Tropospheric NO\textsubscript{2} and O\textsubscript{3} response to COVID-19 lockdown restrictions at the national and urban scales in Germany

Vigneshkumar Balamurugan\textsuperscript{1}, Jia Chen\textsuperscript{1}, Zhen Qu\textsuperscript{2}, Xiao Bi\textsuperscript{1}, Johannes Gensheimer\textsuperscript{1}, Ankit Shekhar\textsuperscript{3}, Shrutilipi Bhattacharjee\textsuperscript{4}, Frank N. Keutsch\textsuperscript{2,5}

\textsuperscript{1}TUM Department of Electrical and Computer Engineering, Technische Universität München, Munich, Germany
\textsuperscript{2}School of Engineering and Applied Science, Harvard University, Cambridge, Massachusetts, USA
\textsuperscript{3}Department of Environmental Systems Science, ETH Zurich, Switzerland
\textsuperscript{4}Department of Information Technology, National Institute of Technology Karnataka, Surathkal, India
\textsuperscript{5}Department of Chemistry and Chemical Biology, Harvard University, Cambridge, Massachusetts, USA

Key Points:

- During the COVID-19 lockdown period, NO\textsubscript{2} concentrations decreased and O\textsubscript{3} concentrations increased in eight German cities
- The degree of NO\textsubscript{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

Corresponding author: Vigneshkumar Balamurugan, Jia chen, vigneshkumar.balamurugan@tum.de, jia.chen@tum.de
Abstract
This study estimates the influence of anthropogenic emission reductions on nitrogen dioxide (NO\textsubscript{2}) and ozone (O\textsubscript{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\textsubscript{2} concentrations by 23 % (\& 16 %) between March 21 and June 30, 2020 after accounting for meteorology, whereas the corresponding mean in-situ O\textsubscript{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\textsubscript{X} saturation of ozone production is stronger than in the summer. This implies that future reductions in NO\textsubscript{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\textsubscript{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\textsubscript{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\textsubscript{2}) and ozone (O\textsubscript{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\textsubscript{2} concentrations during the 2020 lockdown period, an increase in O\textsubscript{3} concentrations during the 2020 spring lockdown, and a decrease in O\textsubscript{3} concentrations during the 2020 early summer lockdown. The increased O\textsubscript{3} concentration in response to the decreased NO\textsubscript{2} concentration implies that future reductions in NO\textsubscript{X} emissions are likely to increase ozone pollution in German metropolitan areas during winter and spring. Furthermore, there was a nationwide decrease in NO\textsubscript{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\textsubscript{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 (\textit{DW COVID-19}, 2020). These measures resulted in a significant reduction in emissions following economic activity and overall mobility (Evangelion 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\textsubscript{2}), carbon monoxide (CO), sulphur dioxide (SO\textsubscript{2}) and particulate matter (PM\textsubscript{10} and PM\textsubscript{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); Deroubai 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; 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 (Doumbia 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 ($\approx$ 21 hours) than in the summer ($\approx$ 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; Stillman, 1999; Stillman 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 meteorological influences, 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 (Supplement figure S3). This study mainly focused on the urban scale to examine the impact of reduced mobility on NO₂ and O₃ concentrations during the 2020 COVID-19 pandemic period. We also extended our study nationwide to investigate other significant NOₓ sources in rural locations.

We used tropospheric NO₂ 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 NO₂ 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 NO₂ precision (error estimate originating from the spectral fit and other retrieval aspects) for each pixel is within the range of 3.6*10¹⁴ to 3.7*10¹⁶ molec. cm⁻² (about 21% to 75 % of tropospheric NO₂ column). The TROPOMI NO₂ 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 NO₂ 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 NO₂ column densities were gridded in 0.25*0.25-degree bins.

We investigate agricultural activities in Germany using ammonia (NH₃) data (Kuttippurath et al., 2020). The “Standard monthly IASI/Metop-B ammonia (NH₃) data set” was downloaded from IASI NH₃ (2020). This data set contains monthly averaged NH₃ 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 NH₃ over Germany (IASI NH₃, 2020).

In-situ surface NO₂ and O₃ 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 NO₂ and O₃ between 2020 and 2019 caused by meteorological changes. The GEOS-Chem model is driven by MERRA-2 assimilated me-
We conduct nested simulations over Germany (41°-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 NOx 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 NOx emissions (Hudman et al., 2012). In all comparisons of the GC model to TROPOMI, GC NO2 vertical profile simulations (at 47 vertical layer) are converted to NO2 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 NO2 and O3 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 NO2, O3 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 NO2 and O3 between 2020 and 2019 are solely due to meteorological influences, e.g., differences in wind speed, boundary layer height, photo-chemistry etc.:

\[
\Delta NO_2(GC) = NO_2(GC,2020) - NO_2(GC,2019)
\]

\[
\Delta O_3(GC) = O_3(GC,2020) - O_3(GC,2019)
\]

The difference between the 2020 and 2019 NO2 and O3 observations for specific time periods include influence from both meteorological and emissions changes:

\[
\Delta NO_2(\text{obs}) = NO_2(\text{obs,2020}) - NO_2(\text{obs,2019})
\]

\[
\Delta O_3(\text{obs}) = O_3(\text{obs,2020}) - O_3(\text{obs,2019})
\]

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),

\[
\Delta NO_2(\text{acc}) = \Delta NO_2(\text{obs}) - \Delta NO_2(GC)
\]

and similarly for ozone:

\[
\Delta O_3(\text{acc}) = \Delta O_3(\text{obs}) - \Delta O_3(GC)
\]

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 NO2 and O3: impact of meteorological conditions

Like previous studies (Çelik & Ibrahim, 2007; Deroubaix et al., 2021; Ordóñez et al., 2020; Voiculescu et al., 2020), we investigated correlations between satellite and in-situ NO2 and O3 concentrations and local meteorological parameters to find the dependency of NO2 and O3 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 NO2 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 positive 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 various 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 emissions, which can raise the ozone level, especially in the summer. Future climate conditions 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; Vautard 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 levels will increase (De Sario et al., 2013; Meehl & Tebaldi, 2004). Furthermore, temperature, boundary layer height and solar radiation, which are considered to be the most related meteorological factors influencing NO$_2$ and O$_3$ concentrations, are interdependent.

### 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 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 February 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 measures 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)). However, 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 (Figure 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 O$_3$ concentrations in January and June 2020, and higher mean O$_3$ 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 NO$_2$ and O$_3$ 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 NO$_2$, O$_3$ and particulate matter, implying that GC accounts for impacts of meteorology when using precise meteorological data and emission 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 concentration 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 surface NO$_2$ concentrations and in-situ surface NO$_2$ concentrations for eight metropolitan areas ($R$, pearson correlation coefficient, $> 0.65$, with high $R$ (0.75) for Cologne). Similar results were obtained for GC surface O$_3$ concentrations, ($R > 0.65$, with a high $R$ (0.74) for Dresden). GC underestimates NO$_2$ surface concentrations, except for Hamburg. The mean bias (GC - in-situ) ranges from +2.9 to -23 %. Except for Hamburg and Hanover, GC overestimates surface O$_3$ concentrations, with mean bias ranges from +24 to -10.3 %. When comparing 2019 GC and TROPOMI NO$_2$ column densities, relatively low correlation ($R$, between 0.24 and 0.55) was found, and the NO$_2$ column densities in metropolitan areas were underestimated by GC (mean bias ranges from -4 to -28 %). However, the GC model is capable of modeling the spatial variability of NO$_2$ column densities at the national scale, emphasizing GC’s ability to represent the distribution of emission source locations (Figure S7). The over/under estimation of NO$_2$ and O$_3$ concentrations are caused by the emission inventory (over/under estimation of emission) used in GC simulation. The low bias in NO$_2$ and high bias in O$_3$ could be consistent with NO$_X$ saturated conditions. Because we use the difference in GC concentrations between 2020 and 2019 ($\Delta$ NO$_2$(GC) and $\Delta$ O$_3$(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 implementation of lockdown restrictions) to determine the extent to which meteorology is responsible for variations in pollutant concentrations. Before accounting for meteorology, the difference in mean in-situ NO$_2$ concentration between 2020 and 2019 is -28 % for the period February 1 and March 20. After accounting for meteorology, the difference is reduced 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 impacts of meteorology on in-situ and TROPOMI NO$_2$ 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 meteorology, the mean in-situ and TROPOMI NO$_2$ 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 meteorological impacts also found that the impact of the COVID-19 pandemic on NO$_X$ emissions lasted at least until June 2020. After accounting for meteorology, we observed a slight increase in mean in-situ O$_3$ concentration between March 21 and May 31, 2020 (consistent with Deroubaix et al. (2021); Ordoñez et al. (2020)), and a slight decrease in mean in-situ O$_3$ concentration in June 2020 compared to 2019. In our study areas (metropolitan areas), the impact of meteorological conditions on in-situ O$_3$ concentrations are clearly noticeable in all periods. Meteorological conditions were favorable for high O$_3$ 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.
trations between February 1 and May 31, 2020 (consistent with Gaubert et al. (2021)), while meteorological conditions were favorable for low O₃ concentrations in January and June 2020. For instance, before accounting for meteorology, mean O₃ 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 (j)). After accounting for meteorology, the difference between mean O₃ concentrations in June 2020 and the same period in 2019 is reduced to -3 %. Meteorology had a different impact on NO₂ and O₃ levels and this impact also varied for different time periods. This demonstrates the complex relationship between O₃, NO₂, and meteorological conditions.

We found large discrepancies between in-situ and TROPOMI NO₂ 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 NO₂ and TROPOMI NO₂ 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 NO₂ to the TROPOMI NO₂ observation. Second, we compare the in-situ NO₂ at the time of TROPOMI overpass with the TROPOMI NO₂, 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 NO₂ 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 primarily 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 NO₂ background on satellite column measurements to infer the changes in surface NOₓ emission. The primary sources of background NO₂ 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 NOₓ emissions at elevated temperatures (Butterbach-Bahl et al., 2001) (discussed below). In addition, subtracting the contribution of the NO₂ 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 NO₂ emissions. In contrast to satellite column measurements, background NO₂ has little influence (5-10 %) on in-situ surface NO₂ concentrations (R. F. Silvern et al., 2019). The discrepancies between in-situ and TROPOMI changes primarily results from unaccounted background NO₂ 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 NOₓ emission changes.

The NO₂ column densities in rural locations were also investigated. During the 2020 stricter lockdown period, after accounting for meteorology, slightly increased NO₂ vertical column densities over North-West Germany are observed compared to 2019 (Figure 4 (c)). We hypothesise that this is due to enhanced soil NOₓ emissions over North-West Germany in the 2020 stricter lockdown period (associated with increased temperature over North-West Germany (Figure S8 (f)); soil NOₓ emissions typically increase with temperature (Oikawa et al., 2015). Soil NOₓ 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\textsubscript{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\textsubscript{X} emissions (Almaraz et al., 2018; S. Liu et al., 2017; Skiba et al., 2021). Figure S11 shows the monthly mean NH\textsubscript{3} total column densities over Germany. High NH\textsubscript{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\textsubscript{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\textsubscript{X} in elevated-temperature environments.

5 Ozone sensitivity to NO\textsubscript{X} changes

Like previous studies that reported the urban NO\textsubscript{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\textsubscript{2} measurements show that weekend NO\textsubscript{2} concentrations are lower than weekday NO\textsubscript{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\textsubscript{3} concentrations helps identify the ozone production regime. As NO\textsubscript{X} emissions decrease on weekends, the response of ozone will demonstrate whether ozone production is NO\textsubscript{X} limited or saturated. Hammer et al. (2002); Gaubert et al. (2021) used the H\textsubscript{2}O\textsubscript{2}/HNO\textsubscript{3} ratio and Sillman et al. (2003) used the O\textsubscript{3}/NO\textsubscript{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\textsubscript{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\textsubscript{X} saturated and NO\textsubscript{X} limited regimes during the year; in the winter, ozone production is usually NO\textsubscript{X} saturated, whereas it is often NO\textsubscript{X} limited in the summer (Jin et al., 2017). The winter and spring O\textsubscript{3} weekend effect is much stronger than the summer O\textsubscript{3} weekend effect (Figure 5, for Munich metropolitan area); reduced NO\textsubscript{X} emission on weekends increase O\textsubscript{3} concentrations, i.e., NO\textsubscript{X} saturated conditions prevail, consistent with above mentioned previous studies, which shows that NO\textsubscript{X} saturated conditions persist to the current time period. Therefore, German metropolitan areas are expected to be in a NO\textsubscript{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 $\sigma$) in the calculation of relative change between weekend and weekday means.

6 Conclusions

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$_X$ emissions from agricultural soils during the 2020 stricter lockdown period. We hypothesize that North-West Germany is a hot spot of soil NO$_X$ emissions in elevated-temperature environments due to intensive agricultural practices (fertilizer applications) during the early spring. The IASI NH$_3$ 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$_3$ increased slightly (4 %) during the 2020 spring lockdown while it decreased slightly (3 %) during the 2020 early summer lockdown, in response to decreased NO$_2$ in both time periods, compared to 2019. This implies that the degree of NO$_X$ saturation of ozone production is weakening from winter to summer. These findings are also supported by the response of O$_3$ to changes in precursor emissions using weekend vs. weekday differences. Therefore, reducing NO$_X$ emissions would benefit summer ozone reduction, whereas reducing NO$_X$ emissions would increase ozone levels during winter and spring. Appropriate NO$_X$ 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$_{10}$, SO$_2$, CO and anthropogenic VOCs (primary pollutant) and PM$_{2.5}$ (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.

Acknowledgments
This work has been supported by Institute for Advanced Study, Technical University of Munich, through the German Excellence Initiative and the European Union Seventh Framework Program (Grant: 291763) and in part by the German Research Foundation (DFG) (Grant: 419317138). The authors declare no conflicts of interest. The TROPOMI NO$_2$ data can be found at https://s5phub.copernicus.eu/. The IASI NH$_3$ data is available at https://iasi.aeris-data.fr/catalog/. Hourly NO$_2$ and O$_3$ concentrations are downloaded from UBA’s website (https://www.umweltbundesamt.de/en/data). Hourly ERA5 meteorological data are freely available at https://cds.climate.copernicus.eu/

References

Almaraz, M., Bai, E., Wang, C., Trousdell, J., Conley, S., Faloona, I., & Houlton, B. Z. (2018). Agriculture is a major source of no$_x$ pollution in california. Science advances, 4(1), eaao3477.

Barré, J., Petetin, H., Colette, A., Guevara, M., Peuch, V.-H., Rouil, L., . . . others (2020). Estimating lockdown induced european no$_2$ changes. Atmospheric Chemistry and Physics Discussions, 1–28.

Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.-F., Van Gent, J., Eskes, H., . . . others (2020). Impact of coronavirus outbreak on no$_2$ pollution assessed using tropomi and omi observations. Geophysical Research Letters, 47(11), e2020GL087978.

Bedi, J. S., Dhaka, P., Vijay, D., Aulakh, R. S., Gill, J. P. S., et al. (2020). Assessment of air quality changes in the four metropolitan cities of india during covid-19 pandemic lockdown. Aerosol and Air Quality Research, 20(10), 2062–2070.

Beekmann, M., & Vautard, R. (2010). A modelling study of photochemical regimes over europe: robustness and variability. Atmospheric Chemistry and Physics, 10(20), 10067–10084.
Beirle, S., Borger, C., Dörner, S., Li, A., Hu, Z., Liu, F., . . . Wagner, T. (2019). Pinpointing nitrogen oxide emissions from space. Science advances, 5(11), eaax9800.

Beirle, S., Platt, U., Wenig, M., & Wagner, T. (2003). Weekly cycle of no2 by gome measurements: a signature of anthropogenic sources. Atmospheric Chemistry and Physics, 3(6), 2225–2232.

Berman, J. D., & Ebisu, K. (2020). Changes in us air pollution during the covid-19 pandemic. Science of the Total Environment, 739, 139864.

Bozem, H., Butler, T. M., Lawrence, M. G., Harder, H., Martinez, M., Kubistin, D., . . . Fischer, H. (2017). Chemical processes related to net ozone tendencies in the free troposphere. Atmospheric Chemistry and Physics, 17(17), 10565–10582.

Butterbach-Bahl, K., Stange, F., Papen, H., & Li, C. (2001). Regional inventory of nitric oxide and nitrous oxide emissions for forest soils of southeast germany using the biogeochemical model pnet-n-dndc. Journal of Geophysical Research: Atmospheres, 106(D24), 34155–34166.

Çelik, M. B., & İbrahim, K. (2007). The relation between meteorological factors and pollutants concentrations in karabük city. Gazi University Journal of Science, 20(4), 87–95.

Chauhan, A., & Singh, R. P. (2020). Decline in pm2.5 concentrations over major cities around the world associated with covid-19. Environmental Research, 187, 109634.

Coates, J., Mar, K. A., Ojha, N., & Butler, T. M. (2016). The influence of temperature on ozone production under varying nox conditions—a modelling study. Atmospheric Chemistry and Physics, 16(18), 11601–11615.

Collivignarelli, M. C., Abbà, A., Bertanza, G., Pedrazzani, R., Ricciardi, P., & Mino, M. C. (2020). Lockdown for covid-2019 in milan: What are the effects on air quality? Science of The Total Environment, 732, 139280.

Copernicus. (2020). Sentinel-5P Pre-Operations Data Hub. https://s5phub.copernicus.eu/dhus/#/home.

Copernicus climate change service (c3s). (2017). ERA5: Fifth generation of ECMWF atmospheric reanalyses of the global climate. Copernicus Climate Change Service Climate Data Store (CDS). https://cds.climate.copernicus.eu/cdsapp#!/home. (Accessed: 2020-12-31)

Council, N. R., et al. (1992). Rethinking the ozone problem in urban and regional air pollution. National Academies Press.

Crutzen, P. J. (1988). Tropospheric ozone: An overview. Tropospheric ozone, 3–32.

Curci, G., Beekmann, M., Vautard, R., Smiatek, G., Steinbrecher, R., Theloke, J., & Friedrich, R. (2009). Modelling study of the impact of isoprene and terpene biogenic emissions on european ozone levels. Atmospheric Environment, 43(7), 1444–1455.

Deroubaix, A., Brasseur, G., Gaubert, B., Labuhn, I., Memut, L., Siour, G., & Tuccella, P. (2021). Response of surface ozone concentration to emission reduction and meteorology during the covid-19 lockdown in europe. Meteorological Applications, 28(3), e1990.

Derwent, R., Jenkin, M., Saunders, S., Pilling, M., Simmonds, P., Passant, N., . . . Kent, A. (2003). Photochemical ozone formation in north west europe and its control. Atmospheric Environment, 37(14), 1983–1991.

De Sario, M., Katsoyannou, K., & Michelozzi, P. (2013). Climate change, extreme weather events, air pollution and respiratory health in europe. European Respiratory Journal, 42(3), 826–843.

Dietrich, F., Chen, J., Voggenreiter, B., Aigner, P., Nachtigall, N., & Reger, B. (2021). Muccnet: Munich urban carbon column network. Atmospheric Measurement Techniques, 14(2), 1111–1126.
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.

Doumbia, T., Granier, C., Elguindi, N., Bouarar, I., Darras, S., Brasseur, G., others (2021). Changes in global air pollutant emissions during the covid-19 pandemic: a dataset for atmospheric chemistry modeling. Earth System Science Data Discussions, 1–26.

Dw covid-19. (2020). Coronavirus: What are the lockdown measures across Europe?. https://www.dw.com/en/coronavirus-what-are-the-lockdown-measures-across-europe/a-52905137. (Accessed: 2020-12-31)

Dwd. (2020). The German Weather Service. https://www.dwd.de/DE/Home/home_node.html.

Engardt, M., Bergström, R., & Andersson, C. (2009). Climate and emission changes contributing to changes in near-surface ozone in europe over the coming decades: results from model studies. Ambio, 452–458.

Esa cci. (2020). ESA-CCI land cover product. https://maps.elie.ucl.ac.be/CCI/viewer/.

Evangeliou, N., Platt, S. M., Eckhardt, S., Lund Myhre, C., Laj, P., Alados-Arboledas, L., others (2021). Changes in black carbon emissions over europe due to covid-19 lockdowns. Atmospheric Chemistry and Physics, 21(4), 2675–2692.

Fiore, A., Jacob, D. J., Liu, H., Yantosca, R. M., Fairlie, T. D., & Li, Q. (2003). Variability in surface ozone background over the united states: Implications for air quality policy. Journal of Geophysical Research: Atmospheres, 108(D24).

Forkel, R., & Knoche, R. (2006). Regional climate change and its impact on photooxidant concentrations in southern germany: Simulations with a coupled regional climate-chemistry model. Journal of Geophysical Research: Atmospheres, 111(D12).

Forster, P. M., Forster, H. I., Evans, M. J., Gidden, M. J., Jones, C. D., Keller, C. A., others (2020). Current and future global climate impacts resulting from covid-19. Nature Climate Change, 10(10), 913–919.

Fu, F., Purvis-Roberts, K. L., & Williams, B. (2020). Impact of the covid-19 pandemic lockdown on air pollution in 20 major cities around the world. Atmosphere, 11(11), 1189.

Gabusi, V., & Volta, M. (2005). Seasonal modelling assessment of ozone sensitivity to precursors in northern italy. Atmospheric Environment, 39(15), 2785–2804.

Gaubert, B., Bouarar, I., Doumbia, T., Liu, Y., Stavrakou, T., Deroubaix, A., others (2021). Global changes in secondary atmospheric pollutants during the 2020 covid-19 pandemic. Journal of Geophysical Research: Atmospheres, 126(8), e2021JD034213.

Gensheimer, J., Turner, A. J., Shekhar, A., Wenzel, A., Keutsch, F. N., & Chen, J. (2021). What are the different measures of mobility telling us about surface transportation co2 emissions during the covid-19 pandemic? Journal of Geophysical Research: Atmospheres, e2021JD034664.

Geos-chem. (2020). The International GEOS-Chem User Community: GEOS-Chem 12.9.2 http://doi.org/10.5281/zenodo.3959279.

Goldberg, D. L., Anenberg, S. C., Griffin, D., McLinden, C. A., Lu, Z., & Streets, D. G. (2020). Disentangling the impact of the covid-19 lockdowns on urban no2 from natural variability. Geophysical Research Letters, 47(17), e2020GL089269.

Grange, S. K., Lee, J. D., Drysdale, W. S., Lewis, A. C., Hueglin, C., Emmenegger, L., & Carslaw, D. C. (2020). Covid-19 lockdowns highlight a risk of increasing ozone pollution in european urban areas. Atmospheric Chemistry and Physics Discussions, 1–25.
Guevara, M., Jorba, O., Soret, A., Petetin, H., Bowdalo, D., Serradell, K., . . . others (2021). Time-resolved emission reductions for atmospheric chemistry modelling in europe during the covid-19 lockdowns. *Atmospheric Chemistry and Physics, 21*(2), 773–797.

Hammer, M.-U., Vogel, B., & Vogel, H. (2002). Findings on $\text{H}_2\text{O}/\text{HNO}_3$ as an indicator of ozone sensitivity in baden-württemberg, berlin-brandenburg, and the po valley based on numerical simulations. *Journal of Geophysical Research: Atmospheres, 107*(D22), LOP–3.

He, G., Pan, Y., & Tanaka, T. (2020). The short-term impacts of covid-19 lockdown on urban air pollution in china. *Nature Sustainability*, 1–7.

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Jaansens-Maenhout, G., Pitkänen, T., . . . others (2018). Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the community emissions data system (ceds). *Geoscientific Model Development, 11*(1), 369–408.

Hudman, R., Moore, N., Mebust, A., Martin, R., Russell, A., Valin, L., & Cohen, R. (2012). Steps towards a mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints. *Atmospheric Chemistry and Physics, 12*(16), 7779–7795.

Ialongo, I., Virta, H., Eskes, H., Hovila, J., & Douros, J. (2020). Comparison of tropomi/sentinel-5 precursor $\text{NO}_2$ observations with ground-based measurements in helsinki. *Atmospheric Measurement Techniques, 13*(1).

Iasi nh3. (2020). IASI NH$_3$ product. https://iasi.aeris-data.fr/catalog/.

Jacob, D. J. (1999). *Introduction to atmospheric chemistry*. Princeton University Press.

Jin, X., Fiore, A. M., Murray, L. T., Valin, L. C., Lamсал, L. N., Duncan, B., . . . others (2017). Evaluating a space-based indicator of surface ozone-$\text{NO}_2\text{-voc}$ sensitivity over midlatitude source regions and application to decadal trends. *Journal of Geophysical Research: Atmospheres, 122*(19), 10–439.

Kang, M., Zhang, J., Zhang, H., & Ying, Q. (2021). On the relevancy of observed ozone increase during covid-19 lockdown to summertime ozone and $\text{PM}_{2.5}$ control policies in china. *Environmental Science & Technology Letters, 8*(4), 289–294.

Keller, C. A., Evans, M. J., Knowland, K. E., Hasenkopf, C. A., Modekurty, S., Luchesi, R. A., . . . others (2021). Global impact of covid-19 restrictions on the surface concentrations of nitrogen dioxide and ozone. *Atmospheric Chemistry and Physics, 21*(5), 3555–3592.

Kleinman, L. I. (2005). The dependence of tropospheric ozone production rate on ozone precursors. *Atmospheric Environment, 39*(3), 575–586.

Kleinman, L. I., Daum, P. H., Lee, J. H., Lee, Y.-N., Nunnermacker, L. J., Springston, S. R., . . . Sillman, S. (1997). Dependence of ozone production on $\text{NO}$ and hydrocarbons in the troposphere. *Geophysical Research Letters, 24*(18), 2299–2302.

Kroll, J. H., Heald, C. L., Cappa, C. D., Farmer, D. K., Fry, J. L., Murphy, J. G., & Steiner, A. L. (2020). The complex chemical effects of covid-19 shutdowns on air quality. *Nature Chemistry, 12*(9), 777–779.

Kuenen, J., Visschedijk, A., Jozwicka, M., & Denier Van Der Gon, H. (2014). Tnomac2li emission inventory: a multi-year (2003–2009) consistent high-resolution european emission inventory for air quality modelling. *Atmos. Chem. Phys., 14*(20), 10963–10976.

Kuttipurath, J., Singh, A., Dash, S., Mallick, N., Clerbaux, C., Van Damme, M., . . . others (2020). Record high levels of atmospheric ammonia over india: Spatial and temporal analyses. *Science of the Total Environment, 740*, 139986.

Le Quéré, C., Jackson, R. B., Jones, M. W., Smith, A. J., Abernethy, S., Andrew, R. M., . . . others (2020). Temporary reduction in daily global $\text{CO}_2$ emissions during the covid-19 forced confinement. *Nature Climate Change, 1*, 1–7.
Lin, X., Trainer, M., & Liu, S. (1988). On the nonlinearity of the tropospheric ozone production. *Journal of Geophysical Research: Atmospheres, 93*(D12), 15879–15888.

Liu, S., Lin, F., Wu, S., Ji, C., Sun, Y., Jin, Y., . . . Zou, J. (2017). A meta-analysis of fertilizer-induced soil no and combined no+ n2o emissions. *Global Change Biology, 23*(6), 2520–2532.

Liu, Y., Wang, T., Stavroulakis, T., Elguindi, N., Doumbia, T., Granier, C., . . . Brasseur, G. P. (2020). Diverse response of surface ozone to covid-19 lockdown in china. *arXiv preprint arXiv:2008.10851*.

Liu, Z., Ciais, P., Deng, Z., Lei, R., Davis, S. J., Feng, S., . . . others (2020). Covid-19 causes record decline in global co2 emissions. *Nature communications, 11*(1), 1–12.

Liu, Z., Deng, Z., Ciais, P., Lei, R., Davis, S. J., Feng, S., . . . others (2020). Near-real-time monitoring of global co2 emissions reveals the effects of the covid-19 pandemic. *Nature communications, 11*(1), 1–12.

Liu, Z., Ciais, P., Deng, Z., Lei, R., Davis, S. J., Feng, S., . . . others (2020). Covid-19 causes record decline in global co2 emissions.
lockdown upon no\textsubscript{2} pollution in spain. \textit{Atmospheric Chemistry and Physics}, \textit{20}(18), 11119–11141.

Potts, D. A., Marais, E. A., Boesch, H., Pope, R. J., Lee, J., Drysdale, W., \ldots others (2021). Diagnosing air quality changes in the uk during the covid-19 lockdown using tropomi and geos-chem. \textit{Environmental Research Letters}, \textit{16}(5), 054031.

Qu, Z., Jacob, D. J., Silvern, R. F., Shah, V., Campbell, P. C., Valin, L. C., & Murray, L. T. (2021). Us covid-19 shutdown demonstrates importance of background no\textsubscript{2} in inferring no\textsubscript{x} emissions from satellite no\textsubscript{2} observations. \textit{Geophysical Research Letters}, e2021GL092783.

Ramanantenasoa, M. M. J., Gilliot, J.-M., Mignolet, C., Bedos, C., Mathias, E., Eglin, T., \ldots Génermont, S. (2018). A new framework to estimate spatio-temporal ammonia emissions due to nitrogen fertilization in france. \textit{Science of the Total Environment}, \textit{645}, 205–219.

Shah, V., Jacob, D. J., Li, K., Silvern, R. F., Zhai, S., Liu, M., \ldots Zhang, Q. (2020). Effect of changing no\textsubscript{x} lifetime on the seasonality and long-term trends of satellite-observed tropospheric no\textsubscript{2} columns over china. \textit{Atmospheric Chemistry and Physics}, \textit{20}(3), 1483–1495.

Sharma, S., Zhang, M., Gao, J., Zhang, H., Kota, S. H., \ldots others (2020). Effect of restricted emissions during covid-19 on air quality in india. \textit{Science of the Total Environment}, \textit{728}, 138878.

Sicard, P., Paoletti, E., Agathokleous, E., Araminiê, V., Proietti, C., Coulibaly, F., \& De Marco, A. (2020). Ozone weekend effect in cities: Deep insights for urban air pollution control. \textit{Environmental research}, \textit{191}, 110193.

Sillman, S. (1999). The relation between ozone, no\textsubscript{x} and hydrocarbons in urban and polluted rural environments. \textit{Atmospheric Environment}, \textit{33}(12), 1821–1845.

Sillman, S., Logan, J. A., \& Wofsy, S. C. (1990). The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes. \textit{Journal of Geophysical Research: Atmospheres}, \textit{95}(D2), 1837–1851.

Sillman, S., Vautard, R., Menut, L., \& Kley, D. (2003). O\textsubscript{3}-no\textsubscript{x}-voc sensitivity and no\textsubscript{x}-voc indicators in paris: Results from models and atmospheric pollution over the paris area (esquif) measurements. \textit{Journal of Geophysical Research: Atmospheres}, \textit{108}(D17).

Silvern, R., Jacob, D., Travis, K., Sherwen, T., Evans, M., Cohen, R., \ldots others (2018). Observed no/no\textsubscript{2} ratios in the upper troposphere imply errors in no\textsubscript{2}-no\textsubscript{2} cycling kinetics or an unaccounted no\textsubscript{x} reservoir. \textit{Geophysical Research Letters}, \textit{45}(9), 4466–4474.

Silvern, R. F., Jacob, D. J., Mickley, L. J., Sulprizio, M. P., Travis, K. R., Marais, E. A., \ldots others (2019). Using satellite observations of tropospheric no\textsubscript{2} columns to infer long-term trends in us no\textsubscript{x} emissions: the importance of accounting for the free tropospheric no\textsubscript{2} background. \textit{Atmospheric Chemistry and Physics}, \textit{19}(13), 8863–8878.

Simpson, D. (1995). Biogenic emissions in europe: 2. implications for ozone control strategies. \textit{Journal of Geophysical Research: Atmospheres}, \textit{100}(D11), 22891–22906.

Skiba, U., Medinets, S., Cardenas, L. M., Carnell, E. J., Hutchings, N., \& Amon, B. (2021). Assessing the contribution of soil no\textsubscript{x} emissions to european atmospheric pollution. \textit{Environmental Research Letters}, \textit{16}(2), 025009.

Solberg, S., Walker, S.-E., Schneider, P., \& Guerreiro, C. (2021). Quantifying the impact of the covid-19 lockdown measures on nitrogen dioxide levels throughout europe. \textit{Atmosphere}, \textit{12}(2), 131.
Tai, A. P., Mickley, L. J., & Jacob, D. J. (2012). Impact of 2000–2050 climate change on fine particulate matter (PM2.5) air quality inferred from a multimodel analysis of meteorological modes. *Atmospheric Chemistry and Physics, 12*(23), 11329–11337.

Travis, K. R., Jacob, D. J., Fisher, J. A., Kim, P. S., Marais, E. A., Zhu, L., . . . others (2016). Why do models overestimate surface ozone in the southeast United States? *Atmospheric Chemistry and Physics, 16*(21), 13561–13577.

Turner, A. J., Kim, J., Fitzmaurice, H., Newman, C., Worthington, K., Chan, K., . . . Cohen, R. C. (2020). Observed impacts of COVID-19 on urban CO2 emissions. *Geophysical Research Letters, 47*(22), e2020GL090037.

Umweltbundesamt. (2020). Federal Environment Agency: current air data. [https://www.umweltbundesamt.de/en/data](https://www.umweltbundesamt.de/en/data).

Van Geffen, J., Boersma, K. F., Eskes, H., Sneep, M., Ter Linden, M., Zara, M., & Veeckind, J. P. (2020). 5p tropomi no2 slant column retrieval: method, stability, uncertainties and comparisons with omi. *Atmospheric Measurement Techniques, 13*(3), 1315–1335.

Vautard, R., Beekmann, M., Desplat, J., Hodzic, A., & Morel, S. (2007). Air quality in Europe during the summer of 2003 as a prototype of air quality in a warmer climate. *Comptes Rendus Geoscience, 339*(11-12), 747–763.

Venter, Z. S., Annan, K., Chowdhury, S., & Lelieveld, J. (2020). COVID-19 lockdowns cause global air pollution declines. *Proceedings of the National Academy of Sciences, 117*(32), 18984–18990.

Viatte, C., Wang, T., Damme, M. V., Dammers, E., Meleux, F., Clarisse, L., . . . others (2020). Atmospheric ammonia variability and link with particulate matter formation: a case study over the Paris area. *Atmospheric Chemistry and Physics, 20*(1), 577–596.

Voiculescu, M., Constantin, D.-E., Condurache-Bota, S., Câlnic, V., Rosu, A., & Dragomir Balanica, C. M. (2020). Role of meteorological parameters in the diurnal and seasonal variation of NO2 in a Romanian urban environment. *International Journal of Environmental Research and Public Health, 17*(17), 6228.

Wang, Y., Hu, B., Ji, D., Liu, Z., Tang, G., Xin, J., . . . others (2014). Ozone weekend effects in the Beijing–Tianjin–Hebei metropolitan area, China. *Atmospheric Chemistry and Physics, 14*(5), 2419–2429.

Weber, J., Shin, Y. M., Staunton Sykes, J., Archer-Nicholls, S., Abraham, N. L., & Archibald, A. T. (2020). Minimal climate impacts from short-lived climate forcers following emission reductions related to the COVID-19 pandemic. *Geophysical Research Letters, 47*(20), e2020GL090326.

Werf, G. R., Randerson, J. T., Giglio, L., Leeuwen, T. T. v., Chen, Y., Rogers, B. M., . . . others (2017). Global fire emissions estimates during 1997–2016. *Earth System Science Data, 9*(2), 697–720.

Zhang, L., Jacob, D. J., Knipping, E. M., Kumar, N., Munger, J. W., Carouge, C., . . . Chen, D. (2012). Nitrogen deposition to the United States: distribution, sources, and processes. *Atmospheric Chemistry and Physics, 12*(10), 4539–4554.

Zhang, R., Zhang, Y., Lin, H., Feng, X., Fu, T.-M., & Wang, Y. (2020). NOx emission reduction and recovery during COVID-19 in East China. *Atmosphere, 11*(4), 433.