Tropospheric NO_2 and O_3 response to COVID-19 lockdown restrictions at the national and urban scales in Germany

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

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

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

Corresponding author: Vigneskumar 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_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 (Evangeliou 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); Deroubaix et al. (2021), compared lockdown period concentration with long-term mean to estimate lockdown 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., 2020; 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 (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; 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 (Supplement 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 $3.6*10^{14}$ to $3.7*10^{16}$ 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 $\pm$ 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 NH3* (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 NH3*, 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 meteorological 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_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_X emissions (Hudman et al., 2012). In all comparisons of the GC model to TROPOMI, GC NO_2 vertical profile simulations (at 47 vertical layer) are converted to NO_2 column densities for TROPOMI footprints by interpolating into TROPOMI measurements pressure levels and applying TROPOMI's averaging kernals. Similar to above, GC column densities were gridded in 0.25*0.25-degree bins at the national scale.

Our methodology to obtain NO_2 and O_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_2, O_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_2 and O_3 between 2020 and 2019 are solely due to meteorological influences, e.g., differences in wind speed, boundary layer height, photo-chemistry etc.:

$$\Delta NO_{2(GC)} = NO_{2(GC,2020)} - NO_{2(GC,2019)} \tag{1}$$

$$\Delta O_{3(GC)} = O_{3(GC,2020)} - O_{3(GC,2019)} \tag{2}$$

The difference between the 2020 and 2019 NO_2 and O_3 observations for specific time periods include influence from both meteorological and emissions changes:

$$\Delta NO_{2(obs)} = NO_{2(obs,2020)} - NO_{2(obs,2019)} \tag{3}$$

$$\Delta O_{3(obs)} = O_{3(obs,2020)} - O_{3(obs,2019)} \tag{4}$$

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),

$$\Delta NO_{2(acc)} = \Delta NO_{2(obs)} - \Delta NO_{2(GC)} \tag{5}$$

and similarly for ozone:

$$\Delta O_{3(acc)} = \Delta O_{3(obs)} - \Delta O_{3(GC)} \tag{6}$$

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 NO_2 and 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_2 and O_3 concentrations and local meteorological parameters to find the dependency of NO_2 and O_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_2 concentrations have a negative correlation with wind speed, temperature and

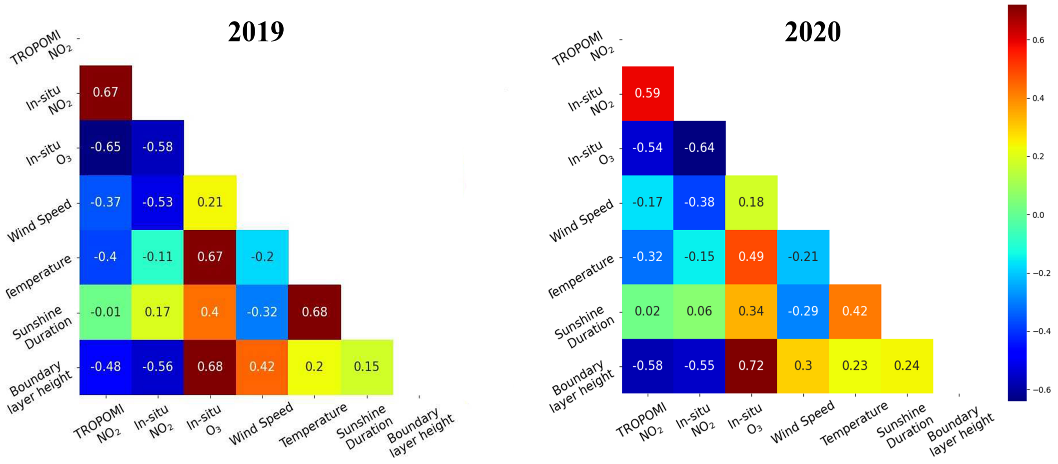

Figure 1. Correlation matrix (R-correlation coefficient) between meteorological parameters and NO_2 and O_3 concentrations (January to June in 2020 and 2019) in Munich.

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, lower NO_2 values are observed com-

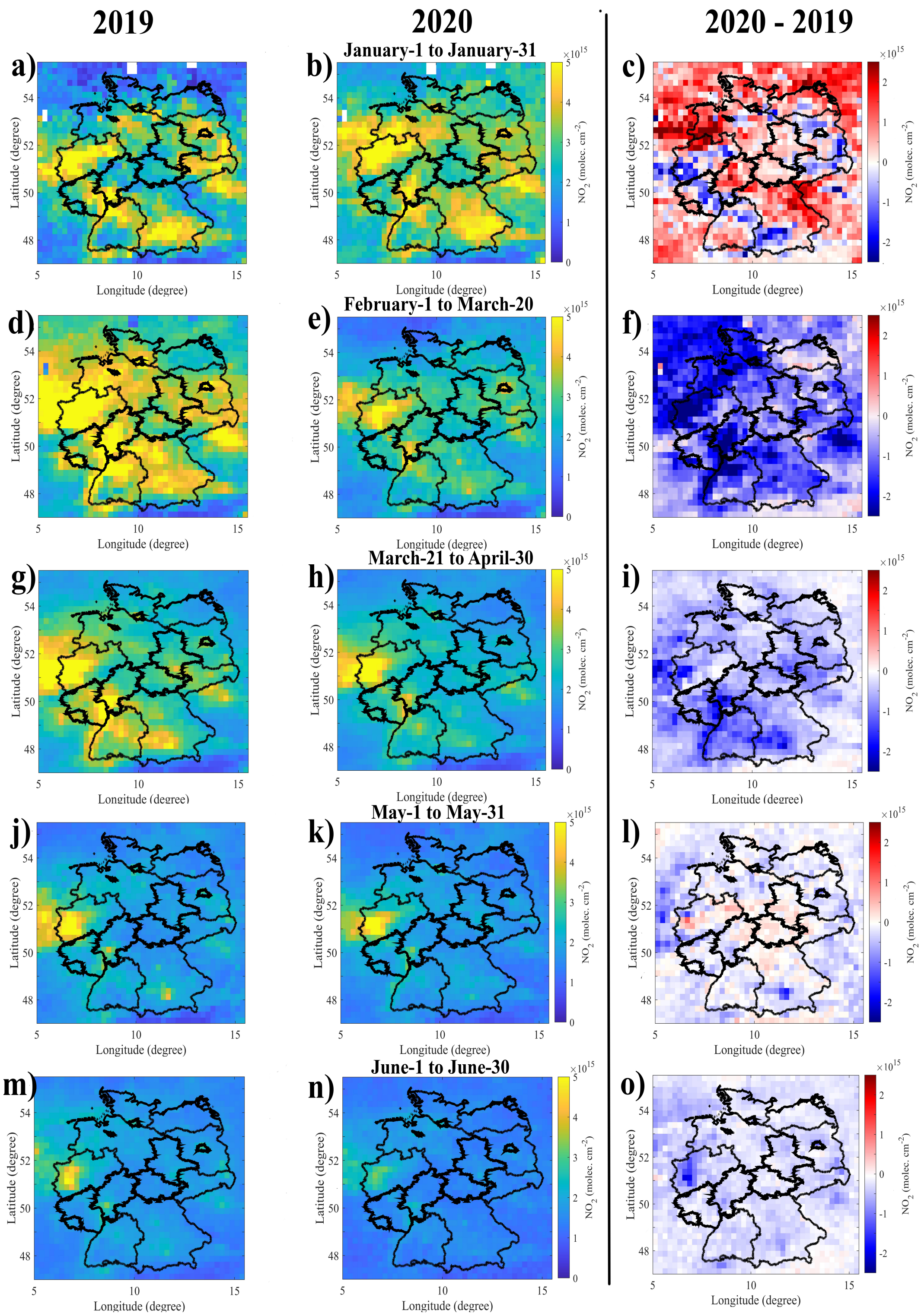

Figure 2. Mean TROPOMI tropospheric NO_2 column densities for 2019 (first column) and 2020 (second column). The absolute differences in TROPOMI tropospheric NO_2 column densities between 2020 and 2019 (third column).

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); Ordóñez 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-

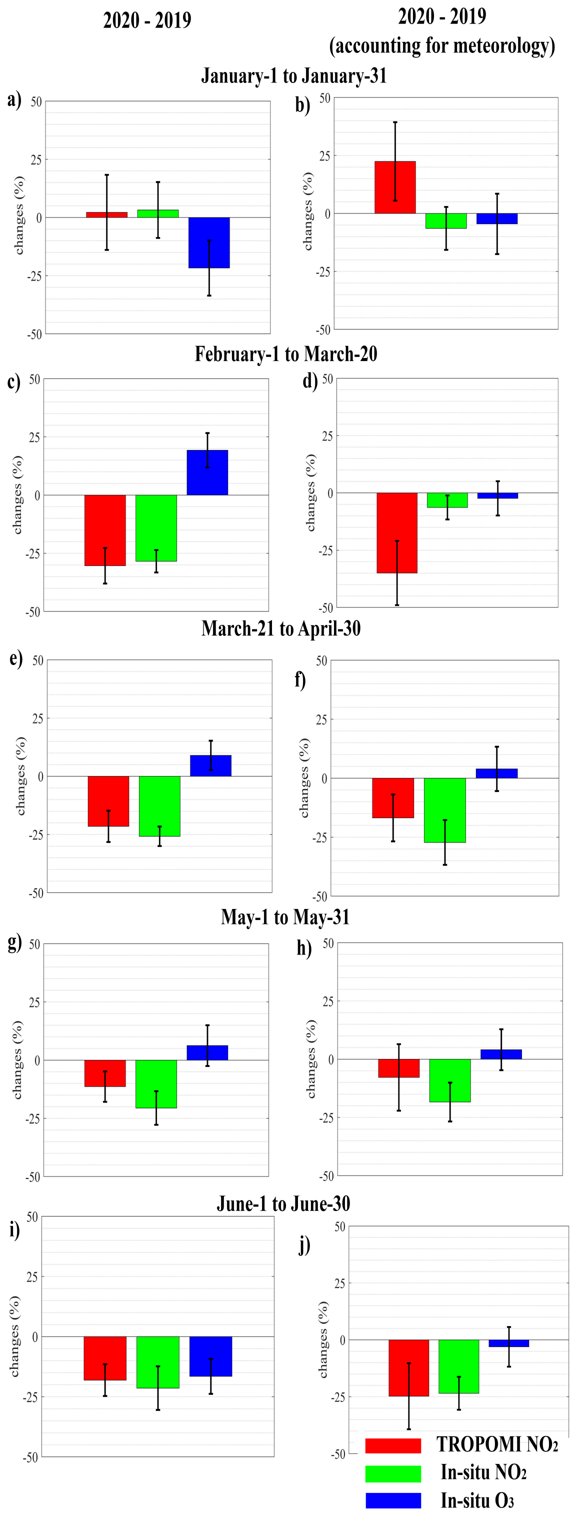

Figure 3. Mean relative changes in meteorological impacts unaccounted (left column) and accounted (right column) NO_2 and O_3 concentrations in eight metropolitan cities between 2020 and 2019. Error bars represent the 1 σ (standard deviation) of mean of eight metropolitan cities.

trations between February 1 and May 31, 2020 (consistent with Gaubert et al. (2021)), while meteorological conditions were favorable for low O_3 concentrations in January and June 2020. For instance, before accounting for meteorology, mean O_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 (j)). After accounting for meteorology, the difference between mean O_3 concentrations in June 2020 and the same period in 2019 is reduced to -3 %. Meteorology had a different impact on NO_2 and O_3 levels and this impact also varied for different time periods. This demonstrates the complex relationship between O_3, NO_2, and meteorological conditions.

We found large discrepancies between in-situ and TROPOMI NO_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_2 and TROPOMI NO_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_2 to the TROPOMI NO_2 observation. Second, we compare the in-situ NO_2 at the time of TROPOMI overpass with the TROPOMI NO_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_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 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 NO_2 background on satellite column measurements to infer the changes in surface NO_X emission. The primary sources of background NO_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_X emissions at elevated temperatures (Butterbach-Bahl et al., 2001) (discussed below). In addition, subtracting the contribution of the NO_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_2 emissions. In contrast to satellite column measurements, background NO_2 has little influence (5-10 %) on in-situ surface NO_2 concentrations (R. F. Silvern et al., 2019). The discrepancies between in-situ and TROPOMI changes primarily results from unaccounted background NO_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_X emission changes.

The NO_2 column densities in rural locations were also investigated. During the 2020 stricter lockdown period, after accounting for meteorology, slightly increased NO_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_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_X emissions typically increase with temperature (Oikawa et al., 2015). Soil NO_X emissions are expected to be high in the early spring (stricter lockdown period), even though the average temperature in May

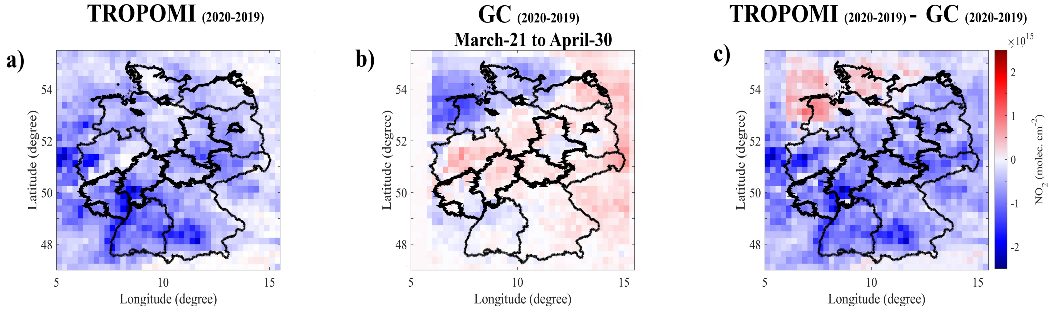

Figure 4. The absolute difference in TROPOMI (a) and GEOS-Chem (b) NO_2 column densities between 2020 and 2019 (stricter lockdown period: March 21 to April 30). The absolute difference between first two columns is shown in panel (c).

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_2O_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-

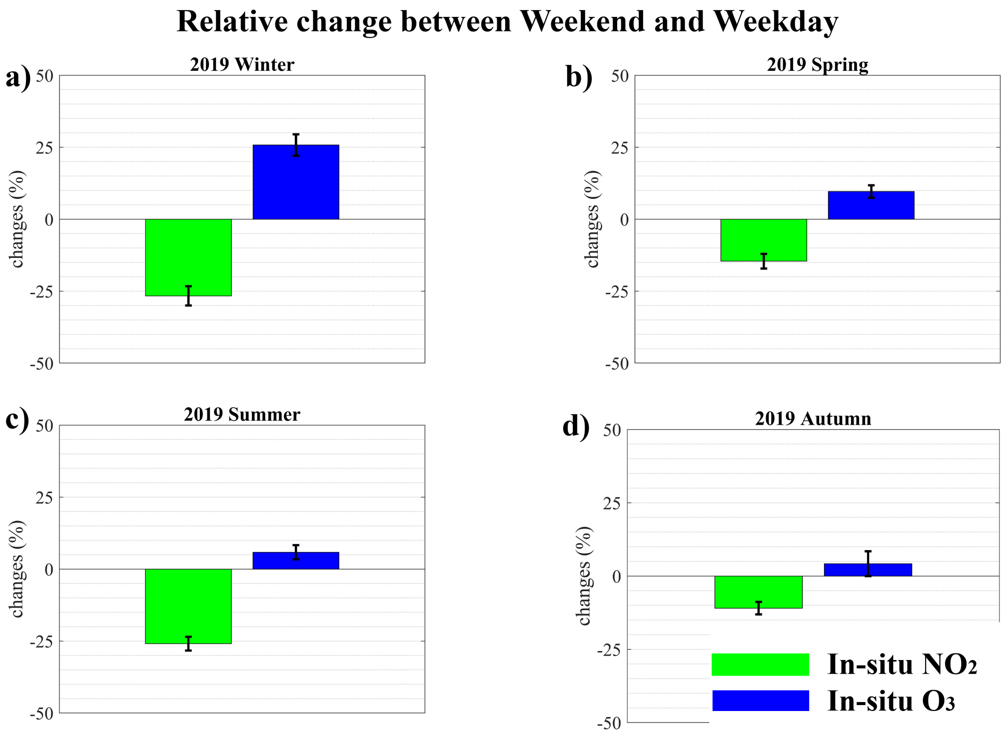

Figure 5. Mean relative change in in-situ NO_2 and O_3 concentrations in Munich between weekends and weekdays. Error bars represent statistical uncertainty (1 σ) in the calculation of relative change between weekend and weekday means.

counted for mean in-situ O_3 concentrations in spring (March 21 and May 31, 2020) and a slight decrease (3 %) in meteorology accounted for mean in-situ O_3 concentrations in early summer (June, 2020) compared to the same period in 2019. This implies that the degree of NO_X saturation of ozone production is weakening from winter to summer (consistent with weekend effects and Jin et al. (2017); Kang et al. (2021)). During the lockdown period, the daily maximum 8-hour mean O_3 concentration in metropolitan areas also exceeded the EU target value (120 $\mu g/m^3$) (2 days in Munich, Berlin, Cologne, Stuttgart metropolitan areas). These exceedances are more attributable to NO_X saturated conditions than to meteorology.

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 hypothesise 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 no_2 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 $pm_{2.5}$ 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 no_x conditions–a modelling study. *Atmospheric Chemistry and Physics*, *16*(18), 11601–11615.

Collivignarelli, M. C., Abbà, A., Bertanza, G., Pedrazzani, R., Ricciardi, P., & Miino, 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., Menut, 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., Katsouyanni, 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.

Dlr covid-19. (2020). 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), 2795–2804.

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), e2020JD034213.

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 co_2 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 no_2 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 h_2o_2/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., Janssens-Maenhout, G., Pitkanen, 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 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., Lamsal, L. N., Duncan, B., ... others (2017). Evaluating a space-based indicator of surface ozone-no_x-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 $pm_{2.5}$ 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., Lucchesi, 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 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). Tno-macc_ii emission inventory; a multi-year (2003–2009) consistent high-resolution european emission inventory for air quality modelling. *Atmos. Chem. Phys*, *14*(20), 10963–10976.

Kuttippurath, 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 co_2 emissions during the covid-19 forced confinement. *Nature Climate Change*, 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+ n_2o emissions. *Global Change Biology*, *23*(6), 2520–2532.

Liu, Y., Wang, T., Stavrakou, 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). Near-real-time monitoring of global co_2 emissions reveals the effects of the covid-19 pandemic. *Nature communications*, *11*(1), 1–12.

Liu, Z., Deng, Z., Ciais, P., Lei, R., Davis, S. J., Feng, S., ... others (2020). Covid-19 causes record decline in global co_2 emissions.

Lorente, A., Boersma, K., Eskes, H., Veefkind, J., Van Geffen, J., de Zeeuw, M., ... Krol, M. (2019). Quantification of nitrogen oxides emissions from build-up of pollution over paris with tropomi. *Scientific reports*, *9*(1), 1–10.

Lorente, A., Folkert Boersma, K., Yu, H., Dörner, S., Hilboll, A., Richter, A., ... others (2017). Structural uncertainty in air mass factor calculation for no 2 and hcho satellite retrievals. *Atmospheric Measurement Techniques*, *10*(3), 759–782.

Marais, E. A., Jacob, D. J., Choi, S., Joiner, J., Belmonte-Rivas, M., Cohen, R. C., ... others (2018). Nitrogen oxides in the global upper troposphere: interpreting cloud-sliced no_2 observations from the omi satellite instrument. *Atmospheric Chemistry and Physics*, *18*(23), 17017–17027.

Marais, E. A., Roberts, J. F., Ryan, R. G., Eskes, H., Boersma, K. F., Choi, S., ... others (2021). New observations of no_2 in the upper troposphere from tropomi. *Atmospheric Measurement Techniques*, *14*(3), 2389–2408.

Martin, R. V., Fiore, A. M., & Van Donkelaar, A. (2004). Space-based diagnosis of surface ozone sensitivity to anthropogenic emissions. *Geophysical Research Letters*, *31*(6).

Meehl, G. A., & Tebaldi, C. (2004). More intense, more frequent, and longer lasting heat waves in the 21st century. *Science*, *305*(5686), 994–997.

Meleux, F., Solmon, F., & Giorgi, F. (2007). Increase in summer european ozone amounts due to climate change. *Atmospheric environment*, *41*(35), 7577–7587.

Melkonyan, A., & Wagner, P. (2013). Ozone and its projection in regard to climate change. *Atmospheric Environment*, *67*, 287–295.

Menut, L., Bessagnet, B., Siour, G., Mailler, S., Pennel, R., & Cholakian, A. (2020). Impact of lockdown measures to combat covid-19 on air quality over western europe. *Science of the Total Environment*, *741*, 140426.

Merra-2. (2020). Modern-Era Retrospective analysis for Research and Applications, Version 2. https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/data_access/.

Mertens, M., Jöckel, P., Matthes, S., Nützel, M., Grewe, V., & Sausen, R. (2021). Covid-19 induced lower-tropospheric ozone changes. *Environmental Research Letters*, *16*(6), 064005.

Oikawa, P., Ge, C., Wang, J., Eberwein, J., Liang, L., Allsman, L., ... Jenerette, G. (2015). Unusually high soil nitrogen oxide emissions influence air quality in a high-temperature agricultural region. *Nature communications*, *6*(1), 1–10.

Ordóñez, C., Garrido-Perez, J. M., & García-Herrera, R. (2020). Early spring near-surface ozone in europe during the covid-19 shutdown: Meteorological effects outweigh emission changes. *Science of the Total Environment*, *747*, 141322.

Petetin, H., Bowdalo, D., Soret, A., Guevara, M., Jorba, O., Serradell, K., & Pérez García-Pando, C. (2020). Meteorology-normalized impact of the covid-19 lockdown upon no_2 pollution in spain. *Atmospheric Chemistry and Physics*, *20*(18), 11119–11141.

Potts, D. A., Marais, E. A., Boesch, H., Pope, R. J., Lee, J., Drysdale, W., ... others (2021). Diagnosing air quality changes in the uk during the covid-19 lockdown using tropomi and geos-chem. *Environmental Research Letters*, *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_2 in inferring no_x emissions from satellite no_2 observations. *Geophysical Research Letters*, e2021GL092783.

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

S5p no2 readme. (2020). S5P Mission Performance Centre Nitrogen Dioxide [L2 NO2] Readme. `https://sentinel.esa.int/documents/247904/3541451/Sentinel-5P-Nitrogen-Dioxide-Level-2-Product-Readme-File`.

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

Sharma, S., Zhang, M., Gao, J., Zhang, H., Kota, S. H., et al. (2020). Effect of restricted emissions during covid-19 on air quality in india. *Science of the Total Environment*, *728*, 138878.

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

Sillman, S. (1999). The relation between ozone, nox and hydrocarbons in urban and polluted rural environments. *Atmospheric Environment*, *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. *Journal of Geophysical Research: Atmospheres*, *95*(D2), 1837–1851.

Sillman, S., Vautard, R., Menut, L., & Kley, D. (2003). O_3-no_x-voc sensitivity and no_x-voc indicators in paris: Results from models and atmospheric pollution over the paris area (esquif) measurements. *Journal of Geophysical Research: Atmospheres*, *108*(D17).

Silvern, R., Jacob, D., Travis, K., Sherwen, T., Evans, M., Cohen, R., ... others (2018). Observed no/no_2 ratios in the upper troposphere imply errors in no-no_2-o_3 cycling kinetics or an unaccounted nox reservoir. *Geophysical Research Letters*, *45*(9), 4466–4474.

Silvern, R. F., Jacob, D. J., Mickley, L. J., Sulprizio, M. P., Travis, K. R., Marais, E. A., ... others (2019). Using satellite observations of tropospheric no_2 columns to infer long-term trends in us no_x emissions: the importance of accounting for the free tropospheric no_2 background. *Atmospheric Chemistry and Physics*, *19*(13), 8863–8878.

Simpson, D. (1995). Biogenic emissions in europe: 2. implications for ozone control strategies. *Journal of Geophysical Research: Atmospheres*, *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_x emissions to european atmospheric pollution. *Environmental Research Letters*, *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. *Atmosphere*, *12*(2), 131.

Tai, A. P., Mickley, L. J., & Jacob, D. J. (2012). Impact of 2000–2050 climate change on fine particulate matter ($pm_{2.5}$) air quality inferred from a multi-model 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 co_2 emissions. *Geophysical Research Letters*, *47*(22), e2020GL090037.

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

Van Geffen, J., Boersma, K. F., Eskes, H., Sneep, M., Ter Linden, M., Zara, M., & Veefkind, J. P. (2020). S5p tropomi no_2 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., Aunan, 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., Călmuc, V., Roșu, A., & Dragomir Bălănică, C. M. (2020). Role of meteorological parameters in the diurnal and seasonal variation of no_2 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). No_x emission reduction and recovery during covid-19 in east china. *Atmosphere*, *11*(4), 433.