Tropospheric NO$_2$ and O$_3$ response to COVID-19
lockdown restrictions at the national and urban scales
in Germany

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Key Points:

• During the COVID-19 lockdown period, NO$_2$ concentrations decreased and O$_3$ concentrations increased in eight German cities

• The degree of NO$_X$ saturation of ozone production is weakening from winter to summer

• Meteorological variability adjusted by GEOS-Chem model simulations driven by the same emissions for 2020 and 2019

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Abstract

This study estimates the influence of anthropogenic emission reductions on nitrogen dioxide (NO$_2$) and ozone (O$_3$) concentration changes in Germany during the COVID-19 pandemic period using in-situ surface and Sentinel-5p (TROPOMI) satellite column measurements and GEOS-Chem model simulations. We show that reductions in anthropogenic emissions in eight German metropolitan areas reduced mean in-situ (\& column) NO$_2$ concentrations by 23 \% (\& 16 \%) between March 21 and June 30, 2020 after accounting for meteorology, whereas the corresponding mean in-situ O$_3$ concentration increased by 4 \% between March 21 and May 31, 2020, and decreased by 3 \% in June 2020, compared to 2019. In the winter and spring, the degree of NO$_X$ saturation of ozone production is stronger than in the summer. This implies that future reductions in NO$_X$ emissions in these metropolitan areas are likely to increase ozone pollution during winter and spring if appropriate mitigation measures are not implemented. TROPOMI NO$_2$ concentrations decreased nationwide during the stricter lockdown period after accounting for meteorology with the exception of North-West Germany which can be attributed to enhanced NO$_X$ emissions from agricultural soils.

Plain Language Summary

Pollutant concentrations in the atmosphere are influenced not only by changes in emissions, but also by meteorology and atmospheric chemical reactions. Because of this, estimating the direct influence of anthropogenic emission reductions on nitrogen dioxide (NO$_2$) and ozone (O$_3$) concentrations during the initial COVID-19 pandemic period is complex. In our study, we used GEOS-Chem model simulations to account for meteorology impacts. For Germany, compared to 2019 we see a decrease in NO$_2$ concentrations during the 2020 lockdown period, an increase in O$_3$ concentrations during the 2020 spring lockdown, and a decrease in O$_3$ concentrations during the 2020 early summer lockdown. The increased O$_3$ concentration in response to the decreased NO$_2$ concentration implies that future reductions in NO$_X$ emissions are likely to increase ozone pollution in German metropolitan areas during winter and spring. Furthermore, there was a nationwide decrease in NO$_2$ concentrations except for North-West Germany during the 2020 stricter lockdown period. We hypothesise that North-West Germany is a hotspot of soil NO$_X$ emissions in elevated-temperature environments due to intensive agricultural practices.

1 Introduction

The outbreak of the novel COVID-19 virus in late 2019 prompted governments to take various measures to prevent the COVID-19 virus from spreading through society. These actions include physical distancing, a ban on large group gatherings, home office work, and international and domestic travel restrictions (DW COVID-19, 2020). These measures resulted in a significant reduction in emissions following economic activity and overall mobility (Evangelou et al., 2021; Gensheimer et al., 2021; Guevara et al., 2021; Le Quéré et al., 2020; Turner et al., 2020; Z. Liu, Ciais, et al., 2020; Z. Liu, Deng, et al., 2020). There has been a lot of interest in studying this time window and its impacts on the Earth system. Numerous studies (Bauwens et al., 2020; Berman & Ebisu, 2020; Chauhan & Singh, 2020; Collivignarelli et al., 2020; Dietrich et al., 2021; He et al., 2020; Keller et al., 2021; R. Zhang et al., 2020) have reported a reduction in major air pollutant concentrations during the COVID-19 lockdown period, including nitrogen dioxide (NO$_2$), carbon monoxide (CO), sulphur dioxide (SO$_2$) and particulate matter (PM$_{10}$ and PM$_{2.5}$), which are primarily emitted by fossil fuel consumption. During the COVID-19 lockdown period, air quality improved in most countries, particularly in urban areas (Bedi et al., 2020; Fu et al., 2020). Previous studies, such as Bauwens et al. (2020); Deroubais et al. (2021), compared lockdown period concentration with long-term mean to estimate lock-
down effects by eliminating the average climatological seasonal cycle. However, a direct comparison of lockdown period pollutant concentrations with pre-lockdown period pollutant concentrations includes both meteorological and COVID-19 emission reduction influences.

Meteorological effects must be considered to determine the actual impact of anthropogenic emission reductions on changes in pollutant concentrations during the COVID-19 lockdown period (Barré et al., 2020; Deroubaix et al., 2021; Gaubert et al., 2021; Goldberg et al., 2020; Petetin et al., 2020; Sharma et al., 2020; Y. Liu et al., 2020), particularly with regard to chemical processes (Kroll et al., 2020). An analysis of pollutant concentration changes over the European networks of surface air quality measurement stations was performed to isolate the lockdown impacts based on a data-driven meteorological adjustment (Ordóñez et al., 2020; Venter et al., 2020). Previous works (Gaubert et al., 2021; Menut et al., 2021; Mertens et al., 2021; Potts et al., 2021; Weber et al., 2020) have used different modelling approaches to investigate the impact of lockdown measures on air quality over Europe. The 2020 emission reduction scenarios were set up using available activity data from various sources (Dounbia et al., 2021; Forster et al., 2020; Guevara et al., 2021). As part of its modeling work, Gaubert et al. (2021) compared the 2020 lockdown period with climatological mean in order to separate the anomalies caused by the weather conditions in 2020, and they have called for more meteorology adjusted studies to avoid the flawed results.

We focus on nitrogen dioxide (NO$_2$) and ozone (O$_3$) concentration changes due to 2020 COVID-19 lockdown restrictions, from March 21 to June 30. We consider NO$_2$ and O$_3$ together from the perspective of atmospheric chemistry, because NO$_2$ and O$_3$ concentrations are functions of each other (Bozem et al., 2017). Nitrogen oxide (NO$_X$ = NO+NO$_2$) emissions have a pronounced seasonal cycle, with higher emissions in the winter than in the summer (Beirle et al., 2019; Kuenen et al., 2014). Half of the NO$_X$ in the troposphere is from fossil fuel consumption in urban areas (e.g. figure S1). Tropospheric NO$_2$ concentrations follow a similar annual cycle, with higher values in the winter than in the summer. This is due to the fact that in addition to the higher emissions mentioned above also the lifetime of NO$_2$ is longer in the winter (≈21 hours) than in the summer (≈6 hours) (Shah et al., 2020). Peak NO$_2$ concentrations in the winter are also influenced by atmospheric inversion conditions. NO$_2$ influences climate by acting as a precursor to the formation of tropospheric O$_3$ (Crutzen, 1988; Jacob, 1999), and both NO$_2$ and O$_3$ have an impact on human health. Tropospheric ozone production is complex and depends strongly and non-linearly on nitrogen oxides (NO$_X$) and volatile organic compound (VOC) concentrations, despite the fact that photolysis of NO$_2$ is the only chemical source of tropospheric ozone (Council et al., 1992; Kleinman, 2005; Lin et al., 1988). Ozone decreases as NO$_X$ decreases in regions with low NO$_X$ and high VOC concentrations, i.e., NO$_X$ limited regimes; however, in high NO$_X$ regions, i.e., VOC limited regimes (or NO$_X$ saturated regimes), a decrease in NO$_X$ results in an increase in O$_3$ concentration (Kleinman et al., 1997; Sillman, 1999; Sillman et al., 1990) (figure S2).

This study uses the TROPOspheric Monitoring Instrument (TROPOMI) on the Sentinel-5 Precursor (Sentinel-5P) satellite and governmental in-situ NO$_2$ measurements as a proxy for changes in NO$_2$, and governmental in-situ O$_3$ measurements as a proxy for changes in O$_3$ concentrations in Germany. To account for the impact of meteorology, we use the same anthropogenic emissions in 2020 and 2019 with 2019 open fire emissions and long-term (1995-2013) monthly lightning NO$_X$ emission climatology for the GEOS-Chem model. We are therefore able to present separate quantitative results for changes in NO$_2$ and O$_3$ concentrations caused by meteorological changes and by reductions in anthropogenic emissions resulting from COVID-19 lockdown measures. To the best of our knowledge, no such study using GC modeling to account for meteorological impacts has been conducted for Germany.
2 Study Regions, Data Sets, Model and Method

Our study region covers a bounding box over the national area of Germany (5-15.5°E, 47-55.5°N), with a particular focus on eight urban areas spread across the country: Munich, Berlin, Cologne, Dresden, Frankfurt, Hamburg, Hanover, and Stuttgart (Supplementary figure S3). This study mainly focused on the urban scale to examine the impact of reduced mobility on NO$_2$ and O$_3$ concentrations during the 2020 COVID-19 pandemic period. We also extended our study nationwide to investigate other significant NO$_X$ sources in rural locations.

We used tropospheric NO$_2$ column data from the TROPOspheric Monitoring Instrument (TROPOMI) aboard the Sentinel-5 Precursor satellite (Copernicus, 2020). The satellite is in a sun synchronous orbit with an equatorial crossing time of 13:30 (local solar time). TROPOMI NO$_2$ data has a spatial resolution of 7*3.5 km (increased to 5.5*3.5 km after 6 August, 2019) and it covers the globe daily due to its wide swath (Van Geffen et al., 2020). TROPOMI NO$_2$ precision (error estimate originating from the spectral fit and other retrieval aspects) for each pixel is within the range of $3.6 \times 10^{14}$ to $3.7 \times 10^{16}$ molec. cm$^{-2}$ (about 21% to 75% of tropospheric NO$_2$ column). The TROPOMI NO$_2$ measurements for winter are highly uncertain (Figure S4). The main source of uncertainty is the calculation of the air mass factor, which is estimated to be on the order of ± 30% (Lorente et al., 2017). Since our study is mainly focusing on the relative difference in NO$_2$ between 2020 and 2019, the systematic errors associated with TROPOMI retrievals (e.g., spectroscopic errors and instrument bias) should cancel out, while random error component is persistent. However, when we apply temporal and spatial averaging, random errors are reduced. We followed S5P NO2 Readme (2020) for the quality filter criteria, which removes cloud-covered scenes in order to avoid high error propagation through retrievals. We averaged the TROPOMI values within a radius of 0.5 degree from the urban center to create time series (& daily observations) at the urban scale. For comparisons between 2020 and 2019 at the national scale, TROPOMI tropospheric NO$_2$ column densities were gridded in 0.25*0.25-degree bins.

We investigate agricultural activities in Germany using ammonia (NH$_3$) data (Kuttippurath et al., 2020). The “Standard monthly IASI/Metop-B ammonia (NH$_3$) data set” was downloaded from IASI NH$_3$ (2020). This data set contains monthly averaged NH$_3$ measurements (total column), from the Infrared Atmospheric Sounding Interferometer (IASI), onboard the Metop satellites, at 1*1 degree resolution. We also used the “Near-real time daily IASI/Metop-B ammonia (NH3) total column dataset (ANNI-NH3-v3)” product to investigate the inter-annual short-term (less than a month) variability in NH$_3$ over Germany (IASI NH$_3$, 2020).

In-situ surface NO$_2$ and O$_3$ concentrations were obtained as hourly averaged measurements from the UBA’s (German Environment Agency) database (Umweltbundesamt, 2020). We collected data from 38 stations in eight German cities, including both urban and rural measurement sites, for 2020 and 2019. In this study, we averaged all 24-hour measurements from stations located within each city.

The ERA5 dataset (Copernicus Climate Change Service (C3S), 2017) is used as a reference data set to discuss meteorological conditions over study areas. We used the “ERA5 hourly data on pressure levels” product for wind speed and direction and temperature. Further, we used the “ERA5 hourly data on single levels” product for boundary layer height. We averaged these values within a radius of 0.5 degree from the urban center to create a time-series (& daily observations) at the urban scale. The sunshine duration (hours per day) data was obtained from Deutscher Wetterdienst (DWD, 2020).

The GEOS-Chem (GC) chemical transport model (GEOS-Chem, 2020) is used to estimate the concentration differences in NO$_2$ and O$_3$ between 2020 and 2019 caused by meteorological changes. The GEOS-Chem model is driven by MERRA-2 assimilated me-
teorological data (MERRA-2, 2020). We conduct nested simulations over Germany (4-17°E, 45-57°N) at a horizontal resolution of 0.5°×0.625° with dynamic boundary conditions generated from a global simulation by 4°×5° resolution. GEOS-Chem assumes the same anthropogenic emissions in 2020 and 2019. We used anthropogenic emissions in 2014 from the Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018) and 2019 open fire emissions from GFED4 (Werf et al., 2017) for both 2019 and 2020 simulations. The spatial and monthly climatology of lightning NO\textsubscript{X} emissions is constrained by LIS/OTD satellite observations averaged over 1995-2013. We used an improved parameterization approach implemented in the GEOS-Chem model to calculate soil NO\textsubscript{X} emissions (Hudman et al., 2012). In all comparisons of the GC model to TROPOMI, GC NO\textsubscript{2} vertical profile simulations (at 47 vertical layer) are converted to NO\textsubscript{2} column densities for TROPOMI footprints by interpolating into TROPOMI measurements pressure levels and applying TROPOMI’s averaging kernels. Similar to above, GC column densities were gridded in 0.25°×0.25-degree bins at the national scale.

Our methodology to obtain NO\textsubscript{2} and O\textsubscript{3} concentration changes between 2020 and 2019 (2020-2019) for which meteorological impacts have been accounted for is as follows. Previous studies (Fiore et al., 2003; R. F. Silvern et al., 2019; Tai et al., 2012) have shown that GC can reproduce the temporal variability of NO\textsubscript{2}, O\textsubscript{3} and particulate matter, implying that GC accounts for meteorological impacts. We conduct GC simulations for 2020 and 2019 with identical emissions but with the respective meteorology from MERRA-2 reanalysis. Since we use the same anthropogenic emission in 2020 and 2019, the GC differences in NO\textsubscript{2} and O\textsubscript{3} between 2020 and 2019 are solely due to meteorological influences, e.g., differences in wind speed, boundary layer height, photo-chemistry etc.:

\[ \Delta \text{NO}_2(\text{GC}) = \text{NO}_2(\text{GC,2020}) - \text{NO}_2(\text{GC,2019}) \]

\[ \Delta \text{O}_3(\text{GC}) = \text{O}_3(\text{GC,2020}) - \text{O}_3(\text{GC,2019}) \]

The difference between the 2020 and 2019 NO\textsubscript{2} and O\textsubscript{3} observations for specific time periods include influence from both meteorological and emissions changes:

\[ \Delta \text{NO}_2(\text{obs}) = \text{NO}_2(\text{obs,2020}) - \text{NO}_2(\text{obs,2019}) \]

\[ \Delta \text{O}_3(\text{obs}) = \text{O}_3(\text{obs,2020}) - \text{O}_3(\text{obs,2019}) \]

In order to account for the differences resulting from meteorology and isolate the impact resulting from emission changes we subtract the difference in the simulations from the difference in the observations as follow (Qu et al., 2021),

\[ \Delta \text{NO}_2(\text{acc}) = \Delta \text{NO}_2(\text{obs}) - \Delta \text{NO}_2(\text{GC}) \]

and similarly for ozone:

\[ \Delta \text{O}_3(\text{acc}) = \Delta \text{O}_3(\text{obs}) - \Delta \text{O}_3(\text{GC}) \]

Where, “acc” refers to meteorology accounted for, “obs” refers to in-situ or TROPOMI measured concentrations, and “GC” refers to GEOS-Chem model simulated concentrations. This approach results in values that have accounted for meteorological influence to estimate the concentration changes resulting only from COVID-19 emission reductions.

3 Tropospheric \text{NO}_2 and \text{O}_3: impact of meteorological conditions

Like previous studies (Çelik & İbrahim, 2007; Deroubaix et al., 2021; Ordóñez et al., 2020; Voiculescu et al., 2020), we investigated correlations between satellite and in-situ NO\textsubscript{2} and O\textsubscript{3} concentrations and local meteorological parameters to find the dependency of NO\textsubscript{2} and O\textsubscript{3} concentrations on meteorology. The correlation matrix is shown in Figure 1 for the Munich metropolitan area. We find similar correlation behaviour between variables for 2019 (no lockdown) and 2020 (lockdown). Generally, satellite and in-situ NO\textsubscript{2} concentrations have a negative correlation with wind speed, temperature and...
boundary layer height, e.g., as pollutants disperse more at high wind speeds than at low
wind speeds. As temperature and sunlight increases, the rate of NO$_2$ photochemical loss
accelerates, and the planetary boundary layer expands resulting in higher dilution. O$_3$
concentrations have a generally negative correlation with NO$_2$ concentrations and posi-
tive correlation with sunshine duration and temperature. This results from the fact that
NO$_2$ and high solar radiation play an important role in regulating O$_3$. Temperature has
been shown to have a significant influence on ozone production over Europe under var-ious NO$_X$ conditions (Coates et al., 2016; Melkonyan & Wagner, 2013). In addition, Curci
et al. (2009) show that increasing temperature causes an increase in biogenic VOC emis-
sions, which can raise the ozone level, especially in the summer. Future climate condi-
tions in Europe (as a result of global warming) will almost certainly have an impact on
ozone pollution (Engardt et al., 2009; Forkel & Knoche, 2006; Meleux et al., 2007; Vau-
tard et al., 2007). Europe may experience more intense and frequent heatwaves and droughts
in the future, which will increase wildfire events and, as a result, background ozone lev-
els will increase (De Sario et al., 2013; Meehl & Tebaldi, 2004). Furthermore, temper-
ature, boundary layer height and solar radiation, which are considered to be the most
related meteorological factors influencing NO$_2$ and O$_3$ concentrations, are interdepen-

4 Changes in NO$_2$ and O$_3$ concentrations in Germany due to COVID-19 lockdown restrictions

In this study we compare January through June of 2020 and 2019. This time pe-
period is divided into five sections: 1) No lockdown restrictions from January 1 to January
31, 2020. 2) No lockdown restrictions with anomalous weather conditions from Febru-
ary 1 to March 20, 2020. 3) Strict lockdown restrictions from March 21 to April 30, 2020
(spring). 4) Loose measures from May 1 to May 31, 2020 (late spring). 5) Loose mea-
sures from June 1 to June 30, 2020 (early summer). The mean TROPOMI and in-situ
NO$_2$ in January of 2020 was slightly higher than in 2019 (Figure 2 (c) and 3(a)). How-
ever, between February 1 and March 20, 2020, prior to the lockdown, mean observed TROPOMI
and in-situ NO$_2$ was already significantly lower than in 2019 at both the national (Fig-
ure 2 (f)) and urban scales (Figure 3(c) and S5). This can be attributed to unusually
high wind speeds caused by storms in February 2020 (DLR COVID-19, 2020). The first
governmental COVID-19 lockdown restrictions went into effect on March 21, 2020. In
the period following the lockdown implementation, lower NO$_2$ values are observed com-
Figure 2. Mean TROPOMI tropospheric NO$_2$ column densities for 2019 (first column) and 2020 (second column). The absolute differences in TROPOMI tropospheric NO$_2$ column densities between 2020 and 2019 (third column).
pared to 2019. In-situ measurements show lower mean O3 concentrations in January and
June 2020, and higher mean O3 concentrations from February 1 through May 31, 2020,
compared to 2019 (Figure 3 and S5).

GEOS-Chem model simulations are used to estimate the difference in NO2 and O3
concentrations between 2020 and 2019 caused by meteorology. Studies (Fiore et al., 2003;
R. F. Silvern et al., 2019; Tai et al., 2012) have demonstrated that GC can reproduce
the observed temporal variability of NO2, O3 and particulate matter, implying that GC
accounts for impacts of meteorology when using precise meteorological data and emis-
sion inventories. In our study, we also compare the GC and observed concentrations from
2019 to verify that the GC can reproduce the temporal variability of observed concen-
tration changes. The 2019 (January to June) period is used to validate the GC model
simulations as unlike 2020 emissions are not affected by changes resulting from COVID
measures. To validate the GC model, we compared GC surface concentrations with in-
situ surface concentrations, and GC column densities with TROPOMI column densities
(Figure S6, for cologne metropolitan area). We find good agreement between GC sur-
face NO2 concentrations and in-situ surface NO2 concentrations for eight metropolitan
areas (R, Pearson correlation coefficient, > 0.65, with high R (0.75) for Cologne). Sim-
ilar results were obtained for GC surface O3 concentrations, (R > 0.65, with a high R
(0.74) for Dresden). GC underestimates NO2 surface concentrations, except for Ham-
burg. The mean bias (GC - in-situ) ranges from +2.9 to -23 %. Except for Hamburg and
Hanover, GC overestimates surface O3 concentrations, with mean bias ranges from +24
to -10.3 %. When comparing 2019 GC and TROPOMI NO2 column densities, relatively
low correlation (R, between 0.24 and 0.55) was found, and the NO2 column densities in
metropolitan areas were underestimated by GC (mean bias ranges from -4 to -28 %). How-
ever, the GC model is capable of modeling the spatial variability of NO2 column den-
sities at the national scale, emphasizing GC’s ability to represent the distribution of emis-
sion source locations (Figure S7). The over/under estimation of NO2 and O3 concen-
trations are caused by the emission inventory (over/under estimation of emission) used
in GC simulation. The low bias in NO2 and high bias in O3 could be consistent with NOX
saturated conditions. Because we use the difference in GC concentrations between 2020
and 2019 (Δ NO2(GC) and Δ O3(GC)), general biases are cancelled out.

Due to the passage of two strong storm systems February 2020 experienced high
winds. We consider the period from February 1 to March 20, 2020 (prior to the imple-
mentation of lockdown restrictions) to determine the extent to which meteorology is re-
sponsible for variations in pollutant concentrations. Before accounting for meteorology,
the difference in mean in-situ NO2 concentration between 2020 and 2019 is -28 % for the
period February 1 and March 20. After accounting for meteorology, the difference is re-
duced to -6 % (consistent with meteorology accounted changes for the period between
January 1 and January 31, 2020 compared to 2019, figure 1 (a,b,c,d)). This emphasizes
the significance of employing our method to account for meteorological impacts. The im-
pacts of meteorology on in-situ and TROPOMI NO2 concentrations are relatively small
(+0.4 % and -0.6 %, respectively) for the period between March 21 and June 30, 2020
(the period after the implementation of lockdown restrictions). After accounting for me-
teorology, the mean in-situ and TROPOMI NO2 values between March 21 and June 30,
2020 were significantly lower (by 23 % and 16 %, respectively) than the same period in
2019 (Figure 3, (f, h, j)). Other studies (Barré et al., 2020; Grange et al., 2020; Solberg
et al., 2021) that used a machine learning and statistical approach to account for me-
teorological impacts also found that the impact of the COVID-19 pandemic on NOX emis-
sions lasted at least until June 2020. After accounting for meteorology, we observed a
slight increase in mean in-situ O3 concentration between March 21 and May 31, 2020
(consistent with Deroubaix et al. (2021); Ordóñez et al. (2020)), and a slight decrease
in mean in-situ O3 concentration in June 2020 compared to 2019. In our study areas (metropoli-
tan areas), the impact of meteorological conditions on in-situ O3 concentrations are clearly
noticeable in all periods. Meteorological conditions were favorable for high O3 concen-
Figure 3. Mean relative changes in meteorological impacts unaccounted (left column) and accounted (right column) NO$_2$ and O$_3$ concentrations in eight metropolitan cities between 2020 and 2019. Error bars represent the 1 $\sigma$ (standard deviation) of mean of eight metropolitan cities.
transitions between February 1 and May 31, 2020 (consistent with Gaubert et al. (2021)), while meteorological conditions were favorable for low O\textsubscript{3} concentrations in January and June 2020. For instance, before accounting for meteorology, mean O\textsubscript{3} concentration in June 2020 is 16.5 % lower than in 2019, which could be attributed to the low temperature (less ozone production) in June 2020 (Figure S8 (jj)). After accounting for meteorology, the difference between mean O\textsubscript{3} concentrations in June 2020 and the same period in 2019 is reduced to -3 %. Meteorology had a different impact on NO\textsubscript{2} and O\textsubscript{3} levels and this impact also varied for different time periods. This demonstrates the complex relationship between O\textsubscript{3}, NO\textsubscript{2}, and meteorological conditions.

We found large discrepancies between in-situ and TROPOMI NO\textsubscript{2} changes for the study period. It is important to note that the number of TROPOMI cloud-free measurements between 2020 and 2019 may have an impact on results (for Munich, TROPOMI measurements are available for 269 days out of 363 days). In addition, the TROPOMI overpass occurs at 13.30 local time, which may make it less sensitive to traffic-related emissions (peak in the morning from 7-9 am and evening from 4-7 pm). We conducted two comparisons between 2019 in-situ NO\textsubscript{2} and TROPOMI NO\textsubscript{2} measurements to determine whether the TROPOMI measurements (overpass occurs around 13.30) could represent traffic-related emissions. First, we compare the mean 24 hour in-situ NO\textsubscript{2} to the TROPOMI NO\textsubscript{2} observation. Second, we compare the in-situ NO\textsubscript{2} at the time of TROPOMI overpass with the TROPOMI NO\textsubscript{2}, which should have better agreement. We use the empirical relationship (Lorente et al., 2019) that includes boundary layer information to convert the surface concentration to column density. The TROPOMI observations agree well with the in-situ observations at the TROPOMI overpass time (mean bias (TROPOMI - in-situ) is about -13 %), whereas TROPOMI underestimates NO\textsubscript{2} compared to the 24-hr mean in-situ value (mean bias is about -41.5 %) (Figure S9, for Munich). This indicates that TROPOMI is not suitable to directly represent the 24-hr mean (daily concentration), which could lead to errors in estimating lockdown effects, because the lockdown primarly reduced traffic-related emissions. Furthermore, the observed satellite column concentration is certainly influenced by the background concentration. The free tropospheric background contributes 70-80 % of the total column observed via satellite (R. Silvern et al., 2018; Travis et al., 2016). R. F. Silvern et al. (2019) and Qu et al. (2021) demonstrate the importance of accounting for the influence of free tropospheric NO\textsubscript{2} background on satellite column measurements to infer the changes in surface NO\textsubscript{X} emission. The primary sources of background NO\textsubscript{2} are lightning, soil, wildfires and long-range transport of pollution (L. Zhang et al., 2012), which are unaffected by lockdown restrictions. The contribution from soil has been shown to increase up to 27 % of total NO\textsubscript{X} emissions at elevated temperatures (Butterbach-Bahl et al., 2001) (discussed below). In addition, subtracting the contribution of the NO\textsubscript{2} background from satellite column observation is complex, because of its non-uniformity (Marais et al., 2018, 2021), thus, using column measurements is challenging for estimates of local changes in NO\textsubscript{2} emissions. In contrast to satellite column measurements, background NO\textsubscript{2} has little influence (5-10 %) on in-situ surface NO\textsubscript{2} concentrations (R. F. Silvern et al., 2019). The discrepancies between in-situ and TROPOMI changes primarily results from unaccounted background NO\textsubscript{2} influence on the satellite observation and that TROPOMI’s overpass time makes it less sensitive to overall diurnal emissions. These discrepancies limit the use of satellite column measurements to infer the surface NO\textsubscript{X} emission changes.

The NO\textsubscript{2} column densities in rural locations were also investigated. During the 2020 stricter lockdown period, after accounting for meteorology, slightly increased NO\textsubscript{2} vertical column densities over North-West Germany are observed compared to 2019 (figure 4 (c)). We hypothesise that this is due to enhanced soil NO\textsubscript{X} emissions over North-West Germany in the 2020 stricter lockdown period (associated with increased temperature over North-West Germany (Figure S8 (f)); soil NO\textsubscript{X} emissions typically increase with temperature (Oikawa et al., 2015). Soil NO\textsubscript{X} emissions are expected to be high in the early spring (stricter lockdown period), even though the average temperature in May
Figure 4. The absolute difference in TROPOMI (a) and GEOS-Chem (b) NO$_2$ column densities between 2020 and 2019 (stricter lockdown period: March 21 to April 30). The absolute difference between first two columns is shown in panel (c).

and June is higher than in the stricter lockdown period, because agricultural practices such as fertilizer application begin and end in the early spring (Ramanantenasoa et al., 2018; Viatte et al., 2020). Fertilized soils have high potential for NO$_X$ emissions (Almaraz et al., 2018; S. Liu et al., 2017; Skiba et al., 2021). Figure S11 shows the monthly mean NH$_3$ total column densities over Germany. High NH$_3$ total column densities were observed in April as agricultural practices (fertilizer application) began in the early spring. Notably, strong enhancements were observed over North-West Germany. The total column of NH$_3$ over North-West Germany in 2020 (strict lockdown period) is higher than in 2019 (Figure S12). This supports our hypothesis that North-West Germany, which is dominated by Grass and Crop land (ESA CCI, 2020), is an agricultural region, with fertilized soil producing NO$_X$ in elevated-temperature environments.

5 Ozone sensitivity to NO$_X$ changes

Like previous studies that reported the urban NO$_2$ weekly cycle (Beirle et al., 2003; Ialongo et al., 2020), we also investigate this at the national (Germany) and urban scale (Figure S13 & S14). Both TROPOMI and in-situ NO$_2$ measurements show that weekend NO$_2$ concentrations are lower than weekday NO$_2$ concentrations, because primary emission activities such as transportation typically decrease on weekends. Studies (Sicard et al., 2020; Wang et al., 2014) have demonstrated that analysing the difference in weekday vs weekend O$_3$ concentrations helps identify the ozone production regime. As NO$_X$ emissions decrease on weekends, the response of ozone will demonstrate whether ozone production is NO$_X$ limited or saturated. Hammer et al. (2002); Gaubert et al. (2021) used the H$_2$O$_2$/HNO$_3$ ratio and Sillman et al. (2003) used the O$_3$/NO$_y$ ratio as a way to identify the ozone production regime over Europe urban regions. Previous studies (Beekmann & Vautard, 2010; Derwent et al., 2003; Gabusi & Volta, 2005; Gaubert et al., 2021; Martin et al., 2004) have demonstrated that European urban regions are characterized as NO$_X$ saturated ozone production regime. The influence of biogenic VOC emissions on ozone is relatively low in Europe (Curci et al., 2009; Simpson, 1995). There also is a shift between NO$_X$ saturated and NO$_X$ limited regimes during the year; in the winter, ozone production is usually NO$_X$ saturated, whereas it is often NO$_X$ limited in the summer (Jin et al., 2017). The winter and spring O$_3$ weekend effect is much stronger than the summer O$_3$ weekend effect (Figure 5, for Munich metropolitan area): reduced NO$_X$ emission on weekends increase O$_3$ concentrations, i.e., NO$_X$ saturated conditions prevail, consistent with above mentioned previous studies, which shows that NO$_X$ saturated conditions persist to the current time period. Therefore, German metropolitan areas are expected to be in a NO$_X$ saturated ozone production regime also during the initial 2020 COVID-19 pandemic period. Notably, we found an increase (4 %) in meteorology ac-
Figure 5. Mean relative change in in-situ NO$_2$ and O$_3$ concentrations in Munich between weekends and weekdays. Error bars represent statistical uncertainty (1 σ) in the calculation of relative change between weekend and weekday means.

A year-to-year comparison of atmospheric pollutant concentrations is widely used to estimate the influence of reductions in anthropogenic emissions on atmospheric pollutant concentration changes during the COVID-19 pandemic period. However, these findings could be misleading if meteorological impacts are not taken into account. We used identical anthropogenic emissions in 2020 and 2019 in GEOS-Chem model simulations, allowing us to separate the changes in NO$_2$ and O$_3$ attributed to meteorological impacts from the observed changes. Finally, we show that, due to reductions in anthropogenic emissions during the COVID-19 pandemic period, meteorology accounted for mean in-situ & TROPOMI NO$_2$ concentrations decreased by 23 % & 16 %, respectively, compared to 2019, in eight German metropolitan cities between March 21 and June 30. After accounting for meteorology, we find a nationwide decrease in TROPOMI NO$_2$ concentrations except for North-West Germany, which can be attributed to enhanced
NO\textsubscript{X} emissions from agricultural soils during the 2020 stricter lockdown period. We hypothesise that North-West Germany is a hot spot of soil NO\textsubscript{X} emissions in elevated-temperature environments due to intensive agricultural practices (fertilizer applications) during the early spring. The IASI NH\textsubscript{3} satellite data also supports our statement that North-West Germany is an intensive agricultural region during the early spring.

After accounting for meteorology, the concentration of O\textsubscript{3} increased slightly (4 \%) during the 2020 spring lockdown while it decreased slightly (3 \%) during the 2020 early summer lockdown, in response to decreased NO\textsubscript{2} in both time periods, compared to 2019. This implies that the degree of NO\textsubscript{X} saturation of ozone production is weakening from winter to summer. These findings are also supported by the response of O\textsubscript{3} to changes in precursor emissions using weekend vs. weekday differences. Therefore, reducing NO\textsubscript{X} emissions would benefit summer ozone reduction, whereas reducing NO\textsubscript{X} emissions would increase ozone levels during winter and spring. Appropriate NO\textsubscript{X} and VOCs emission control strategies are required to mitigate ozone pollution in German metropolitan areas during winter and spring; otherwise, it may lead to incorrect environmental regulation policies that are closely linked to public health. Despite a sharp decrease in emissions from the transportation sector, emissions from natural sources (dust storms, wildfires) and agriculture sectors were unaffected by 2020 COVID-19 lockdown restrictions. Changes in other pollutants such as PM\textsubscript{10}, SO\textsubscript{2}, CO and anthropogenic VOCs (primary pollutant) and PM\textsubscript{2.5} (secondary pollutant) may provide further insight on air quality during the COVID-19 pandemic period. Extensive studies on air quality during the lockdown period could pave the way for an improved understanding of pollution formation. Those findings will be useful in understanding how reductions in primary emissions affect secondary pollutant formation.

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References


Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.-F., Van Gent, J., Eskes, H., ... others (2020). Impact of coronavirus outbreak on no\textsubscript{2} pollution assessed using tropomi and omi observations. Geophysical Research Letters, 47(11), e2020GL087978.


Despite the influence of weather patterns, the effect of the Coronavirus on air quality is now visible. https://www.dlr.de/content/en/articles/news/2020/02/20200505_effect-of-the-coronavirus-on-air-quality-is-now-visible.html.


lockdown upon \( \text{PO}_{2} \) pollution in Spain. *Atmospheric Chemistry and Physics*, 20(18), 11119–11141.


