these gaseous impurities into the atmosphere including via continuous measurements is a key research area concerning the climate change problem.

Aerosols also have a significant influence on the climate, but the extent of this influence is still discussible. At global level, aerosols seem to reduce the greenhouse gas-induced warming. However, many climate effects from aerosols are regional rather than global, and can vary greatly (Samset 2018). The total radiative forcing of aerosols was estimated to reach 10% (IPCC 2013). Aerosols significantly affect the precipitation processes and can influence on the human health. The main hazard of aerosols as suspended particles is their micron size. Due to the small size and weight, aerosols can be long-range transported in the atmosphere what can increase their negative impact on the environment and

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humans.  $PM_{2.5}$  particles pose the greatest danger to humans, as they can be transported deeply inside the lungs (Ginzburg et al. 2009; Song et al. 2018).

The amount of local anthropogenic releases of GHGs and aerosols at a given area is obviously attributed to the degree of its urbanization and industrial development. Therefore, in recent years, the atmospheric scientists and climatologists particularly focus on the study of the gaseous and aerosol compositions of the atmosphere in urban agglomerations considering them as significant contributors to global and regional climate change (Chandra et al. 2016; Gubanova et al. 2018; Liu et al. 2019, 2016; Super et al. 2017). In this regard, special relevance attaches to the events which can drastically alter the usual course of urban life. A pandemic of COVID-19 (scientifically referred to as the severe acute respiratory syndrome–coronavirus 2 or SARS-CoV-2) became such an event in 2020. The spread of COVID-19, identified in more than 160 countries around the world, caused unprecedented enforced and voluntary restrictions (such as "lock-downs") for the population and business. The reduced economic activity and people's mobility in turn should have led to a decrease in environmental impact. For example, some reduction in regional and global emissions of both GHGs and air pollutants was brought out (Lamprecht et al. 2021; Le Quéré et al. 2020). An associated decrease in air pollution and  $CO_2$  concentration has been observed in satellite data and by local ground-based observations (Buchwitz et al. 2021; Shi and Brasseur 2020). Nevertheless, it is difficult to detect and quantify the decrease in  $CO_2$  concentration in urban environment directly from sparse ground observations affected by many other factors, such as synoptic conditions, natural fluxes and background  $CO_2$  fluctuations (Kutsch et al. 2020; Liu et al. 2021).

In this study, continuous in situ measurements of atmospheric  $CO_2$ ,  $CH_4$  and  $PM_{2.5}$  from December 1, 2017 to May 31, 2020 in the city of Ekaterinburg, Russia (56.85 N; 60.65 E) were analyzed. To test the hypothesis about the impact of significant but relatively short-term decline in economic activity on the local concentration of GHGs and anthropogenic aerosols, the period of severe and moderate restrictions associated with COVID-19 was examined in more detail.

## Materials and methods

### Site overview

Since 2015 the continuous monitoring of atmospheric  $CO_2$ ,  $CH_4$  and  $PM_{2.5}$  has been carried out at the Institute of Industrial Ecology, Ural Branch of the Russian Academy of Sciences in Ekaterinburg (56.85 N; 60.65 E, 283 m a.s.l.). Ekaterinburg is the administrative center of the Sverdlovsk

region (Ural Federal District) and one of the largest cities in the Russian Federation with a developed transport and industrial infrastructure (RFCCI 2019). The climate is moderately continental bordering on continental with sharp variability of weather conditions and pronounced seasonality. The average annual temperature is  $+3\text{ }^\circ\text{C}$  with a tendency to increase rapidly (with the rate of about 1 degree in 25 years) (Vasiliev et al. 2020). The average temperature for the coldest month (January) is  $-12.6\text{ }^\circ\text{C}$ , and for the warmest (July) is  $+19.0\text{ }^\circ\text{C}$ . West winds prevail (ClimExp 2021). The measurement system is located 4 km northeast of the city center of Ekaterinburg. Low-rise buildings dominate the urban development of this location. There is a dendrological park with some ponds in the immediate vicinity.

### Instrumentations

The measurement system is hosted at the Siemens monitoring post, which includes a pavilion, a system for sampling trace gases and aerosols, a life support system for maintaining the indoor temperature and distributing power to devices, and a meteorological mast. The main devices are the Picarro G2401 laser gas analyzer, the Panasonic  $PM_{2.5}$  optical aerosol sensor, and the Meteo-2 automated ultrasonic meteorological complex. Meteo-2 measures the surface air temperature, wind speed and direction, atmospheric pressure, and relative humidity.

Picarro G2401 (Picarro Inc., USA) is a cavity ring-down spectrometer, which enables monitoring of  $CO_2$ ,  $CH_4$ ,  $CO$ , and water vapor in the surface atmosphere to be made in continuous unattended mode. Such a system has proven its suitability for high-precision and stable measurements (Crosson 2008). The sampling inlet with rain guard is kept at the height of 10 m above the ground. The sample air is drawn into the measurement cell through a 6 mm fluoroplastic tube and is cleaned of dust and aerosols by a cascade of 40, 7 and 2  $\mu\text{m}$  filters. The signal recording frequency is more than 1 Hz. The instrument is calibrated once a month using reference gas mixtures produced in Russia. Three standard gases cover the analyzer operating range for each component with concentrations of 296, 396 and 500 ppm for  $CO_2$  and 1.2, 3.1 and 4.2 ppm for  $CH_4$ . Recorded calibration data are additionally processed. For each standard gas this procedure includes isolating and averaging the calibration interval, during which the analyzer provides stable measurements. Then, the coefficient of linear regression between assigned reference and measured concentrations is found allowing corrections for the latter. The units of mole fraction are  $\mu\text{mol mol}^{-1}$  or  $\text{nmol mol}^{-1}$  (abbreviated ppm or ppb, respectively).

Panasonic  $PM_{2.5}$  aerosol sensor is designed to register particles with a diameter of 0.3 to 2.5  $\mu\text{m}$  in the concentration range from 0 to 300  $\mu\text{g m}^{-3}$  using the scattering of light emitted by a red LED. The location of the sensor is similar

as for the gas sampling inlet. To protect against precipitation, the device is placed in a protective case. The sensor has no active pumping, the flow of aerosol particles is provided due to a constant temperature gradient created by the additionally installed heater (Nakayama et al. 2018). Raw data are recorded every ten seconds to a PC connected via a USB cable. Calibrations with aerosol spectrometers DAS or GRIMM are regularly performed to ensure the validity of the sensor measurements. If necessary, the data are corrected using the obtained calibration coefficients.

All instrumental observations are binned at a temporal resolution of 1 h and synchronized with meteorological data measured simultaneously. For the hourly average CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations, the anomalous values associated with technical fault of the instruments are statistically filtered based on hypothesis of a lognormal distribution of initial data. Values outside the 1–99th percentile range are flagged as outliers and excluded from further analysis. Initial data are also used to assess the intra-hour variations ( $\sigma$ ) which are applied to determine background concentrations (see text below).

## Data analysis methods

An important tool for analysis of local and regional GHG processes based on instrumental measurements is the extraction of so-called background levels. The “background” is conventionally understood as the concentration of a given species in a pristine air mass in which anthropogenic impurities are not present. Direct results of CO<sub>2</sub> and CH<sub>4</sub> measurements in an area being not affected by local emissions may indicate background levels at a given time, but this is impossible in an urban environment. Therefore, background concentrations of these gases in Ekaterinburg were determined by the method described in details elsewhere (Aalto et al. 2007; Ivakhov et al. 2019). The method allows to estimate the “background” taking into account the influence of the horizontal wind speed on measurements.

Previously it was found that in Ekaterinburg the highest mole fractions and variations of atmospheric CO<sub>2</sub> and CH<sub>4</sub> correspond to wind speeds up to 2.5 m s<sup>-1</sup> (Gulyaev et al. 2020). When wind speeds are higher than this threshold, polluted urban air is apparently quickly replaced by cleaner air resulting in a decrease in CO<sub>2</sub> and CH<sub>4</sub> mole fractions and its variations. This indirectly confirms the influence of local anthropogenic sources on the observations at low wind speeds (Elansky et al. 2015). Time intervals corresponding to wind speed up to 2.5 m s<sup>-1</sup> were excluded from the data set for calculation of the background. Mole fractions with intra-hour variations exceeding 1 $\sigma$  (> 1 ppm for CO<sub>2</sub> and > 8 ppb for CH<sub>4</sub>) were also discarded. Further, for each calendar day a four-hour continuous interval was selected with a minimum averaged mole fraction, which was

stipulated to be the background level for this day. If such an interval was not found within a day the background was not calculated. It should be noted that the choice of the interval duration is rather subjective and depends on the characteristics of a monitoring station (Stephens et al. 2013).

To recognize the potential sources and intensity of GHG emission, the ratio of  $\Delta\text{CH}_4/\Delta\text{CO}_2$ , ppb ppm<sup>-1</sup>, between the excess of CH<sub>4</sub> relative to the excess of CO<sub>2</sub> was used as it was proposed previously (Chandra et al. 2016; Mahata et al. 2017; Worthy et al. 2009). For each species, the excess values were estimated from the initial time series by subtracting the matching background curve from the respective hourly values. The background curve was calculated as the fifth percentile of the hourly data for a 24 h moving window. Negative hourly residuals were discarded from the analysis. The background curve determined in this way is likely to underestimate the true “background” concentrations (see above) since the influence of meteorological factors is neglected. However, this method avoids gaps in fixing daily averages and characterizes only simultaneous variability in CH<sub>4</sub> and CO<sub>2</sub> relative to their own background. Certain statistical indicators for the  $\Delta\text{CH}_4/\Delta\text{CO}_2$ , aggregated over specified period, can be useful for identifying potential emission sources and sinks.

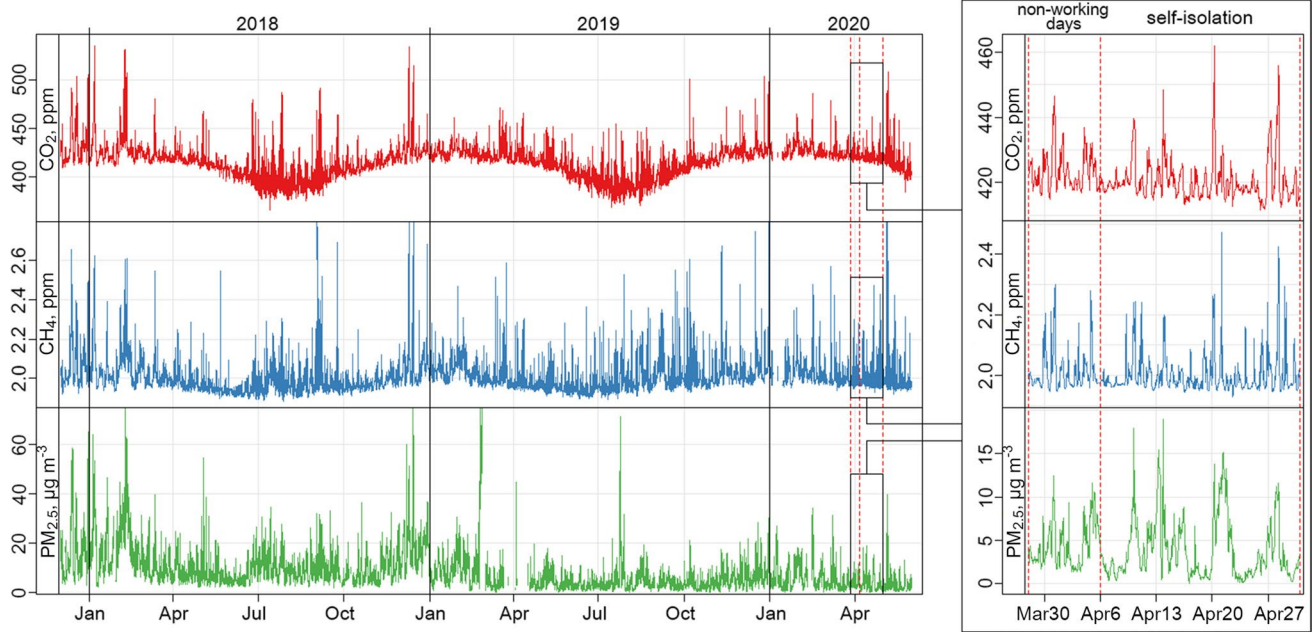
## Results and discussion

### Hourly and monthly average dynamics of atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub>

To assess the effect of the reduced economic activity and population mobility associated with COVID-19 restrictions of 2020 in Ekaterinburg on atmospheric GHGs and aerosols, hourly average concentrations for the period from December 1, 2017 to May 31, 2020 were analyzed (Fig. 1). The lockdown period of 2020 declared in the Russian Federation and, consequently, in the Sverdlovsk region was considered in more detail. This period included two stages, namely “non-working days” from March 28 to April 5, 2020 (including weekends) (Order 2020a) and “self-isolation” from April 6 to April 30, 2020 (Order 2020b, d). Overall, CO<sub>2</sub> time series reflects the characteristic seasonal dynamics with minimum mole fractions in July–August due to the active phenological phase of local ecosystem. A similar trend with minimum values in May–June and maximum values in January was observed for CH<sub>4</sub>. There was no such pronounced tendency for PM<sub>2.5</sub>.

Monthly aggregated descriptive statistics of the hourly average data are summarized in Table 1. During the specified lockdown period, no significant decrease in monthly mean values was observed compared to the same months in previous years, except for aerosol. Thus, the data averaging





**Fig. 1** Time series of hourly average atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations in Ekaterinburg from December 1, 2017 to May 31, 2020 (left panel) and in more detail during active phase of COVID-19 lockdown from March 28 to April 30, 2020 (right panel)

**Table 1** Summary of monthly aggregated CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations observed in Ekaterinburg from December 1, 2017 to May 31, 2020 (mean and standard deviation)

Month	Mean CO <sub>2</sub> (SD), ppm			Mean CH <sub>4</sub> (SD), ppb			Mean PM <sub>2.5</sub> (SD), µg m <sup>-3</sup>		
	2018	2019	2020	2018	2019	2020	2018	2019	2020
Dec <sup>a</sup>	431.7 (18.8)	430.0 (16.2)	428.0 (12.5)	2063 (113)	2081 (154)	2044 (87)	17.1 (11.5)	19.0 (9.6)	7.1 (5.1)
Jan	427.1 (16.7)	425.5 (6.3)	428.4 <sup>b</sup> (8.1)	2043 (115)	2046 (59)	2032 <sup>b</sup> (61)	16.6 (10.0)	8.8 (4.6)	6.1 (5.2)
Feb	434.5 (20.6)	425.0 (5.9)	428.2 (8.5)	2096 (118)	2037 (60)	2031 (66)	22.38 (11.9)	10.4 (9.7)	6.6 (5.4)
Mar	421.4 (7.9)	422.9 (8.7)	423.4 (5.9)	2003 (67)	2007 (69)	2002 (59)	10.0 (5.7)	4.7 <sup>b</sup> (3.4)	3.7 (3.3)
Apr	418.8 (5.9)	419.4 (8.4)	421.0 (6.4)	1981 (48)	1979 (54)	2003 (67)	7.9 (3.8)	3.5 <sup>b</sup> (3.7)	3.9 (3.4)
May	412.3 (8.2)	413.6 (9.7)	416.3 (17.6)	1953 (48)	1964 (47)	2001 (117)	7.1 (5.4)	4.4 (3.0)	3.0 (3.9)
Jun	403.3 (11.1)	399.6 (8.2)	–	1940 (39)	1946 (49)	–	6.9 (4.2)	2.8 (2.0)	–
Jul	398.2 (20.2)	392.9 (14.4)	–	1992 (75)	1974 (66)	–	11.1 (5.7)	5.3 (7.7)	–
Aug	393.9 (11.0)	395.4 (14.0)	–	1972 (58)	1998 (68)	–	7.9 (3.5)	4.4 (3.0)	–
Sep	409.2 (19.2)	405.2 (8.0)	–	2005 (118)	2023 (81)	–	9.9 (6.0)	3.4 (2.1)	–
Oct	409.8 (6.5)	416.9 (11.7)	–	1965 (34)	2001 (74)	–	7.2 (5.1)	3.6 (3.1)	–
Nov	418.6 (5.6)	423.5 (7.7)	–	1995 (36)	2029 (64)	–	10.0 (5.3)	5.4 (3.4)	–

<sup>a</sup>In each specified period, December corresponds to the previous year (i.e. December of 2017, 2018 and 2019)

<sup>b</sup>Monthly average estimated from incomplete data set due to instrument maintenance

over each month does not reveal the effect of GHG's concentration reduction. But it is noteworthy that CO<sub>2</sub> and CH<sub>4</sub> mole fractions increased significantly in May 2020 over the same period in previous years at a rate higher than 10 years mean global annual growth of 2.26 ppm and 7.0 ppb, respectively (WMO 2019). The variability of these values (in terms of standard deviation) jumped unexpectedly and became

comparable to those in the periods with high energy consumption (winter months). This effect was likely attributed to the gradual lifting of lockdown restrictions in accordance with executive order of Russia authorities (Order 2020c), as well as to the May holidays during which the residents of Ekaterinburg repeatedly violated the self-isolation regime (TASS 2020).





To specify the possible short-term effect of COVID-19 restrictions on atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations, the entire lockdown period from March 28 to April 30, 2020 was divided into three-day intervals (Table 2). The stage of “non-working days” (from March 28 to April 5), as expected, did not affect the content of the impurities in the urban atmosphere, since by that time no distinct lockdown measures had been implemented. The observations were greatly influenced at the beginning of “self-isolation” period (from April 6 to April 8) when economic and population activities were significantly reduced by mandatory confinements of all but key workers and organizations. The effect was manifested in a multiple decrease in CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> variability and in minimum values of CH<sub>4</sub> and PM<sub>2.5</sub> concentrations as well. The mean CO<sub>2</sub> mole fraction was not minimal during these days suggesting the seasonal peculiarities of natural ecosystem in the city. In early April, biological CO<sub>2</sub> sink is insufficient and is getting more intense later, in late April–early May. During the rest of “self-isolation” (from April 9 to April 30), the effect of the reduced variability and concentration was not observed, which probably indicates the influence of local anthropogenic sources of GHGs and aerosols. In the course of this period the residents of Ekaterinburg have adapted to regulatory measures and developed mechanisms to bypass severe restrictions. Apparently, this was typical for all Russian cities (Thu et al. 2020). At the same time, the dynamics of industrial production did not statistically drop in April–May 2020, which means the normal operation of the main enterprises of the region (Hartwell et al. 2021; Zubarevich and Safronov 2020). Therefore, the obtained results are mostly accounted for by the changes in population activity, rather than in industry.

### Diurnal variations of CO<sub>2</sub>, CH<sub>4</sub>, PM<sub>2.5</sub> and CO

For better demonstration of the influence of local anthropogenic sources on the observed GHG and aerosol content, mean diurnal cycles for each component were estimated (Fig. 2). The values for carbon monoxide (CO), which is a good air pollution marker as a product of incomplete combustion of fossil fuel and biomass, were also added. Taking

into account the above results, the mean diurnal cycles were plotted separately for three stages of 2020 lockdown, viz.

- (1) “non-working days” (from March 28 to April 5);
- (2) the beginning of “self-isolation” period (from April 6 to April 8);
- (3) the rest of “self-isolation” period (from April 9 to April 30).

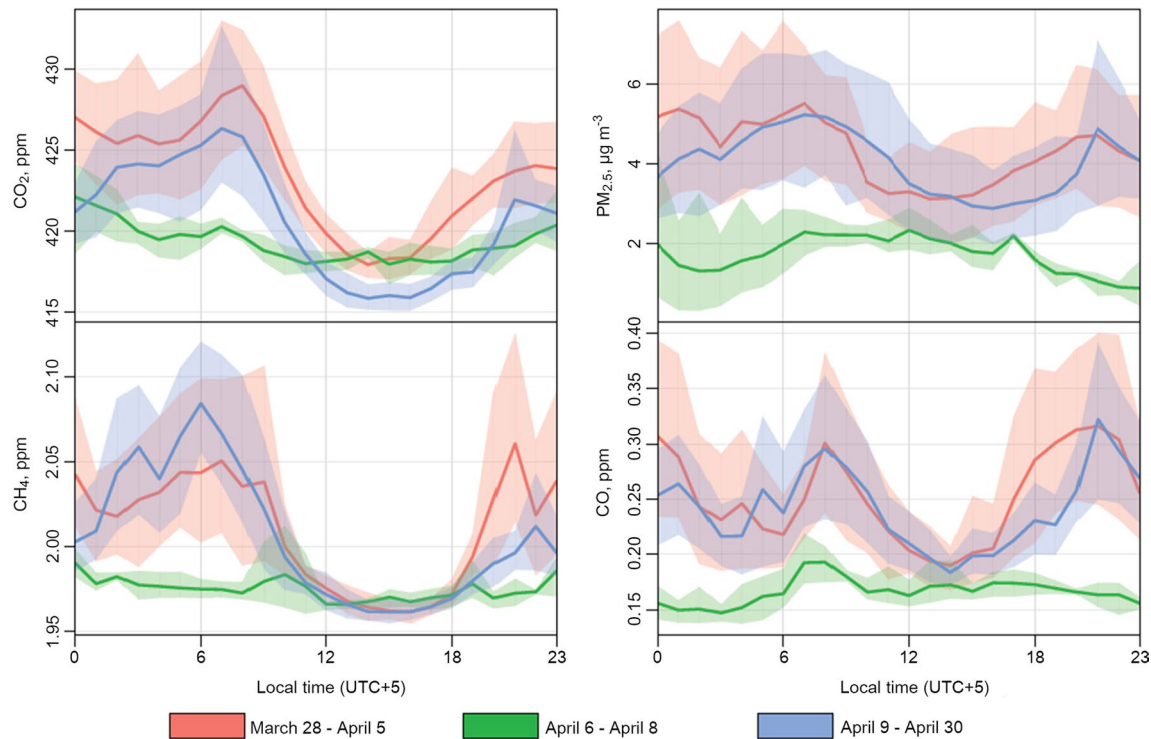
Diurnal variations of CO<sub>2</sub>, CH<sub>4</sub>, and PM<sub>2.5</sub> at stages (1) and (3) were in good agreement with typical characteristics of this season in the presence of local sources. Higher concentrations at night and in the morning are usually due to the local emissions and a stable surface atmosphere especially when thermal inversion occurs. In the daytime, concentrations decrease because the emissions are diluted through vertical mixing of surface air with cleaner air from upper tropospheric layers (Dhaka et al. 2020; Vinogradova et al. 2007). In addition, the emissions are partly offset by CO<sub>2</sub> photosynthetic assimilation and CH<sub>4</sub> hydroxyl oxidation. At stage (2), the mean diurnal cycles of all measured components tended to be flat with average concentrations being lower than corresponding values at stages (1) and (3). Such an effect is known to occur in the absence of local sources of GHG (Ishizawa et al. 2019). If there are strong local emissions, the surface concentrations are better modulated by the daily planetary boundary layer resulting in enhanced diurnal variability. Thus, at stage (2), the influence of the reduced economic and population activities on atmospheric CO<sub>2</sub>, CH<sub>4</sub>, and PM<sub>2.5</sub> was significant.

In the diurnal dynamics of CO, the maximum values were observed at 8:00 a.m. and after 6:00 p.m. at stages (1) and (3). These moments in chronology coincided with the rush hours, which usually associated with the growth of CO mole fractions in the surface atmosphere because of the enhanced anthropogenic emission (Kashin et al. 2010; Rakitin et al. 2011). At stage (2), diurnal cycle of CO, like for other components, did not show a clear tendency and lied below the cycles of stages (1) and (3). A small increase in CO mole fraction was registered only in the morning hours,

**Table 2** Mean values and standard deviations (SD) of hourly average CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> concentrations for 3 days periods during lockdown restrictions in Ekaterinburg

Date	Mean CO <sub>2</sub> (SD)	Mean CH <sub>4</sub> (SD)	Mean PM <sub>2.5</sub> (SD)	Date	Mean CO <sub>2</sub> (SD)	Mean CH <sub>4</sub> (SD)	Mean PM <sub>2.5</sub> (SD)
Mar 28–Mar 30	422.0 (4.0)	2005 (60)	3.5 (1.4)	Apr 15–Apr 17	418.6 (3.6)	1984 (27)	3.8 (2.5)
Mar 31–Apr 2	425.0 (7.3)	2013 (76)	4.0 (2.7)	Apr 18–Apr 20	421.5 (9.0)	2010 (79)	3.3 (3.6)
Apr 3–Apr 5	423.4 (5.1)	2013 (62)	5.5 (2.9)	Apr 21–Apr 23	419.3 (3.8)	1992 (75)	5.4 (5.0)
Apr 6–Apr 8	419.3 (1.6)	1975 (11)	1.7 (0.9)	Apr 24–Apr 26	417.2 (3.5)	1996 (63)	1.9 (1.2)
Apr 9–Apr 11	422.4 (6.7)	2029 (75)	4.7 (3.2)	Apr 27–Apr 29	424.3 (10.9)	2027 (107)	4.3 (3.3)
Apr 12–Apr 14	422.7 (5.7)	2008 (57)	5.9 (3.9)	Apr 30–May 2	418.0 (3.0)	1987 (31)	2.2 (1.4)





**Fig. 2** Mean diurnal cycles of hourly  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{PM}_{2.5}$  and  $\text{CO}$  concentrations during lockdown of 2020. Shadows indicate 95% confidence intervals

that apparently reflects the mobility of key workers released from COVID-19 restrictions.

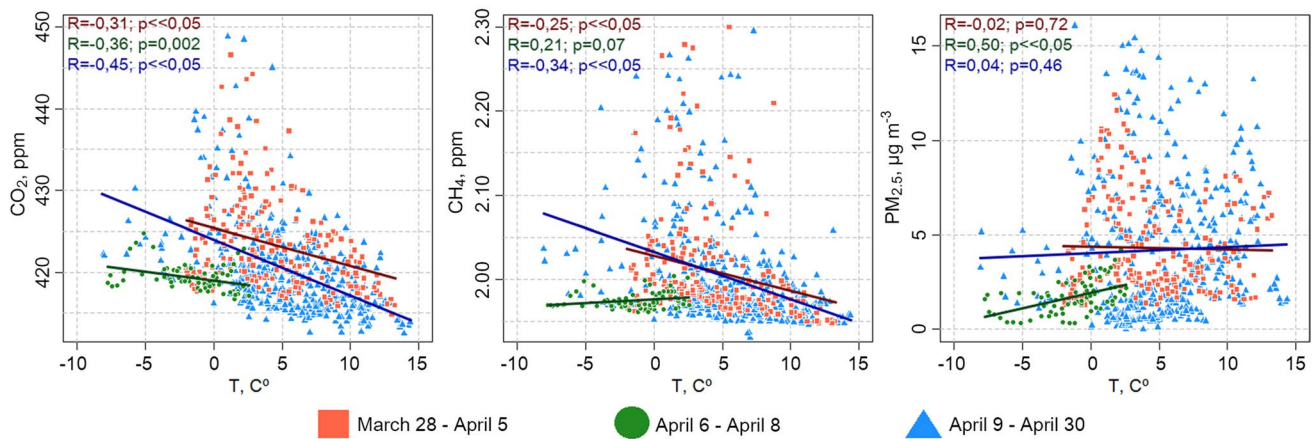
### Influence of air temperature

Along with diurnal cycles, synoptic variations in atmospheric  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{PM}_{2.5}$  can provide information on interaction between local- and regional-scale emissions and the atmosphere. During the growing season, an increase in air temperature within  $20^\circ\text{C}$  is known to intensify  $\text{CO}_2$  photosynthetic sequestration (Baldocchi et al. 2001; Law et al. 2002) and, consequently, to decrease its atmospheric concentration. In early spring, in the range of  $0\text{--}5^\circ\text{C}$ ,  $\text{CO}_2$  mole fraction can grow due to effluxes from snow cover melting (Sullivan et al. 2012). The  $\text{CH}_4$  content in the surface atmosphere in the absence of strong anthropogenic sources usually tends to grow with an increase in temperature due to the methanogenesis. This process occurs most actively at air temperature of  $30\text{--}40^\circ\text{C}$ , but in some cases increased methane effluxes are also observed immediately after the snow cover melts (Ginzburg et al. 2011; Harriss et al. 1982; Vinogradova et al. 2007). The anthropogenic factors in urban areas can significantly distort the expected positive correlation between atmospheric  $\text{CH}_4$  and air temperature. For

instance, at low temperatures, concentrations may increase due to vehicle emissions (Anisimov et al. 2014; Weilenmann et al. 2009). With a relatively constant industrial activity, this is the main factor affecting the temperature dependencies. As for  $\text{PM}_{2.5}$  in urban environment, it can serve as an anthropogenic marker, since the main sources of its emission are enterprises of the fuel-energy complex, vehicles, construction, etc. (ChooChuay et al. 2020; Zhang and Cao 2015). With an increase in air temperature, the conditions for the formation of secondary finely dispersed aerosols are improved, especially due to particle resuspension in drier weather; therefore, an increase in  $\text{PM}_{2.5}$  concentration in the atmosphere can be expected (Gubanova et al. 2018). However, in spring, an increase in  $\text{PM}_{2.5}$  concentration can be partly compensated by enhanced air convection due to surface temperature growth, leading to a significant weakening of the positive correlation.

At the above described stages of the lockdown in Ekaterinburg, the measurements demonstrate different patterns of “concentration–air temperature” correlations (Fig. 3). For  $\text{CO}_2$ , significant negative correlations at all stages ( $R$  varies from  $-0.31$  to  $-0.45$ ,  $p < 0.05$ ) apparently indicate the dominance of natural processes in forming the surface concentrations. At low temperatures, some contribution can





**Fig. 3** Hourly average concentrations of the investigated components as a response to air temperature during each period of COVID-19 restrictions in Ekaterinburg

also be made by anthropogenic emissions, but this effect is less noticeable at stage (2).  $\text{CH}_4$  showed negative response to air temperature ( $p < 0.05$ ) at stages (1) and (3), which, on the contrary, may indicate a strong influence of man-made emissions. At stage (2), when there was a substantial reduction in vehicular traffic, the correlation tended to be positive, i.e. similar to natural patterns (due to spring methanogenesis). For  $\text{PM}_{2.5}$ , a positive significant correlation ( $R = 0.50$ ,  $p < 0.05$ ) with air temperature was also found only at stage (2), which was likely caused by the effect of industrial emissions and drier weather associated with an increase in temperature. As mentioned above, industrial activity in Ekaterinburg did not decrease during the lockdown period. Thus, the greatest changes in “concentration–air temperature” correlations (toward natural patterns) occurred precisely at stage (2), when a strict “self-isolation” regime was enacted followed by a sharp reduction of people’s mobility. These results confirm the short-term effect of COVID-19 lockdown on  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{PM}_{2.5}$  concentrations in the surface atmosphere of Ekaterinburg.

### Difference between measured and background concentrations

To distinguish the anthropogenic signal in the observed GHGs in urban environment during COVID-19 lockdown, the measured daily average data were compared with corresponding background levels. Background levels were estimated as described above (see “Data analysis methods” for details). The mean difference between measured and background mole fractions for a given time interval can be simply referred to as “the excess”. Evolution of the excess allows to detect the change in the anthropogenic contribution to the

observed concentrations. For example, it was shown earlier (Gulyaev et al. 2020) that the mean excess for urban area is minimal in spring comparing with other seasons, which is apparently associated with phenological effect of local ecosystems, providing partly an offset of anthropogenic emissions. In 2017–2018 in Ekaterinburg, the spring excess was approximately 4–5 ppm for  $\text{CO}_2$  and 20–30 ppb for  $\text{CH}_4$ , which was 1–1.5% relative to the background levels for both gases (Gulyaev et al. 2020).

The monthly average excesses of measured over background  $\text{CO}_2$  and  $\text{CH}_4$  mole fractions for the spring period of 2020 are shown in Table 3. In general, the values in March–April for  $\text{CO}_2$  were close to the previously obtained estimations of 4–5 ppm. For  $\text{CH}_4$ , values higher than range of 20–30 ppb were observed with a significant increase in May up to 58 ppb (3% relative to the background), suggesting the possible influence of additional methane sources which was not previously manifested in measured data. Moreover, “the mean excess” of 10.8 ppm (2.7%) for  $\text{CO}_2$  in May was also more than twice those in March–April of 2020, which can be accounted for by mitigation of severe COVID-19 restrictions and the subsequent increase in population activity.

A detailed study of the lockdown period in Ekaterinburg showed that “the mean excess” at stages (1) and (3) did not statistically differ (for 95% confidence interval) from

**Table 3** The mean excess of measured daily average  $\text{CO}_2$  and  $\text{CH}_4$  mole fractions (in ppm, ppb and percentage) over corresponding background levels for the spring months of 2020 in Ekaterinburg

Component	March	April	May
$\text{CO}_2$ , ppm (%)	4.4 (1.1)	4.7 (1.1)	10.8 (2.7)
$\text{CH}_4$ , ppb (%)	31 (1.6)	43 (2.2)	58 (3.0)

**Table 4** The mean excesses and standard errors for three stages of 2020 lockdown in Ekaterinburg

Component	(1) From March 28 to April 5	(2) From April 6 to April 8	(3) From April 9 to April 30
CO <sub>2</sub> , ppm	5.7 ± 1.0	1.3 ± 0.2	5.1 ± 0.8
CH <sub>4</sub> , ppb	48 ± 10	8 ± 4	49 ± 5

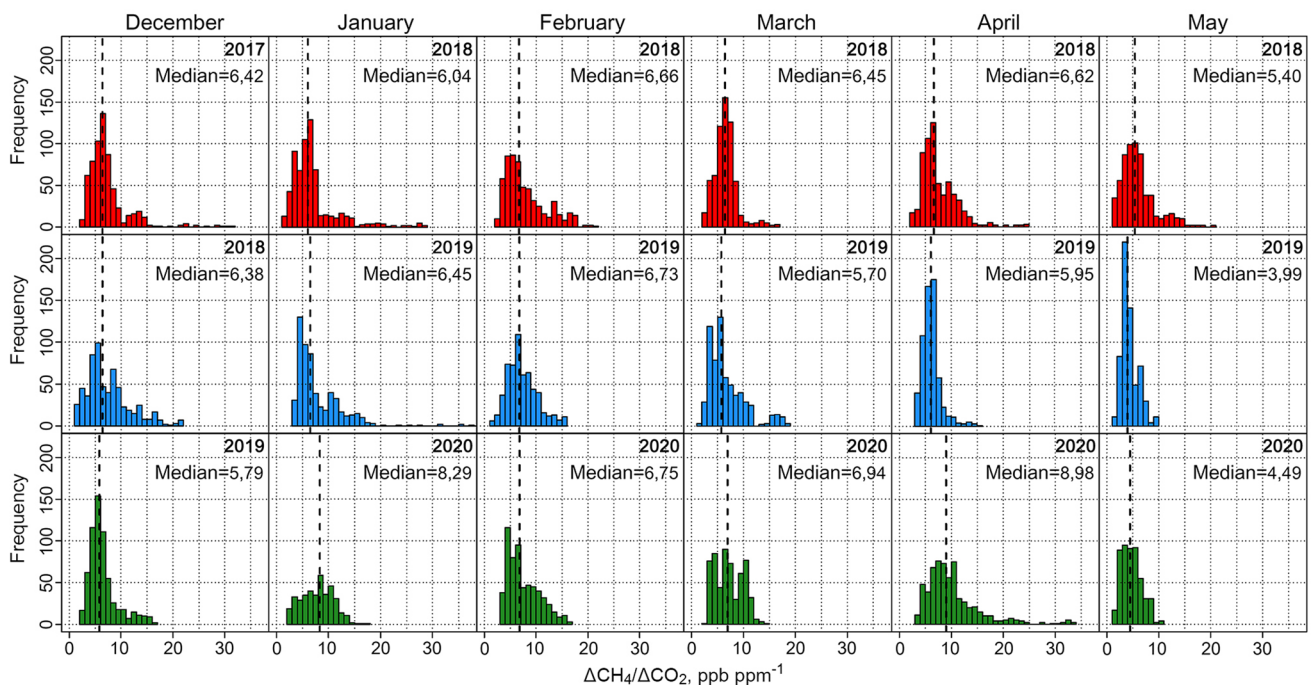
the monthly average values in March–April for both CO<sub>2</sub> and CH<sub>4</sub> (Table 4). While at stage (2) there was a multiple decrease in the differences between measured and background concentrations for both components. Thus, the short-term effect of COVID-19 restrictions on atmospheric CO<sub>2</sub> and CH<sub>4</sub> content is further evident from the revealed patterns of a decrease in anthropogenic excess.

### $\Delta\text{CH}_4/\Delta\text{CO}_2$ ratio: potential emission sources

Figure 4 shows frequency distributions of  $\Delta\text{CH}_4/\Delta\text{CO}_2$  ratios, ppb ppm<sup>-1</sup>, aggregated for each month of winter and spring of 2017–2020 in Ekaterinburg. The used method for calculation of the  $\Delta\text{CH}_4/\Delta\text{CO}_2$  enables indirectly assessment of the relative intensity of CO<sub>2</sub> and CH<sub>4</sub> emission by various sources (Chandra et al. 2016; Mahata et al. 2017). As can be seen, for all months,  $\Delta\text{CH}_4$  and  $\Delta\text{CO}_2$  ratios tended to be lognormal distributed with right-sided asymmetry. The

greatest variability of this indicator was typical for the winter months and achieved 30–35 ppb ppm<sup>-1</sup> in January 2019. At the same time, the medians changed insignificantly from 5.8 to 6.8 ppb ppm<sup>-1</sup>, excluding 8.3 ppb ppm<sup>-1</sup> in January 2020. This enhanced value may be caused by the incomplete data set for the specified period because of mains interruption and instrument maintenance. In winter, the influence of natural fluxes on atmospheric GHG concentrations is known to be minimal due to inactive phenological phase. Therefore, the highly variable  $\Delta\text{CH}_4/\Delta\text{CO}_2$  rather characterizes anthropogenic sources with different ratios of methane and carbon dioxide emissions, such as municipal boiler and sewage facilities, vehicles, construction, etc.

It is to be noted that increased variability of  $\Delta\text{CH}_4/\Delta\text{CO}_2$  was also observed in April 2020 when the strict lockdown was enacted. The frequency distribution was expanded significantly both relative to the previous months of the year and relative to the same periods of previous years. The median value in April 2020 also increased significantly up to 9.0 ppb ppm<sup>-1</sup>, which was higher than winter levels. This phenomenon may be attributed to a decrease in CO<sub>2</sub> emissions from fossil fuels and vehicles associated with the lockdown regime in Ekaterinburg at that period. Moreover, the effect of a local methane source was manifested, which was previously less significant.

**Fig. 4** Monthly frequency distributions of the  $\Delta\text{CH}_4/\Delta\text{CO}_2$  in winter and spring of 2017–2020 in Ekaterinburg. Dashed lines indicate median values



## Conclusion

Hourly and monthly average CO<sub>2</sub> and CH<sub>4</sub> mole fractions in the surface atmosphere of Ekaterinburg from December 1, 2017 to May 31, 2020 demonstrated the typical seasonal dynamics, despite the presence of a large number of local anthropogenic sources in the urban environment. The annual quasi-harmonic fluctuations of the mole fractions had highest values in winter months and lowest values in July–August for CO<sub>2</sub> and in May–June for CH<sub>4</sub>. But the seasonal amplitude was less pronounced for methane. Fine aerosol PM<sub>2.5</sub> was characterized by increased concentrations in winter; however, during the rest of the year, there was no clear trend in changes in its atmospheric content. For all components, the data averaged over each month did not show the influence of COVID-19 restrictions on atmospheric concentrations. At the same time, there was a significant increase in CO<sub>2</sub> and CH<sub>4</sub> mole fractions in May 2020 compared to the same periods of previous years, which could be a consequence of the gradual lifting of the enforced confinements, as well as of the violation of “self-isolation” regime (voluntary restrictions) during May holidays in Russia.

The detailed examination of the lockdown period from March 28 to April 30, 2020 allowed to reveal a multiple decrease in the inter-hourly variations of all studied components at stage (2) (at the beginning of strict “self-isolation” period from April 6 to April 8). The absolute CH<sub>4</sub> and PM<sub>2.5</sub> concentrations at this stage were minimum for the entire lockdown period and amounted to 1975 ppb and 1.7 μg m<sup>-3</sup>, respectively. The CO<sub>2</sub> mole fraction of 419.3 ppm was not the lowest on these days which indicates the phenological features of this season. During the spring 2020, the decrease in CO<sub>2</sub> mole fraction was due to both “self-isolation” effect and natural sink and it was difficult to quantitatively distinguish the contribution of these processes.

The short-term effect of COVID-19 lockdown on atmospheric CO<sub>2</sub>, CH<sub>4</sub> and PM<sub>2.5</sub> was further shown from the mean diurnal cycles of hourly concentrations. At stage (2), there was a significant decrease in daily variations in gas and aerosol concentrations. Given the relatively constant industrial activity, the results obtained can be associated with a decrease in the mobility of urban population and, accordingly, with substantial reduction in vehicular traffic. This conclusion was also confirmed by the analysis of correlation between CO<sub>2</sub>, CH<sub>4</sub>, PM<sub>2.5</sub> concentration and air temperature during each period of the restrictions in Ekaterinburg. Thus, the development of urban public transport can become a possible instrument for reducing air pollution, namely renewal of the transport fleet, creation of dedicated lanes, development of the route network, transition to more environment-friendly fuels, etc. Availability of the

developed public transport can spur some people to stop using private vehicular regularly and therefore can lead to a decrease in the traffic load.

The mean excess of measured daily average CO<sub>2</sub> and CH<sub>4</sub> mole fractions over corresponding background levels in May 2020 was 10.8 ppm and 58 ppb, respectively. These values were significantly higher than those in March–April 2020 suggesting the increase in economic activity of the population during the period of voluntary restrictions in May. The study of the strict lockdown period showed that at stage (2) there was a multiple decrease in the difference between measured and background concentrations of both components. The excess was 1.3 ± 0.2 ppm (mean ± SE) for CO<sub>2</sub> and 8 ± 4 ppb for CH<sub>4</sub>. This also proves the short-term effect of quarantine restrictions on atmospheric concentrations of the considered trace gases. Moreover, the higher median value (9.0 ppb ppm<sup>-1</sup>) and variability of ΔCH<sub>4</sub>/ΔCO<sub>2</sub> ratios in April 2020, when the strict lockdown was enacted, were obtained. This effect was likely to bring about by the short-term decrease in CO<sub>2</sub> anthropogenic emissions, as well as by the influence of a local source of CH<sub>4</sub>.

The COVID-19 pandemic has altered the lives of millions of people around the world. Obviously, the most urgent task was to overcome the growing crisis of public health and socio-economic development. On the other hand, the pandemic forced confinements represent an unexpected global “experiment” on the climate system. The obtained data make it possible to investigate the response of the climate system to relatively short-term changes in atmospheric emissions. But many recent studies have failed to establish lockdown effect on concentrations of trace gases and aerosols in the atmosphere. In this work, we have succeeded it by analyzing different types of concentration variability and a specific complex of methane and carbon dioxide mole fractions. Certainly, these results are rather local, but the approaches can be used in other areas for evaluating GHG and aerosol fluctuations caused by weak and short-term changes in its sources and sinks. The findings showed that the reduction of negative human impact on the atmosphere resulted from enforced and voluntary restrictions can be insufficient and short-term and quickly resumes when economic and public activity recovers. When meeting a deferred demand, the impact on the environment may even be more powerful than in the pre-pandemic times. Thus, in order to improve urban air quality, measures are required to ensure long-term emissions reduction. Instrumental data obtained during lockdowns provide experimental information on the effect size of improving air quality that can be expected with a reduced level of emissions during these periods.



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**Availability of data and material** The datasets generated and analyzed in the current study are available from the corresponding author on reasonable request.

**Code availability** Not applicable.

## Declarations

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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