

The impact of COVID-19 lockdowns on urban photochemistry as inferred from TROPOMI

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HIGHLIGHTS

- The impact of Covid-19 lockdowns on the NO_x lifetime is derived using TROPOMI retrieved NO₂/CO ratios for six megacities.
- Covid-19 lockdowns reduced the NO_x lifetime compared to the year before across the six megacities.
- TROPOMI derived reduction in NO_x lifetime over Denver agrees with CLASS model initialized using ground-based observations.

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ABSTRACT

Movement restrictions were imposed in 2020 to mitigate the spread of Covid-19. These lock-down episodes provide a unique opportunity to study the sensitivity of urban photochemistry to temporary emission reductions and test air quality models. This study uses Tropospheric Monitoring Instrument (TROPOMI) nitrogen dioxide/carbon monoxide (NO₂/CO) ratios in urban plumes in combination with an exponential fitting procedure to infer changes in the NO_x lifetime (τ_{NO_x}) during Covid-19 lock-downs in the cities of Denver, Chicago, New York, Riyadh, Wuhan and Sao Paulo compared with the year before.

The strict lockdown policy in Wuhan led to a 65–80% reduction in NO₂, compared to 30–50% in the other cities that were studied. In New York and Wuhan, CO concentration was reduced by 10–15%, whereas over Riyadh, Denver, Chicago, and Sao Paulo the CO background concentration increased by 2–5 ppb. τ_{NO_x} has been derived for calm ($0.0 < U \text{ (m/s)} < 3.5$) and windy ($5.0 < U \text{ (m/s)} < 8.5$) days to study the influence of wind speed. We find reductions in τ_{NO_x} during Covid-19 lockdowns in all six megacities during calm days. The largest change in τ_{NO_x} during calm days is found for Sao-Paulo ($31.8 \pm 9.0\%$), whereas the smallest reduction is observed over Riyadh ($22 \pm 6.6\%$). During windy days, reductions in τ_{NO_x} are observed during Covid-19 lockdowns in New York and Chicago. However, over Riyadh τ_{NO_x} is almost similar for windy days during the Covid-19 lockdown and the year before.

Ground-based measurements and the Chemistry Land-surface Atmosphere Soil Slab (CLASS) model have been used to validate the TROPOMI-derived results over Denver. CLASS simulates an enhancement of ozone (O₃) by 4 ppb along with reductions in NO (38.7%), NO₂ (25.7%) and CO (17.2%) during the Covid-19 lockdown in agreement with the ground-based measurements. In CLASS, decreased NO_x emissions reduce the removal of OH in the NO₂ + OH reaction, leading to higher OH concentrations and decreased τ_{NO_x} . The reduction in τ_{NO_x} inferred from TROPOMI ($28 \pm 9.0\%$) is in agreement with CLASS. These results indicate that TROPOMI derived NO₂/CO ratios provide useful information about urban photochemistry and that changes in photochemical lifetimes can successfully be detected.

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1. Introduction

The movement restriction policy implemented by national governments to control the spread of the Covid-19 virus in 2020 had many social, economic and environmental impacts (Bonaccorsi et al., 2020; Chauhan and Singh, 2020; Dhaka et al., 2020; McKibbin and Fernando, 2021). Among the environmental impacts are improvements in air quality, due to reduced emissions of air pollutants associated with the reduced economic activity (Le et al., 2020; Misra et al., 2021). In the past few years, new satellites have been deployed that improve the capacity to monitor air pollution world-wide from space. These measurements offer a great opportunity to investigate the impact of Covid-19 lockdowns on urban air quality. Several studies have demonstrated that trends in urban NO_x emissions can be quantified using satellite observations of NO₂ (Beirle et al., 2019; Liu et al., 2017; Lorente et al., 2019). Urban CO emissions can be quantified using satellites also, but less accurately than NO_x due to its longer lifetime, complicating the separation of the influence of urban emissions on the observed total column abundances from the background. Nevertheless, a few studies have quantified changes in CO emission from cities using satellite observations (Beirle et al., 2019; Borsdorff et al., 2019; Dekker et al., 2017, 2019; Pommier et al., 2013).

The reduced energy use during Covid-19 lockdowns provides a 'natural experiment' to test the capacity of the existing measurement and modelling infrastructure to monitor the energy transition and its influences on urban air quality. Over central Chinese cities, satellite observations show a decline in tropospheric column NO₂ by 30.3%–65.2% (Le et al., 2020). CO mixing ratios decreased by 16.2%–37.9% in ground based measurements (Xu et al., 2020). In the European city of Milan, ground based measurements show a decrease of NO₂ by 40%–55% and CO by 25%–30% (Bray et al., 2020; Romano et al., 2021; Virghileanu et al., 2020). Similar reductions in NO₂ over Milan were detected by Ozone Monitoring Instrument (OMI) observations (Bray et al., 2020). In New York, OMI show reduction in NO₂ tropospheric column by 28%–50% and CO concentration decreased by 7%–10% in ground based measurement (Bray et al., 2020). This confirms that reductions in air pollution levels during Covid-19 lockdowns have been measured on ground, but can also be observed from space (Filonchik et al., 2020; Goldberg et al., 2020; Lange et al., 2021). Miyazaki et al. (2021) studied the change in global photochemistry in response to Covid-19 lockdown and indicate that the free troposphere became less oxidative in response to reductions in NO_x emissions. So far, however, changes in the urban photochemical regime and the chemical turn over time of pollutants such as NO_x in urban plume received less attention.

NO_x plays an important role in atmospheric chemistry as it regulates the photochemical production of ozone (O₃), for instance in photochemical smog formation, and atmospheric oxidation rates through the recycling of HO_x (OH + HO₂). During daytime, the NO_x lifetime (τ_{NO_x}) is controlled by the reaction between the hydroxyl radical (OH) and NO₂ forming HNO₃. The tropospheric OH concentration is strongly influenced by the NO_x concentration due to the non-linear recycling of HO_x, causing τ_{NO_x} to be depended on its own concentration (Valin et al., 2013). Additionally, NO_x is removed in the formation of dinitrogen penta-oxide (N₂O₅), peroxyacetyl nitrate (PAN), and organic nitrates (RONO₂).

In the past, OMI NO₂ retrievals have been used to determine the NO_x emission and lifetime in urban plumes using the exponential modified gaussian (EMG) method by Beirle et al. (2011). The EMG method was modified by Liu et al. (2016) for application to complex emission distributions. The variation of τ_{NO_x} in urban plumes with wind speed has been demonstrated by Valin et al. (2011). The impact of strengthened air pollution mitigation measures in US cities on τ_{NO_x} has been estimated by Laughner and Cohen (2019) using an exponential fitting method applied to OMI NO₂ observations. Recently, Lama et al. (2022) presented a method for estimating τ_{NO_x} and the hydroxyl radical in urban plumes, using the regional chemistry transport model WRF in combination with

the satellite retrievals of NO₂ and CO. This method has the advantage that it can be applied to single satellite overpass. However, the use of an atmospheric transport model is computationally expensive. In this study we adopt a simplified approach using the Tropospheric Monitoring Instrument (TROPOMI) derived NO₂/CO ratio to estimate τ_{NO_x} in urban plumes during Covid-19 lockdowns in comparison with other years. On 13 October 2017, the Sentinel-5 Precursor satellite was launched with TROPOMI instrument on board (Veefkind et al., 2012). TROPOMI measures NO₂ tropospheric column and CO total column with high sensitivity, high spatial resolution and daily global coverage and is therefore well suited for this task. In this method, CO is used due to its longer lifetime than NO₂ and can be considered as inert tracer over the urban area of interest. The difference in the rate of decay between NO₂ and CO provides information about the photochemical oxidation of NO₂, because atmospheric dispersion is expected to have a very similar impact on both tracers and therefore cancels out in their ratio.

This paper is organized as follows: Section 2 describes the TROPOMI NO₂ and CO data that were used, the method used to estimate τ_{NO_x} using TROPOMI derived NO₂/CO ratios and the sensitivity tests that are performed to evaluate the robustness of the method. The impact of Covid-19 lockdowns on urban τ_{NO_x} is presented in section 3, followed by its validation using ground based observations and CLASS. The main outcomes of this study are discussed and summarized in sections 4 and 5. Additional figures supporting the result of this study can be found in the supplement.

2. Data and method

2.1. TROPOMI NO₂ tropospheric column

We used the S5P-PAL reprocessed NO₂ [mole/m²] data product available at <https://data-portal.s5p-pal.com/products/no2.html>. In December 2020, the NO₂ processor v1.3.2 was upgraded to v1.4.0. The updated FRESCO cloud retrieval in v1.4.0. increased the tropospheric NO₂ column by 0%–50% in polluted areas under clear sky conditions (Eskes and Eichmann, 2022; Riess et al., 2022; Van Geffen et al., 2022). In contrast, adjustments in surface albedo in v2.2.0 increased the tropospheric NO₂ columns by 10%–15% for polluted cloud free scenes (Eskes and Eichmann, 2022). Therefore, to study the impact of Covid-19 lockdowns on the observed trends in pollutants, NO₂ data products v1.2.x and 1.3.x cannot be combined with v1.4.0 and v2.2.0. To restore the consistency of the full dataset, the TROPOMI NO₂ data set has been reprocessed for the period from May 1, 2018 to Nov 14, 2021 using the latest operational NO₂ processor version 2.3.1.

The TROPOMI NO₂ DOAS software developed by KNMI is used to process NO₂ slant column densities (van Geffen et al., 2019). The slant column densities are converted into tropospheric column densities using an improved version of the QA4ECV NO₂ algorithm (Boersma et al., 2011, 2018). Here, tropospheric slant column densities are obtained by subtracting the stratospheric contribution from slant column densities. Tropospheric slant columns are converted into tropospheric vertical column densities by applying air mass factor (AMF), as function of surface albedo, terrain height, cloud height, cloud fraction and a priori vertical NO₂ profile from TM5 model at 1° × 1° resolution (Eskes et al., 2018; Lorente et al., 2017). MAXDOAS ground based measurements over European cities indicate that TROPOMI underestimates tropospheric column NO₂ by 7%–29.7% (Lambert et al., 2019). This bias arises from the coarse resolution a priori NO₂ profiles used in the AMF calculation.

2.2. TROPOMI CO

In this study, offline level 2 CO data version 1.3.x has been used available at <https://cophub.copernicus.eu/s5pexp> (last access: 20 September 2020). The total CO column [molec cm⁻²] is retrieved from TROPOMI 2.3 μm spectra using the SICOR algorithm (Landgraf et al.,

2016). This algorithm uses the profile scaling approach in which TROPOMI-observed spectra are fitted by scaling a reference vertical profile of CO using the Tikhonov regularization technique (Borsdorff et al., 2014). TM5 transport model based reference CO profiles are used in this method (Huijnen et al., 2019; Krol et al., 2005). The sensitivity of TROPOMI-derived total CO columns to the vertical profile of CO is defined by the averaging kernel that is obtained from each retrieval. TROPOMI inferred CO is compared with 28 different Total Carbon Column observing Network (TCCON) measurements, which showed a difference of $9.1 \pm 3.3\%$ (Sha et al., 2021).

2.3. Satellite data selection and filtering criteria

To study the impact of Covid-19 lockdowns on the urban lifetime of NO_x (τ_{NO_x}) six different megacities i.e. Wuhan, Riyadh, Sao Paulo, New York, Denver, and Chicago have been selected. These megacities have been selected to compare τ_{NO_x} under NO_x limited (US cities) and VOC limited regimes (Asian and South American cities: Wuhan, Riyadh and Sao Paulo). Table 1 shows the time periods that have been selected for these six megacities, which vary because the Covid-19 lockdown period differs from country to country. TROPOMI NO_2 and CO data selected during the Covid-19 lockdown in 2020 are compared with same period in 2019 to eliminate seasonal influences, except for Wuhan. In Wuhan, the Covid-19 lockdown period started from 23rd Jan to 23rd March 2020 but TROPOMI data from 2nd Feb to 23rd March 2020 and 10th Feb to 23rd March 2019 have been selected to avoid the Chinese new year holiday in both years, and the associated reductions in anthropogenic emission (Gong et al., 2014). Note that hereafter the time period selected in 2020 is referred as “Covid-19” and the 2019 reference period as “pre-Covid-19”.

TROPOMI NO_2 and CO are derived using different channels in TROPOMI and different retrieval algorithms. Because of this, the NO_2 and CO data products have different spatial resolutions and filtering criteria. For data filtering, both algorithms provide a quality assurance value (qa), which ranges from 0 to 1. NO_2 retrievals with $qa > 0.75$ have been selected representing clear sky conditions. For CO, retrievals with $qa \geq 0.70$ have been selected indicating clear sky conditions or the presence of low level clouds. The application of the SICOR algorithm to SCIAMACHY data showed that the number of valid observations could be increased by accounting for low level clouds (Borsdorff et al., 2018b, a). The CO data were used after stripe filtering following Borsdorff et al. (2018c). The surface pressure in the CO and NO_2 data is used to translate total column CO densities and tropospheric NO_2 column densities into dry column mixing ratios of XCO (ppb) and XNO_2 (ppb). After data filtering, CO and NO_2 data have been co-located by allocating the centre co-ordinate of NO_2 retrievals to the CO footprints to account for the difference in spatial resolution between NO_2 data (at $3.5 \times 7 \text{ km}^2$) and CO data (at $5.5 \times 7 \text{ km}^2$) as in Lama et al. (2022, 2020).

2.4. Separation of calm and windy days

The boundary layer averaged wind speed and wind direction are

Table 1

Cities and time periods selected to study the impact of Covid-19 lockdowns on the NO_x lifetime.

City	Selected time period in 2019 (Pre-Covid-19)	Selected time period in 2020 (Covid-19)
Riyadh	1 st April to 10 th May	1 st April to 31 st May
Denver	1 st April to 31 st May	1 st April to 31 st May
New York	15 th March to 31 st of May	15 th March to 31 st of May
Chicago	26 th March to 31 st of May	26 th March to 31 st of May
Wuhan	10 th Feb to 23 rd March	2 nd Feb to 23 rd March
Sao Paulo	24 th March to 10 th May	24 th March to 10 th May

taken from ECMWF Reanalysis v5 (ERA5) data (retrieved at <https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-pressure-levels?tab=overview>, last access: 10th March 2021) at a spatial resolution of $0.25^\circ \times 0.25^\circ$ and 1 hourly temporal resolution. ERA5 wind vectors have been spatially and temporally interpolated to the footprints of the TROPOMI pixels. Days are labelled as calm or windy when the average wind speed within a 50 km radius from the centre of city is in the range of $0 \leq U \text{ [m/s]} \leq 3.5$ and $5.0 \leq U \text{ [m/s]} \leq 8.5$, respectively. This segregation is used to study the dependence of the NO_x lifetime on the wind speed as in Valin et al. (2013). There are 6–17 clear sky observation for calm and windy days across the six megacities to quantify the NO_x lifetime change during pre-Covid-19 and Covid-19 (see Table S1).

2.5. Method for quantifying the NO_x lifetime

The box rotation method from Lama et al. (2022) has been used to determine the NO_2/CO ratio in the urban plume as function of the downwind distance from the city centre. The boxes have a width of 200 km, and lengths of 100 km in upwind direction and 300 km in downwind direction. The box is rotated for every overpass in the direction of the wind using the averaged wind direction at 100 km radius from the centre of city. The rotated box B1 (see Fig. S1) is divided into N rectangular boxes, with lengths (Δx) of 11 km. The XNO_2 , XCO and wind speed grid cells that fall within the N rectangular boxes are selected to compute averaged NO_2 , CO and NO_2/CO ratios across the wind for each small boxes. The enhancement of XNO_2 and XCO over the background declines at larger distance from the city resulting in inaccurate $\Delta\text{XNO}_2/\Delta\text{XCO}$ ratios. To avoid this, we use the mean XNO_2/XCO ratio instead of enhancement ratios. The travel time [hr] from the city centre for each small box is calculated by dividing the downwind distance by the averaged wind speed for each small box (see Fig. S1).

The decline in the mean of single TROPOMI orbit XNO_2/XCO ratios with the travel time is used to determine the lifetime of NO_x . An exponential function is fitted to the XNO_2/XCO ratios starting from the peak value in the city centre up to 300 km in downwind direction in the form of $y = Ae^{(-x/K)} + c$ (see Fig. 1). Here, A is a fit coefficient quantifying the size of the urban perturbation in the background ratio, x is the travel time, K is τ_{NO_x} and c is the constant background ratio. Note that K represents τ_{NO_x} as the NO_x/NO_2 ratio presumably does not vary significantly in the downwind direction of the urban plume.

2.6. Synthetic performance test

To test the robustness of the method, a sensitivity test has been performed using WRF simulations of NO_x and CO from Lama et al. (2022) for the city of Riyadh. These results were obtained using the passive tracer transport option applied to EDGAR NO_x and CO emissions and CAMS-derived OH (for more details see Text S1 and Fig. S2). To determine the NO_x lifetime at mid-day, Lama et al. (2022) computed the first order loss of NO_x from the reaction between NO_2 and OH forming HNO_3 , ignoring other pathways of NO_x loss. In summer, the NO_x lifetime estimated by Lama et al. (2022) was within 10% of full chemistry computations in WRF. The WRF simulated summer (June to July 2018) averaged NO_2/CO ratio at the time of TROPOMI overpass for windy days ($5.0 \leq U \text{ [m/s]} \leq 8.5$) in the urban downwind plume defines test Case 1. In Case 2, emissions of NO_x and CO are reduced 50% and 20%, respectively, but the OH concentration is same as in Case 1. In Case 3, the OH concentration is increased by 40%, whereas NO_x and CO emissions are the same as in Case 1. Case 1 is compared with Case 2 and Case 3, to understand the impact of changed emissions and OH concentrations on the estimated τ_{NO_x} . The settings of the cases and the resulting τ_{NO_x} estimates derived from WRF and the exponential curve fitting method are listed in Table 2.

Fig. 1 shows WRF simulated XNO_2 , XCO, XNO_2/XCO and the exponential fit to the XNO_2/XCO ratio starting from the peak value until 300 km downwind of the city centre for Case 1 to 3. The width of the band of

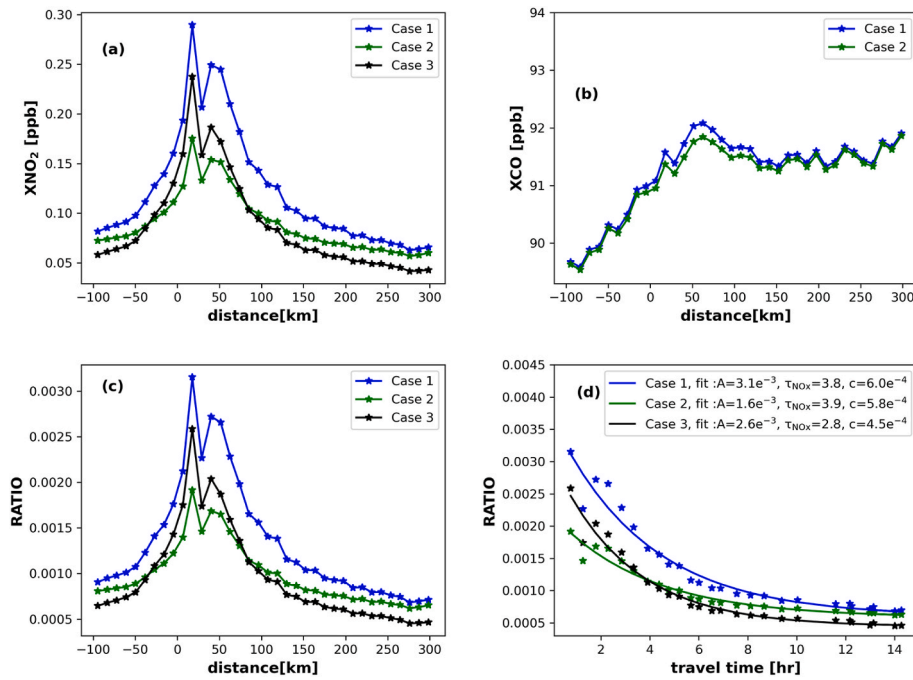


Fig. 1. WRF derived a) XNO_2 , b) XCO , c) XNO_2/XCO and d) exponential fit to the XNO_2/XCO ratio for Case 1–3. EDGAR NO_x and CO emissions and CAMS OH are input variables for the reference Case 1. In Case 2, NO_x and CO emissions are reduced by 50% and 20% respectively compared to Case 1. In the Case 3, CAMS OH is increased by 40% compared to Case 1. The travel time from the centre of the city is calculated as the ratio of the travel distance and the average wind speed.

Table 2

Evaluation of the exponential curve fitting method with WRF simulations for the city of Riyadh.

Sensitivity test	Description	τ_{NO_x} derived from WRF [hr] (A)	τ_{NO_x} derived from Exponential Fit [hr] (B)
Case 1	EDGAR NO_x and CO emission, CAMS derived OH, NCEP meteorological conditions and CAMS derived NO_x and CO boundary condition is used.	3.7 ± 0.25	3.8 ± 0.5
Case 2	EDGAR NO_x emission is reduced by 50% and EDGAR CO emission is reduced by 20%.	3.7 ± 0.25	3.9 ± 0.6
Case 3	CAMS derived OH is increased by 40%, EDGAR NO_x and CO emission as Reference run.	2.7 ± 0.1	2.8 ± 0.3

TROPOMI data we consider across the city is wide enough for the diffusing plume to stay within the band. In this case a constant CO enhancement is expected. As discussed in the introduction, Fig. 1 illustrates the exponential decay of NO_2 in the downwind urban plume, while CO concentrations remain relatively constant. By calculating the ratio of NO_2/CO , valuable insights can be gained regarding the lifetime of NO_x . We performed the bootstrapping method for this test to quantify the uncertainty with decreasing data availability on τ_{NO_x} . The 1000 samples are generated repetitively by reducing the WRF data size (N) to 50% of the original datasets (N_R). Then, τ_{NO_x} is estimated for each of the sample using the exponential curve fitting technique and subsequently, the relative difference between τ_{NO_x} derived for Case 1 compared to other cases is quantified. Using this 1000 samples, the mean and the standard deviation for τ_{NO_x} , and relative difference is derived for Case 1 to 3. For all the tests, τ_{NO_x} derived from WRF and the exponential curve fitting method differs $<6\%$ (see Table 2). The τ_{NO_x} derived from an exponential fit to NO_2 only is longer by 7.5–10.8% for Case 1 to 3 than using the NO_2/CO ratio. This confirms the benefit of applying the

exponential curve fitting method to the NO_2/CO ratio to estimate τ_{NO_x} in the downwind urban plume. The exponential curve fit successfully recovers the applied OH increase between Case 3 and Case 1 of 40% (37.0

± 3.7 , i.e. within 10%). Overall, the comparison between Case 1 and the other cases confirms that the exponential curve fitting method is sensitive to changes in OH, with sufficient skill in recovering the size of these changes, whereas τ_{NO_x} is insensitive to changes in NO_x and CO emissions (see Table 2).

2.7. Sensitivity test: using TROPOMI data

Lama et al. (2022) estimated the summer τ_{NO_x} over Riyadh is shorter by 30%–37.5% compared to winter using the EMG and WRF optimization methods. At northern mid-latitudes, the seasonal variation in the incoming solar radiation is larger, increasing the seasonal variation in τ_{NO_x} . To test whether our exponential fitting method is capable to resolve variations in τ_{NO_x} using TROPOMI data, it has been applied to TROPOMI derived NO_2/CO ratios over Riyadh and Denver in summer (June to August 2019) and winter (November 2019 to January 2020).

Summer and winter averages of TROPOMI retrieved XNO_2 and XCO are shown in Fig. 2 and S3 for a domain of $500 \times 500 \text{ km}^2$ centred around Riyadh and Denver. The XNO_2 and XCO enhancements over the background due to city emissions are detected sufficiently well by the satellite to study the seasonal variation in τ_{NO_x} . Summer XNO_2 is higher by 20%–50% compared to winter XNO_2 whereas XCO shows $<10\%$ seasonal changes over Riyadh. This is consistent with the use of air conditioning during hot days in Riyadh, increasing the power consumption in summer compared to winter (Lange et al., 2021). The summer and winter averaged XNO_2 , XCO and XNO_2/XCO ratios are derived using the box rotation method for windy days over Riyadh (see Fig. 3). Only windy days could be used for Riyadh in winter, due to insufficient data for calm days. τ_{NO_x} estimates obtained from XNO_2/XCO are lower by 7.1% in summer and 16.6% in winter than those obtained using XNO_2 only. The exponential fit to XNO_2/XCO shows that τ_{NO_x} is shorter in summer compared to winter by $50 \pm 14.7\%$ (see Fig. 3d). This means that the seasonal cycle in τ_{NO_x} derived from the exponential curve

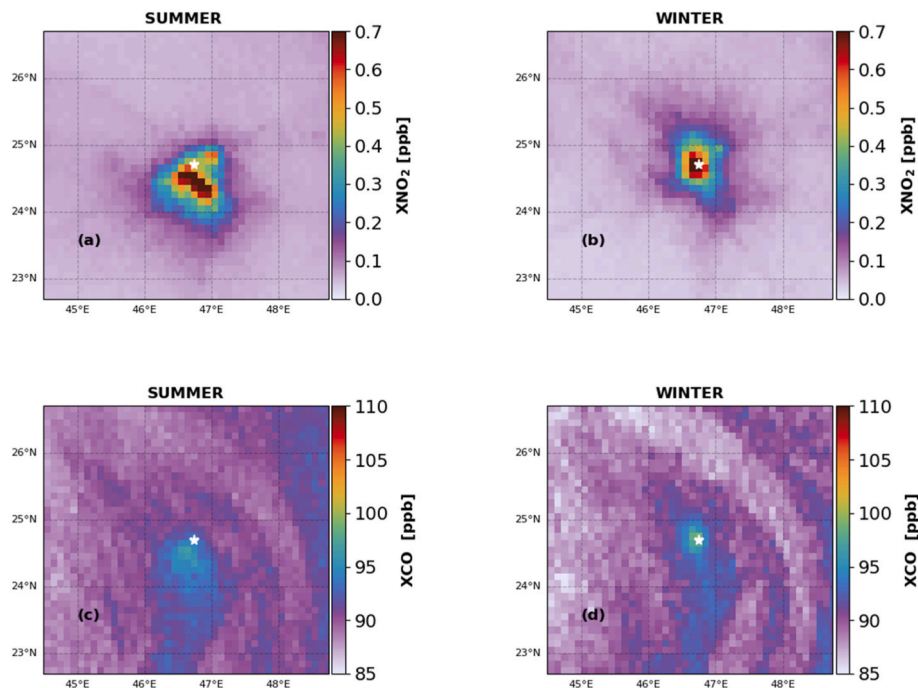


Fig. 2. TROPOMI retrieved XNO_2 (above) and XCO (below) in summer (June to August 2019) and winter (November 2019 to Jan 2020) over Riyadh. The white star represents the centre of Riyadh.

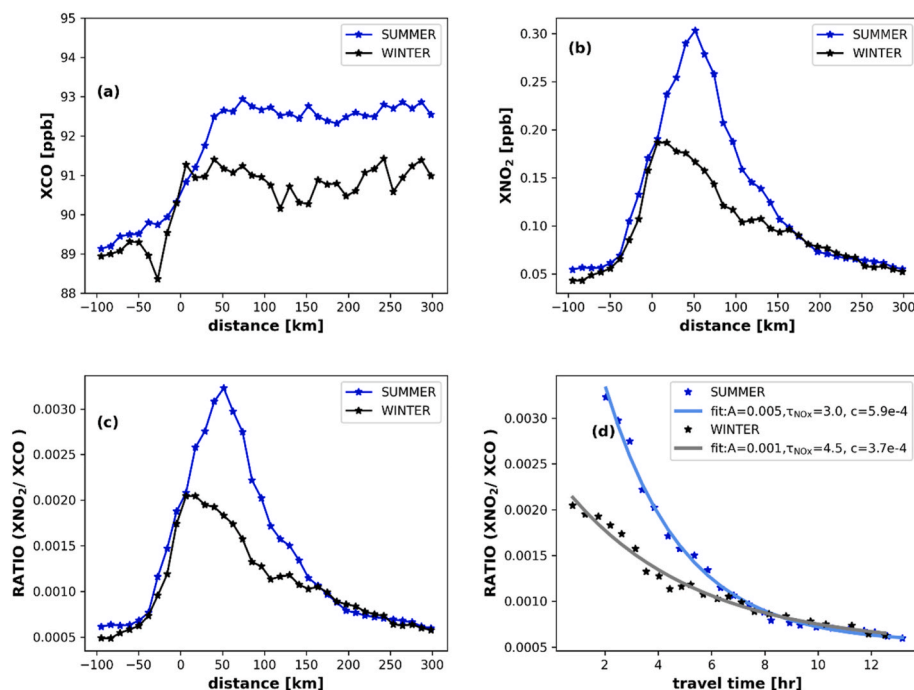


Fig. 3. A comparison of summer (June to August) and winter (Nov 2019 to Jan 2020) averages of TROPOMI for windy condition a) XCO , b) XNO_2 , c) XNO_2/XCO , and d) the exponential fit to the XNO_2/XCO ratio from the peak value to 300 km downwind distance from Riyadh. The travel time is computed from the ratio of the downwind distance and the averaged wind speed for each box.

fitting applied to TROPOMI XNO_2/XCO agrees with [Lama et al. \(2022\)](#). Additionally, boundary layer averaged OH from CAMS over Riyadh at the TROPOMI overpass time is higher by 51.8% in summer compared to winter, in agreement with the seasonal variation in τ_{NOx} derived from TROPOMI (see [Fig. S4a](#)).

In Denver, summer XNO_2 close to the centre of city is smaller by a factor 2.1 to 3.5 compared to winter, whereas the seasonal change in

XCO is 6%–12% (see [Figs. S3 and S5](#)). During winter, the increased power consumption due to residential heating contributes to higher XNO_2 and XCO concentrations compared to summer. The mean XNO_2 , XCO , and XNO_2/XCO ratio are derived for calm days due to insufficient data for windy days. Similar to Riyadh, τ_{NOx} derived from XNO_2/XCO over Denver is shorter by 17.9% in summer and 14% in winter than using XNO_2 only. [Fig. S5](#) shows that for calm days τ_{NOx} is shorter by 65.3

$\pm 11\%$ in summer compared to winter. This sensitivity test confirms that the exponential curve fitting method applied to TROPOMI derived XNO_2/XCO ratios in the downwind urban plume can be used to study the impact of Covid-19 lockdowns on τ_{NOx} . Additionally, CAMS OH is higher by 62% over Denver compared to winter in agreement with TROPOMI (see Fig. S4a).

2.8. Boundary layer atmospheric chemistry modelling

In this study, the Chemistry Land-surface Atmosphere Soil Slab (CLASS) model (van Stratum et al., 2012) has been used to support the interpretation of TROPOMI-estimated changes in the urban photochemistry due to the Covid-19 lockdown in Denver. In CLASS the day-time boundary layer growth is determined mostly by the sensible and latent heat fluxes and the entrainment of air from free troposphere. CLASS is a box model for well mixed atmospheric conditions, capable of successfully reproducing the observed mixing ratios of air pollutants and meteorological variables in the turbulent boundary layer (Van Stratum et al., 2012).

CLASS has a simplified Ox-NOx-VOC-HOx photochemistry scheme with 15 reactive species and 28 reactions. CLASS includes night-time chemistry such as the N_2O_5 formation and subsequent transformation to HNO_3 (Vila-Guerau de Arellano et al., 2009). The simplified chemistry scheme in CLASS incorporates isoprene and its oxidation products but neglects other volatile organic carbon compounds and aerosols (Ouwersloot et al., 2012). Nevertheless, CLASS is able to simulate the observed diurnal cycle and concentrations of important reactants in the polluted atmosphere, as has been shown such for the Amazon and Paris (Lorente et al., 2019; Van Stratum et al., 2012; Vila-Guerau de Arellano and Pino, 2009). CLASS has the advantage over other models that it is computationally cheap, but still includes diurnal changes in the boundary layer and can be used to interpret the various observations (Lorente et al., 2019; Zara et al., 2021).

CLASS has the advantage over 3D chemistry transport models that the settings can be adjusted relatively easily to reproduce observed air pollutants. The CLASS simulation starts at 8 LT (sunrise) with initial conditions for NO, NO_2 , CO and O_3 constrained from ground based

observations over Denver (see section 3.4 for details). Additionally, an initial boundary layer height at 8 LT is taken from ERA5 over Denver. The soil moisture is setup such that CLASS simulates a 2000 m boundary layer height at 13–14 LT as in ERA5. In our idealized CLASS simulation for clear sky conditions (without meteorological differences between pre-Covid-19 and Covid-19 lockdown), NO_x , CO, isoprene emissions and the dry deposition velocity of O_3 are adjusted to reproduce the observed diurnal variability of NO, NO_2 , CO, and O_3 mixing ratios.

3. Results

3.1. XNO_2 and XCO changes during Covid-19

To study an impact of Covid-19 lockdowns on urban photochemistry, TROPOMI data within a domain of $500 \times 500 \text{ km}^2$ over Riyadh are averaged from 1st April to 31st May 2020 and for the same period in 2019 (see Fig. 4). TROPOMI-derived XNO_2 and XCO enhancements are separated clearly from the background concentration values. During pre-Covid-19 and Covid-19, the longer CO lifetime explains why the CO plumes extend further in the south eastward direction than the corresponding NO_2 plumes. During the Covid-19 lockdown XNO_2 is reduced by 20%–40% over Riyadh compared with the previous year. In contrast, the XCO background is increased by 3 ppb–5 ppb during Covid-19 compared to pre-Covid-19.

TROPOMI data from April to May 2019 and 2020 are averaged over Denver to analyse the effect of the Covid-19 lockdown (see Fig. S6). During this period, the wind is mostly southerly. TROPOMI retrieved XNO_2 reduces by 40–50% due to the lockdown compared with the same period a year earlier. Like Riyadh, the background is higher by 2–6 ppb than during Covid-19. In the other analyzed cities Chicago, New York, and Sao-Paulo, XNO_2 is reduced by 30–50%, whereas XNO_2 is more strongly reduced Wuhan (65–80%) due to the more strict home stay order during the Covid-19 lockdown (see Figs. S7–S10). In New York, Sao Paulo and Wuhan, XCO close to city centre is reduced by 10–12% during Covid-19 lockdowns. For Chicago, XCO in the urban plume is higher by 5–7%, while the background XCO is higher by 2–5 ppb like in Denver and Riyadh (Fig. S8). The CO background has increased in a few cities due to the influence of regional biomass burning, however, with a

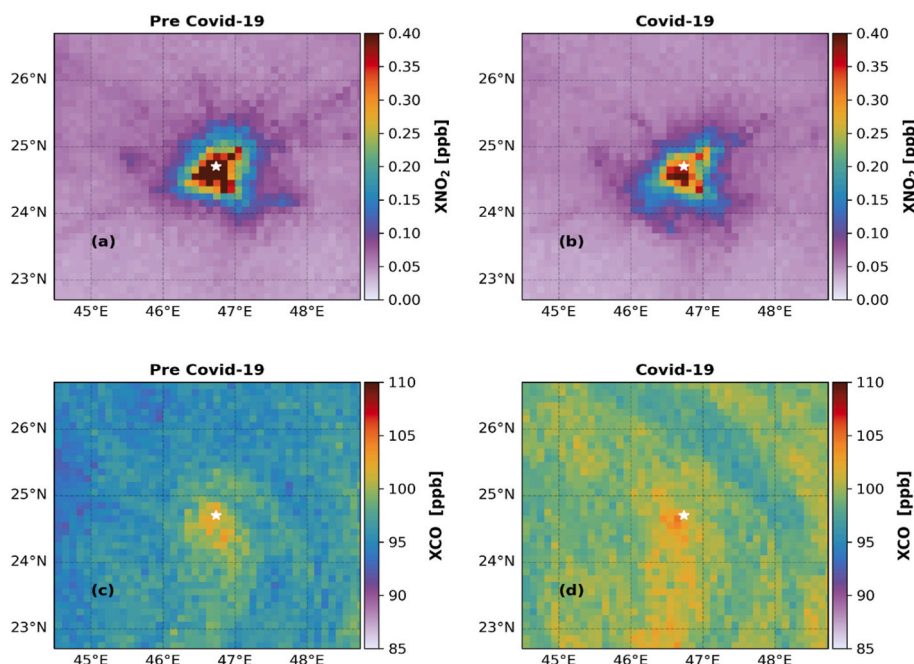


Fig. 4. TROPOMI retrieved XNO_2 (above) and XCO (below) for pre-Covid-19 (1st April to 31st May 2019) and Covid-19 (1st April to 31st May 2020) over Riyadh. The white star represents the centre of Riyadh.

limited influence on the urban photochemistry. A recent study by Miyazaki et al. (2021) showed that the free troposphere has become less oxidative in response to NO_x emission reductions resulting in a decrease in OH at large scales. This could also explain the increase in background CO.

3.2. NO_x lifetime (τ_{NO_x}) in urban plume during pre-covid-19 Vs Covid-19

Using the box rotation method, daily averaged XNO₂, XCO and XNO₂/XCO ratios have been derived for Riyadh for calm (see Fig. 5) and windy days (see Fig. S11) during pre-Covid-19 and Covid-19. During calm days, the peak XNO₂/XCO ratio is close to the centre of the city. The XNO₂/XCO ratio decays most rapidly at intermediate downwind distance (30 km < x < 60 km). At further downwind distance XNO₂/XCO decays again more slowly, due to the combined effect of spatial variation of emission sources and diverse NO_x removal pattern within the downwind urban plume (see Fig. 5c). On calm days during Covid-19, the XNO₂/XCO ratio close to the city centre is reduced by 20–30%, explained mostly by XNO₂. On calm days over Riyadh during Covid-19, τ_{NO_x} is 3.9 h, which is shorter by $22 \pm 6.6\%$ than during pre-Covid-19 (see Fig. 5 d). τ_{NO_x} has been derived for the six megacities by fitting an exponential function to TROPOMI derived XNO₂/XCO ratios in the downwind urban plume during calm and windy days (see Fig. 6 and Fig. S11 to Fig. S17). XNO₂/XCO for data during windy days were available days could only be derived for Riyadh, New York, and Chicago because of insufficient windy data were available for the other cities. The uncertainty in τ_{NO_x} for the six cities has been quantified by adding the contributions of the fitted parameters (A, τ_{NO_x} and C), wind speed, width, length of the box and the background in quadrature (see Tables S2 and S3 and Text S2). The total uncertainty for TROPOMI derived τ_{NO_x} ranges from 10 to 20% across the six megacities.

Over Denver, τ_{NO_x} for calm days is shorter by $28 \pm 9\%$ during Covid-19 than pre-Covid-19, explained mostly by reductions in XNO₂ (Table S4). In other US cities, i.e. Chicago, we also observe shorter τ_{NO_x} during Covid-19 lockdowns. Over Wuhan, Covid-19 τ_{NO_x} derived by fitting XNO₂/XCO ratio is 4.7 ± 0.7 h which differs <15% from Zhang et al. (2023). This difference is within the uncertainty range and

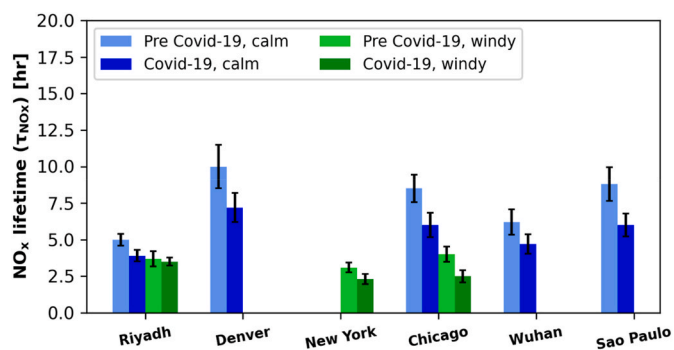


Fig. 6. Comparison of NO_x lifetimes derived using the exponential fitting method applied to TROPOMI derived XNO₂/XCO ratios in downwind urban plumes during pre-Covid-19 and Covid-19 for calm condition (shades of blue) and windy conditions (shades of green). Light shades are for pre-Covid-19 and dark shades for Covid-19. Error bars represent the uncertainty in τ_{NO_x} derived by adding the contributions from wind speed, fitted parameters, width, length of the box and background correction in quadrature.

confirms that the exponential curve fitting method yields realistic estimates of τ_{NO_x} in the urban plume. The more strict home order policy over Wuhan results in stronger reductions in NO₂ (65–80%) but τ_{NO_x} is only reduced by $24.2 \pm 5\%$ during Covid-19. The largest difference in τ_{NO_x} during pre-Covid-19 and Covid-19 is observed during calm days over Sao-Paulo ($31.8 \pm 9\%$) whereas the change in τ_{NO_x} is relatively small over Riyadh compared to other cities (see Fig. 6, S17 and Table S4). Overall, reductions in τ_{NO_x} are found for all the six megacities during Covid-19 lockdowns during calm days (see Fig. 6 and Table S4). On windy days τ_{NO_x} is reduced also during Covid-19 in New York and Chicago, consistent with the calm days. On windy days over Riyadh, however, τ_{NO_x} is almost similar during Covid-19 and pre-Covid-19 (see Fig. S11d).

The τ_{NO_x} for calm and windy days is compared to determine the impact of wind on the NO_x removal in urban plumes. On windy days over Riyadh, the peak in the XNO₂/XCO ratio is shifted 30 km away from

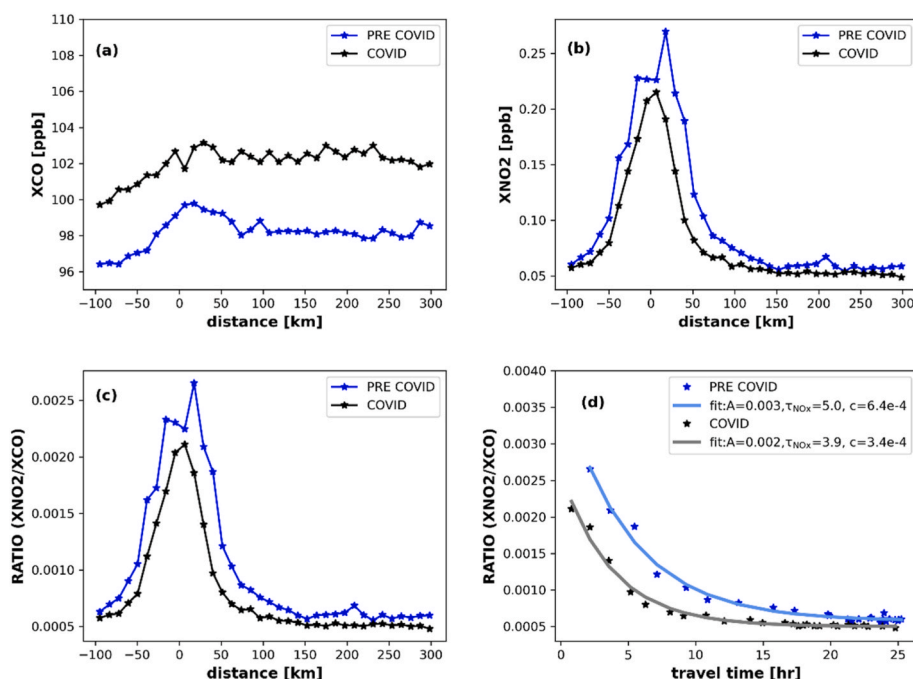


Fig. 5. Pre-Covid-19 (1st April to 31st May 2019) vs Covid-19 (1st April to 31st May 2020) TROPOMI averaged across the wind direction for each small boxes for calm conditions a) XCO, b) XNO₂, c) XNO₂/XCO and d) the exponential fitting of the XNO₂/XCO from the peak point to the downwind distance over Riyadh. The travel time is derived by taking the ratio of travel distance by the average windspeed for each of the small boxes.

the centre of city and is lower by a factor 1.6 compared to calm days, which can be explained by the difference in atmospheric advection and mixing (see Fig. 5 and Fig. S11). As expected, on windy days the NO_2 and CO plume extend >200 km in the downwind direction (see Fig. S11). This demonstrates how TROPOMI can detect the impact of atmospheric transport on the shape of NO_2 and CO plumes. Over Riyadh, τ_{NO_x} is shorter on windy days than on calm days pointing to differences in the NO_x removal, which is expected to be slower during strong pollution episodes. On windy days, the plume is dispersed more quickly leading to lower NO_x concentrations. Lower, but still enhanced NO_x leads to more O_3 and, in turn, higher OH in the plume of a strong emission source (Vinken et al., 2011). On calm days, pollution accumulates increasing τ_{NO_x} , as discussed in the supplement of Lorente et al. (2019). The differences in τ_{NO_x} between calm and windy days are higher during pre-Covid-19 (26.0%) than during Covid-19 (12.5%). Over Chicago, shorter τ_{NO_x} is found during the windy days consistent with Riyadh. The difference in τ_{NO_x} between calm and windy conditions is a factor 2 to 2.5 larger for Chicago than Riyadh. The ERA5 boundary layer over Chicago is shallower during calm days (1106 m) compared to windy days (1737.2 m) by a factor 1.6. The reduced vertical and horizontal mixing during calm days, suppressing OH in reaction with NO_2 can explain the longer τ_{NO_x} (Lorente et al., 2019). This demonstrates that TROPOMI derived XNO_2/XCO can be used to study the functional dependence of the NO_x lifetime on wind speed as mentioned by Valin et al. (2013).

3.3. Comparison between TROPOMI derived τ_{NO_x} and CAMS derived OH

TROPOMI derived τ_{NO_x} during calm and windy conditions has been compared to CAMS derived OH. NO_x and CO emission reductions during the Covid-19 lockdown are not accounted for in CAMS, except over Europe. Therefore, with limit the comparison between TROPOMI derived τ_{NO_x} and CAMS OH to the pre-Covid-19 period. During calm days, boundary layer averaged OH in CAMS is highest over Riyadh (465 ppq) and lowest over Sao Paulo (290 ppq) among the six selected megacities (see Fig. S4). TROPOMI-derived τ_{NO_x} is shortest for Riyadh (5.0 h), consistent with CAMS. Comparing Sao Paulo to Riyadh, CAMS OH is lower by 43% in excellent agreement with TROPOMI derived τ_{NO_x}

being longer by 38%. A similar agreement is found between CAMS OH and TROPOMI-derived τ_{NO_x} over Chicago. Over Denver, however, CAMS OH is 398.5 ppq, which is smaller by 15% compared to Riyadh, whereas TROPOMI shows the longest τ_{NO_x} (10 h) among the selected six megacities. Over Wuhan we also find a disagreement between CAMS OH and TROPOMI-derived τ_{NO_x} , where TROPOMI shows a shorter τ_{NO_x} during windy days than during calm days in contrast to CAMS OH (see Fig. S4). This might be explained by the resolution of the CAMS model being too coarse ($40 \text{ km} \times 40 \text{ km}$) to resolve the urban accumulation of air pollutants.

3.4. Validation using ground-based measurements

To better understand the impact of Covid-19 lockdowns on the photochemical regime in urban plumes and τ_{NO_x} , ground-based measurements from Denver have been analyzed (https://aqsw.ea.gov/aqsw/b/airdata/download_files.html, accessed on 10th Jan 2022) for the period of April to May in 2019 and 2020. Denver has nine ground-based measurement stations for ozone (O_3) and six stations for NO_2 and NO measurements. For CO, data from four ground-based measurement stations are available (see Fig. S18 for a map of measurement stations).

The hourly data are spatially averaged to obtain average diurnal cycles of O_3 , NO_2 , NO, and CO during Covid-19 and pre-Covid-19 (see Fig. 7). In Denver, NO and NO_2 concentrations were reduced by 38.7% and 25.7%, respectively, during the Covid-19 lockdown. This means that the satellite measurements show a larger NO_2 reduction (40–55%) compared to the ground-based measurements. During the Covid-19 lockdown, CO mixing ratios decreased by 17.2%. The O_3 concentration increased by 4 ppb at the time of TROPOMI overpass compared to pre-Covid-19. To analyse the impact of the Covid-19 lockdown in Denver in further detail, we divide the lockdown period into two parts: a) The beginning of the Covid-19 lockdown in the month of April and b) at the midst of Covid-19 lockdown in the month of May. In the beginning of the Covid-19 lockdown, mid-day NO and NO_2 decreased by $\sim 28.2\%$ and O_3 increased by 2.4 ppb (see Fig. S19). However in May, NO shows a larger reduction (54.8%) compared to April at the time of TROPOMI overpass (see Fig. S20). NO_x and volatile organic compounds (VOCs) are the main precursors of photochemical O_3 production in the PBL.

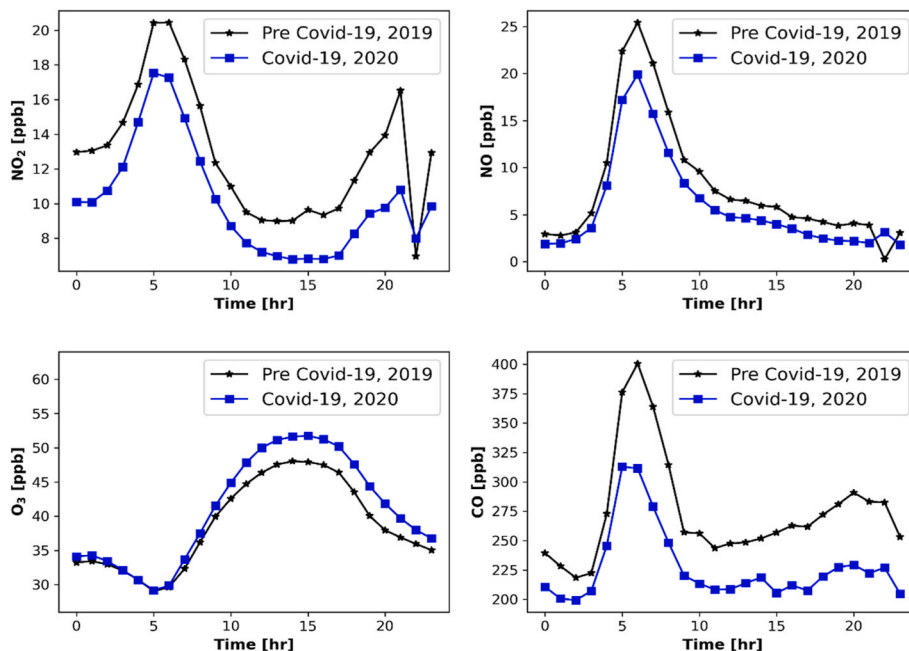


Fig. 7. Averaged diurnal cycles for the period of April to May for Covid-19 and pre-Covid-19 of a) NO_2 , b) NO, c) O_3 and d) CO from ground-based measurements in Denver.

Reductions in NO lead to less O₃ degradation (through $\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2$) under NO_x saturated conditions, causing O₃ to increase by 4.7 ppb at mid-day (see Fig. S19). The increase in O₃ concentration during the conditions of reduced NO_x concentrations is also known as the “week-end effect”. Under high NO_x conditions, less NO₂ leads to less consumption of OH in the NO₂ + OH reaction, increasing OH. To quantify the expected change in OH concentration due to the decrease in NO_x and CO emission, the CLASS box model is used (described in section 3.5).

Yearly variations in emission and meteorological conditions influence O₃ in the urban plume. To account for this variability, five-year averaged ground-based measurement data (2015–2019) are used to derive a reference climatology for comparison to 2020 (Fig. S21). The comparison of five year averaged data (2015–2019) with 2020 shows that NO, NO₂, and CO decrease by 71.4%, 52.1%, and 30% respectively whereas O₃ increases by 1.5 ppb. The year 2017 had extremely high ozone concentrations in April compared to 2020 and resulted in a small decrease in O₃ concentration. However, in May 2020 O₃ concentrations are higher by 3.5 ppb in the afternoon compared to the five-year averaged data (Fig. S22).

3.5. CLASS vs ground-based measurements

To test if the CLASS model can reproduce the observed changes in Denver during 2020, modelled diurnal cycles of NO, NO₂, O₃, and CO are compared to measurements from three ground based stations i.e. Denver 1, Denver 2, and Adams that measure NO, NO₂, O₃ and CO simultaneously, averaged over April to May in 2019 and 2020. The CLASS model is run to simulate the spatially average NO, NO₂, O₃, and CO concentrations over Denver during 8h (8 a.m.–16 p.m.) for pre-Covid-19 and Covid-19 (see Tables S5 and S6 for details of the CLASS setup that has been used).

CLASS can reproduce the morning rush hour peak in NO, NO₂, and CO, and subsequent reductions during the afternoon (see Fig. 8) with a mean absolute error of −0.73 ppb, 0.44 ppb and 0.04 ppb respectively. CLASS captures the increase of O₃ concentration, reaching maximum in the afternoon due to photochemical production (see Fig. 8). To simulate the average NO, NO₂, and O₃ concentrations during Covid-19, the NO_x

and CO emissions are reduced by 70% and 40%, respectively, compared with pre-Covid-19 (see Table S5). CLASS is able to reproduce the observed reduction in NO, NO₂, and CO concentrations and increase in O₃ (see Fig. 8) during the Covid-19 lockdown. As expected from the diurnal cycle in solar insolation, the CLASS simulated diurnal cycle of OH reaches a maximum in the early afternoon (see Fig. 9). Reductions in NO_x and CO emissions during Covid-19 increase OH by 51.3% during the mid-day (12 LST to 14 LST). This OH change in CLASS compares well to TROPOMI ($28 \pm 9\%$). A sensitivity test in which emissions of isoprene are increased by a factor of 1.5 along with NO_x and CO emission reductions also shows an increase in midday OH by 62.4% (see Fig. S23).

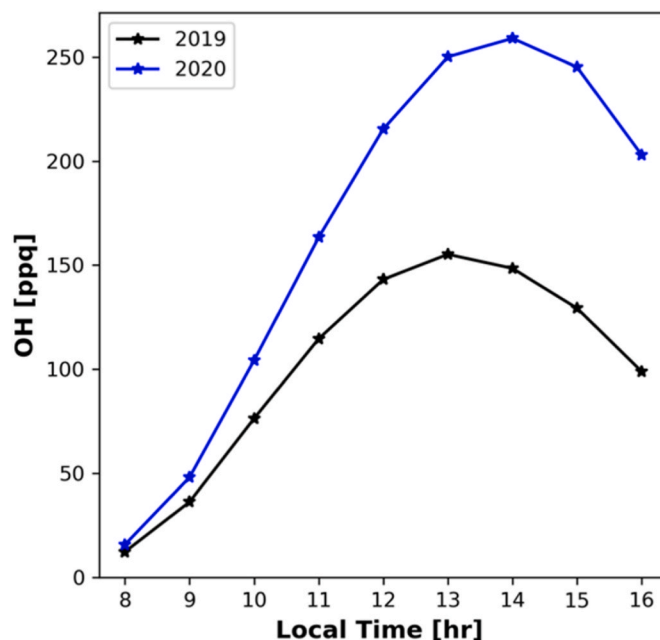


Fig. 9. CLASS simulated OH for 2019 and 2020 over Denver.

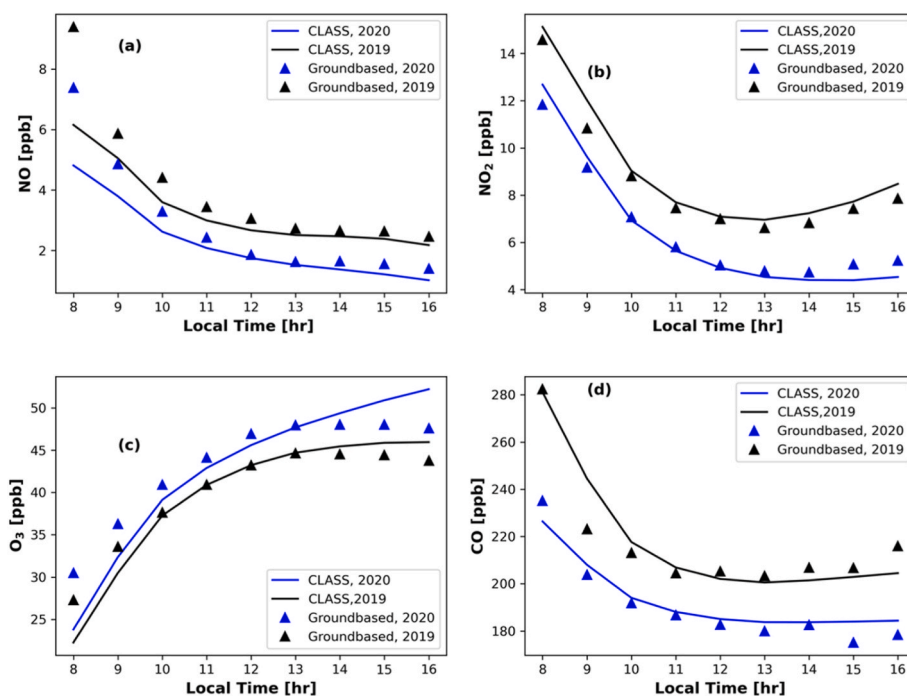


Fig. 8. The mean mixing ratio of a) NO, b) NO₂, c) O₃ and d) CO in Denver simulated by the CLASS model (solid line) and observed by the surface network (triangles) using measurement stations Denver 1, Denver 2, and Adams averaged from April to May in 2019 and 2020.

Another sensitivity test is performed to isolate the impact of the NO_x emission reduction on OH production. This test indicates that reduction in NO_x emission contributes to 95.3% of the OH increase.

4. Discussion

From TROPOMI retrieved NO₂/CO ratios we infer a substantial reduction in τ_{NO_x} over and directly downwind of large cities during Covid-19 lockdowns. For Denver, the satellite observations, ground-based observation, and the CLASS model agree on a τ_{NO_x} decrease by $28 \pm 9\%$. Sensitivity tests in the CLASS model indicate that the decrease in τ_{NO_x} is driven by reduced NO_x emission, which eventually increase OH concentration in urban plume because less OH is consumed by the NO₂ + OH reaction, and more OH is recycled. OH₂ is the main factor determining the lifetime of NO_x during noon, this explains the reduction in τ_{NO_x} . The same mechanism can explain the decreases in τ_{NO_x} inferred for other cities using TROPOMI data. A recent global study by Miyazaki et al. (2021) on urban photochemistry demonstrated that the free troposphere has become less oxidative in response to reductions in NO_x emissions resulting in an increase of the CH₄ lifetime. In the NO_x limited free troposphere, a NO_x decrease is indeed expected to reduce OH, contrasting with the NO_x rich urban conditions of our study.

Meteorological conditions such as temperature, radiation, and wind speed also influence the urban photochemistry, and therefore could also explain spatial and temporal variations in τ_{NO_x} (Lang et al., 2015; Romer et al., 2018). Under clear sky conditions, τ_{NO_x} is systematically lower due to the higher photolysis rates. Because of this, the selection of satellite data for clear sky conditions introduces a sampling bias. However, this bias is present both in the pre-Covid-19 and Covid-19 time windows and the sampling bias is therefore unlikely to explain differences between the two periods, unless cloud cover was significantly different. Differences in meteorology between pre-Covid-19 and Covid-19 can certainly contribute to the differences in τ_{NO_x} . However, meteorology is unlikely to have influenced τ_{NO_x} in the same way across all the six megacities, increasing the likelihood that the τ_{NO_x} changes are primarily caused by changes in NO_x emissions. In city centres with strong emissions from traffic, the photochemical regime is known to be NO_x saturated. Under these conditions, the oxidative capacity is limited by the removal of OH in reaction with NO₂, and decreasing NO_x emission increase OH. Improvements in visibility due to reduced emissions and secondary formation of aerosols could also have contributed to τ_{NO_x} decreases during Covid-19. However, our CLASS computations, which do not simulate changes in aerosol loading, indicate that reduced emissions of reactive trace gases are sufficient to explain the observed τ_{NO_x} changes.

Further downwind of the city, the chemical conditions become favourable for OH formation and recycling, reducing τ_{NO_x} (Romer-Prent et al., 2019; Sobanski et al., 2017; Valin et al., 2013). The exponential fit to the TROPOMI derived NO₂/CO ratio in the downwind urban plume provides an average estimate of τ_{NO_x} and cannot distinguish between τ_{NO_x} in the centre of city and its outskirts, despite our attempts to resolve this difference (not shown). However, future satellite instruments observing at higher temporal and spatial resolutions could provide more independent points over city, so the gradient in NO₂/CO ratio can be captured better. This can provide the information needed to study chemical processing in city plumes.

We do not have emission inventory estimates for 2020 but assume that traffic emissions reduced the most. Therefore, a sensitivity test has been performed for Denver in which traffic NO_x and CO emissions are reduced by 45%, while keeping the other emissions sectors unchanged. According to EDGAR, this traffic emission reduction would increase the NO₂/CO ratio of the emissions by 28.5%. However, TROPOMI shows stronger reductions in NO_x compared to CO resulting in a decrease in the NO₂/CO ratio close to centre of city during Covid-19 lockdown in all the cities we analyzed. Increases in OH due to reductions in NO_x emissions under VOC limited photochemical conditions in city centres could

partially explain the difference between TROPOMI and EDGAR derived NO₂/CO ratio changes. This provides another line of evidence that NO_x lifetimes reduced during Covid-19 lockdowns. Besides traffic emissions, some industrial emission sectors likely also reduced during lockdowns, which could explain another part of the mismatch between TROPOMI and inventory data.

We verified the consistency of the observed NO₂ decrease and estimated OH increase with the EDGAR emission inventory. During the lockdown in Wuhan, EDGAR shows negligible emissions from aviation and road transport, and 80% reduction in the manufacturing industries. The combined impact is a reduction of the total NO_x emission by 50%. TROPOMI shows stronger reduction in NO₂ of 65–80%, which is consistent with the simultaneous increase in OH that we derive.

This study shows that the reduction in emissions results in an increase in the urban atmospheric oxidative capacity, primarily driven by the presence of hydroxyl radicals (OH). This increased oxidative capacity enhances the degradation of reactive species through oxidation processes, ultimately contributing to improved air quality. However, it is important to note that the increased oxidative capacity can also promote additional photochemical reactions, leading to the formation of ozone (O₃) and secondary aerosols. Elevated levels of O₃ can have adverse effects on human health and ecosystems. Thus, while emission reductions have positive implications for air quality through enhanced oxidative degradation, careful consideration is necessary to mitigate the potential negative impacts of increased photochemical reactions and the subsequent formation of O₃ and secondary aerosols. This study shows that NO₂/CO ratio from satellite observations can be used to test how well photochemistry models are capable of simulating the dynamics in urban OH.

In this study, strong isolated sources of NO₂ and CO have been selected to estimate the NO_x lifetime. In regions with more complex emission distributions, calm conditions can be selected to derive τ_{NO_x} when the NO₂ and CO plumes are concentrated close to the source. In addition, our method needs NO_x and CO emissions that are large enough to be readily detectable from space. In European cities, CO emission have been reduced in the past to levels that are barely detectable by TROPOMI. In such cases our method cannot be used.

5. Conclusion

In this study, TROPOMI derived XNO₂, XCO, and XNO₂/XCO ratios are used to study changes in the urban photochemistry during Covid-19 lockdowns. An exponential function fitted method is used to estimate the chemical lifetime of NO_x from TROPOMI retrieved NO₂/CO ratios in the downwind urban plume of large megacities. τ_{NO_x} has been estimated for six different megacities (Riyadh, Wuhan, Sao-Paulo, Chicago, Denver, and New York) during Covid-19 and the year before, to analyse the impact of Covid-19 lockdowns on urban photochemistry. To account for influences of wind speed, τ_{NO_x} is determined for calm and windy day separately. For windy days, τ_{NO_x} has been derived for Riyadh, Chicago and New York only, because the data availability was insufficient for the other cities.

The Covid-19 lockdown reduced urban NO₂ concentrations by 30–50% in the cities of Riyadh, Denver, Chicago, Sao-Paulo, and New York. Wuhan adopted a stricter lockdown policy compared to the other megacities leading to a larger reduction in NO₂ concentration (65–80%). Close to centre of city, XCO showed smaller reductions (10–15%) over Wuhan and New York. In contrast, the XCO background concentration increased during Covid-19 in Riyadh, Denver, Chicago, and Sao-Paulo by 5–10%.

Except over Riyadh during windy days, our results show a systematic reduction in τ_{NO_x} during Covid-19 lockdowns. For the calm days, a large difference in τ_{NO_x} is observed over Sao-Paulo ($31.8 \pm 9.0\%$), whereas a relatively small change in τ_{NO_x} is found for Riyadh ($22 \pm 6.6\%$). Similarly, windy days over New York and Chicago also show shorter τ_{NO_x} during the Covid-19 lockdown than pre-Covid-19. Only in the case of

windy days over Riyadh a similar τ_{NO_x} is observed during Covid-19 and pre-covid-19.

The ground-based measurement and the CLASS model have been used to study the impact of the covid lockdown on the urban photochemistry in Denver, confirming the satellite derived τ_{NO_x} reductions. The CLASS model reproduced the observed increase in O_3 concentration by 4.0 ppb in response to the observed reductions in NO (38.7%) and NO_2 (25.7%) concentration during covid. The decreased NO_x emission results in a reduced consumption of OH in the $\text{NO}_2 + \text{OH}$ reaction, eventually increasing OH in the urban plume which is displayed by CLASS and indicating the reason for shorter τ_{NO_x} during Covid-19 lockdown at mid-day. The relative change in τ_{NO_x} estimated by TROPOMI ($28 \pm 9\%$) is consistent with the CLASS simulations, initialized using the ground-based measurements in Denver.

This study demonstrates how satellite measurements of NO_2 and CO can be used to study urban photochemistry in polluted cities around the world, providing a new observation-based method to evaluate the models that are used for monitoring urban air quality. The changes that are detected during Covid-19 lockdowns provide an observational based quantification of air quality benefits that are to be expected from the energy transition, which can be monitored from space using the method presented in this study.

CRedit authorship contribution statement

Srijana Lama: Formal analysis, performed the data analysis and data interpretation and wrote the paper, discussed the results. **Sander Houweling:** Supervision, supervised the study, discussed the results. **K. Folkert Boersma:** discussed the results. **Ilse Aben:** discussed the results. **Hugo A.C. Denier van der Gon:** discussed the results, All co-authors commented on the paper and improved it. **Maarten C. Krol:** discussed the results.

Declaration of competing interest

The authors declare that they have no conflict of interest.

Data availability

I have shared the data link in the manuscript.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2023.120042>.

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