Secondary PM decreases significantly less than NO₂ emission reductions during COVID lockdown in Germany

Vigneshkumar Balamurugan¹, Jia Chen¹, Zhen Qu², Xiao Bi¹, and Frank N. Keutsch²,³
¹Environmental Sensing and Modeling, Technical University of Munich (TUM), Munich, Germany
²School of Engineering and Applied Science, Harvard University, Cambridge, MA, USA
³Department of Chemistry and Chemical Biology, Harvard University, Cambridge, MA, USA

Correspondence: Vigneshkumar Balamurugan (vigneshkumar.balamurugan@tum.de), Jia Chen (jia.chen@tum.de)

Abstract. This study estimates the influence of anthropogenic emission reductions on the concentration of particulate matter with a diameter smaller than 2.5 µm (PM₂.₅) during the 2020 lockdown period in German metropolitan areas. After accounting for meteorological effects, PM₂.₅ concentrations during the spring 2020 lockdown period were 5% lower compared to the same time period in 2019. However, during the 2020 pre-lockdown period (winter), meteorology accounted for PM₂.₅ concentrations were 19% lower than in 2019. Meanwhile, meteorology accounted for NO₂ concentrations dropped by 23% during the 2020 lockdown period compared to an only 9% drop for the 2020 pre-lockdown period, both compared to 2019. Meteorology accounted for SO₂ and CO concentrations show no significant changes during the 2020 lockdown period compared to 2019. GEOS-Chem (GC) simulation with a COVID-19 emission reduction scenario based on the observations (23% reduction in NOₓ emission with unchanged VOC and SO₂) are consistent with the small reductions of PM₂.₅ during the lockdown and are used to identify the underlying drivers for this. Due to being in a NOₓ saturated ozone production regime, GC OH radical and O₃ concentrations increased (15 and 9%, respectively) during the lockdown compared to a Business As Usual (no lockdown) scenario. The increased O₃ results in increased NO₃ radical concentrations, primarily during the night, despite the large reductions in NO₂. Thus, the oxidative capacity of the atmosphere is increased in all three important oxidants, OH, O₃, and NO₃. PM nitrate formation from gas-phase nitric acid (HNO₃) is decreased during the lockdown as the increased OH concentration cannot compensate for the strong reductions in NO₂ resulting in decreased day-time HNO₃ formation from the OH + NO₂ reaction. However, night-time formation of PM nitrate from N₂O₅ hydrolysis is relatively unchanged. This results from the fact that increased night-time O₃ results in significantly increased NO₃ which roughly balances the effect of the strong NO₂ reductions on N₂O₅ formation. Ultimately, the only small observed decrease in lockdown PM₂.₅ concentrations can be explained by the large contribution of night-time PM nitrate formation, generally enhanced sulfate formation and slightly decreased ammonium. This study also suggests that high PM₂.₅ episodes in early spring are linked to high atmospheric ammonia concentrations combined with favorable meteorological conditions of low temperature and low boundary layer height. North-West Germany is a hot-spot of NH₃ emissions, primarily emitted from livestock farming and intensive agricultural activities (fertilizer application), with high NH₃ concentrations in the early spring and summer months. Based on our findings, we suggest that appropriate NOₓ and VOC emission controls are required to limit ozone, and that should also

https://doi.org/10.5194/acp-2022-87
Preprint. Discussion started: 16 February 2022
© Author(s) 2022. CC BY 4.0 License.
help reduce PM$_{2.5}$. Regulation of NH$_3$ emissions, primarily from agricultural sectors, could result in significant reductions in PM$_{2.5}$ pollution.

1 Introduction

To halt the spread of the COVID-19 virus, various strict measures such as social isolation, curfews, and travel restrictions were implemented around the world in early 2020 (Steinmetz et al., 2020). As a result of these restrictions, anthropogenic emissions decreased significantly (Schumann et al., 2021; Le Quéré et al., 2020; Turner et al., 2020). Reduced primary emission activities from road transportation and industrial activities were expected to improve air quality. Numerous studies using satellite and in-situ measurements have reported significant reductions in primary air pollutant concentrations during the COVID-19 lockdown period compared to pre-lockdown period in various parts of the world (Bauwens et al., 2020; Biswal et al., 2020; Collivignarelli et al., 2020; Dietrich et al., 2021; Field et al., 2020; He et al., 2021; Pathakoti et al., 2020; Mendez-Espinosa et al., 2020), but also emphasize the importance of accounting for the effects of different meteorological conditions between the study period and the reference period (Barré et al., 2020; Grange et al., 2020; Kroll et al., 2020; Koukouli et al., 2021; Ordóñez et al., 2020; Solberg et al., 2021). Anomalies in air pollutant concentrations caused by changes in meteorological conditions were also separated from observed changes using modeling work to estimate the actual influence of COVID-19 lockdown restrictions on air pollutant concentration changes (Balamurugan et al., 2021; Goldberg et al., 2020; Kang et al., 2020; Petetin et al., 2020; Qu et al., 2021; Yin et al., 2021). Secondary pollutant concentrations (O$_3$ and PM$_{2.5}$), which are primarily produced by precursor gases through complex atmospheric chemical reactions, remarkably increased or did not reduce commensurate to precursor emission reductions seen in some parts of the world during the COVID-19 lockdown period (Campbell et al., 2021; Deroubaix et al., 2021; He et al., 2021; Huang et al., 2021; Keller et al., 2021; Lee et al., 2020; Putaud et al., 2021; Souri et al., 2021; Wang et al., 2020, 2021).

Particulate Matter (PM) is the sum of all particles (solid and liquid) suspended in air, and can be classified based on aerodynamic behavior, i.e., aerodynamic diameter (AD). Particles with an AD smaller than 10 µm are referred to as PM$_{10}$, while particles smaller than 2.5 µm AD are referred to as PM$_{2.5}$. Understanding of seasonal and inter-annual variability of PM, particularly over urban areas, remains a challenge (Fuzzi et al., 2015). This is mainly due to a lack of understanding in the attribution of PM sources. PM sources include both direct/primary sources (vehicle and industrial emissions, wind-blown dust, pollen, wildfires, etc.) as well as secondary formation (gas-to-particle conversion process) via atmospheric chemical reaction of precursor compounds such as NO$_X$, SO$_2$, NH$_3$, VOCs and other organic compounds, including compounds that have partitioned from primary aerosol back to the gas-phase, followed by partitioning to the condensed phase (Allen et al., 2015; Ayres et al., 2015; Fisher et al., 2016; Hallquist et al., 2009; Jacob, 1999; Jacobson, 1999; Marais et al., 2016; Seinfeld and Pankow, 2003; Steinfeld, 1998; Zhang et al., 2015). The composition of PM thus varies greatly depending on time and location; for example, in urban areas nitrate and organic aerosol often dominate in winter time (Cesari et al., 2018; Zhai et al., 2021).

In this study, we mainly focus on the response of urban surface PM$_{2.5}$ to COVID-19 lockdown restrictions in Germany. Because major anthropogenic emissions are reduced, this unplanned intervention can test the understanding of the contribu-
tion of secondary PM$_{2.5}$ sources, as well as the processes important in secondary PM$_{2.5}$ formation. Despite of significant reductions in some anthropogenic activities, natural and agricultural air pollutant sources were not affected by the COVID-19 lockdown measures. Ammonia (NH$_3$) emissions (agricultural sources) are a significant source of PM$_{2.5}$ in Germany in the spring (Fortems-Chiney et al., 2016), when lockdown restrictions are implemented. Secondary inorganic aerosols such as ammonium sulfate and ammonium nitrate are the largest contributors to PM$_{2.5}$ in Europe (Pay et al., 2012; Petetin et al., 2016). In comparison to sulfate formation, nitrate formation is more dependent on NH$_3$ concentration (Erisman and Schaap, 2004; Sharma et al., 2007; Wu et al., 2008). In the winter and spring (low temperature and high relative humidity), the role of NH$_3$ in PM$_{2.5}$ formation is greater than in the summer (high temperature and low relative humidity) (Schiferl et al., 2016; Squizzato et al., 2013; Viatte et al., 2020). Primary components of PM$_{2.5}$ are directly proportional to primary emission but secondary components of PM$_{2.5}$ are not directly proportional to secondary precursor emissions or concentrations as they are produced by non-linear complex atmospheric chemical reactions (Shah et al., 2018). Observational and modeling evidence is required to estimate the influence of change in precursor emissions on PM$_{2.5}$ concentrations. To this end, we used ground and space-based measurements of PM$_{2.5}$, NO$_2$, O$_3$, SO$_2$, CO and NH$_3$ in conjunction with GEOS-Chem simulations to investigate the influence of lockdown restrictions on PM$_{2.5}$ concentrations.

Modelling studies such as Gaubert et al. (2021); Hammer et al. (2021); Matthias et al. (2021); Menut et al. (2020) have already reported the PM$_{2.5}$ changes across Europe including Germany, during the COVID-19 lockdown period. The activity data (e.g., transportation, industrial activities and energy production) were used in the above mentioned studies to create a COVID-19 emission reduction scenario (Doumbia et al., 2021; Guevara et al., 2021). However, there are large discrepancies between various activity data sets (Gensheimer et al., 2021), necessitating different approaches to estimating the actual emission reduction caused by the COVID-19 lockdown restrictions. In this study, GEOS-Chem simulations (using identical anthropogenic emission for 2020 and 2019) were used to estimate the meteorology accounted for observed pollutant concentrations changes between 2020 and 2019, which were then used as a proxy for emissions reductions caused by COVID-19 lockdown measures to create a COVID-19 emission scenario in GEOS-Chem model for simulating the lockdown pollutant concentrations (Fig. 1). In addition to looking at the impact of lockdown restrictions on air pollutant concentrations (Sect. 4.1), we focus on process level analysis of the impact of changes in precursor emissions (NO$_X$) on PM$_{2.5}$ formation (Sect. 4.2), as well as the role of ammonia (NH$_3$) emissions in PM$_{2.5}$ formation (Sect. 4.3).

2 Data and Model

Data sets used in this study are summarized in Table 1. We focused on ten metropolitan areas in Germany (Bremen, Cologne, Dresden, Dusseldorf, Frankfurt, Hamburg, Hanover, Leipzig, Munich and Stuttgart) and used surface air pollutant concentration data (PM$_{2.5}$, NO$_2$, O$_3$) for all of these while SO$_2$ data was only available for five of these areas (Bremen, Dresden, Frankfurt, Hamburg and Leipzig) and CO data was limited to six metropolitan areas (Bremen, Frankfurt, Hamburg, Hanover, Munich and Stuttgart). We use data for 2019 and 2020 in this work (data-obtained from https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm).
Table 1. Data sets used in this study.

| Data source                              | Data                        | Temporal resolution | Spatial resolution | Data availability                                                        |
|------------------------------------------|-----------------------------|---------------------|--------------------|-------------------------------------------------------------------------|
| Governmental in-situ measurements        | NO\textsubscript{2}, O\textsubscript{3}, PM\textsubscript{2.5} | 1 h                 |                    | Bremen, Cologne, Dresden, Dusseldorf, Frankfurt, Hamburg, Hanover, Leipzig, Munich and Stuttgart metropolitan areas |
|                                          | SO\textsubscript{2}         | 1 h                 |                    | Bremen, Dresden, Frankfurt, Hamburg and Leipzig metropolitan areas       |
|                                          | CO                          | 1 h                 |                    |                           |
| TROPOMI satellite measurements           | SO\textsubscript{2}         | daily               | 7*3.5 km           | Bremen, Frankfurt, Hamburg, Hanover, Munich and Stuttgart metropolitan areas |
|                                          |                             |                     | (5.5*3.5 km, after August 6, 2019) |                           |
| IASI satellite measurements              | NH\textsubscript{3}        | twice a day         | 12 km diameter     | All of Germany                                                          |
| ERA 5 (ECMWF reanalysis)                | Temperature, relative humidity, boundary layer height and wind speed | monthly          | 1 degree            | All of Germany                                                          |
|                                          | Precipitation               | daily               | 1 degree            | All of Germany                                                          |
| GEOS-Chem (GC) chemical transport model  | All species                 | 1 h                 | 0.5 * 0.625 degree | All of Germany                                                          |

TROPOMI tropospheric SO\textsubscript{2} (Theys et al., 2017) column products are also used (offline products-obtained from https://s5phub.copernicus.eu). The TROPOMI SO\textsubscript{2} product provides the total SO\textsubscript{2} column between the surface and the top of troposphere. The TROPOMI overpass occurs around 13.30 local time. At the start of the mission, the TROPOMI product provided data at a resolution of 7*3.5 km, while after August 6, 2019 the resolution improved to 5.5*3.5 km. Stricter quality filtering criteria (quality assurance value (qa) >= 0.5) was applied to the dataset. A daily mean of SO\textsubscript{2} is calculated by averaging these values within 0.5-degree radius of the urban center.

The daily atmospheric NH\textsubscript{3} variability in Germany was studied using the “near-real time daily IASI/Metop-B ammonia (NH\textsubscript{3}) total column (ANNI-NH\textsubscript{3}-v3)” dataset (products-obtained from https://iasi.aeris-data.fr/catalog/). The data used are from the IASI instrument aboard the Metop-B satellite, which has a local solar overpass time of 9:30 a.m and 9:30 p.m (Clerbaux et al., 2009). We only used day-time (9.30 am) measurements in this study. Night-time measurements (9.30 pm) were excluded due to their large relative errors. A daily mean is calculated by averaging the values within 0.5-degree radius of the urban center. The monthly atmospheric NH\textsubscript{3} variability in Germany was studied using the “standard monthly IASI/Metop-B ULB-LATMOS ammonia (NH\textsubscript{3}) L3 product (total column)” dataset. This product contains a monthly averaged NH\textsubscript{3} total column with a spatial resolution of 1*1 degree (products-obtained from https://iasi.aeris-data.fr/catalog/).

Temperature, relative humidity, boundary layer height and wind information are obtained from the ERA 5 product (Hersbach et al., 2020). This product’s native spatial and temporal resolutions are 0.25 degree and 1 hour, respectively. For precipitation information, the GPCP daily gridded product from ERA 5 is used, which provides global gridded data at 1-degree resolution (products-obtained from https://cds.climate.copernicus.eu/).

We used the GEOS-Chem (GC) chemical transport model (http://doi.org/10.5281/zenodo.3959279) to simulate the pollutant concentration for 2020 and 2019. The GC simulation conducted over Germany (4-17°E, 45-57°N) had a horizontal resolution of 0.5°*0.625° with dynamic boundary conditions generated from a global simulation with 4°*5° resolution. We ran the GC simulation for two cases. In the first case, anthropogenic emissions from the 2014 CEDS inventory (Hoesly et al., 2018)
Figure 1. Schematic diagram of our methodology for calculating the meteorology accounted for observed pollutant concentration changes between 2020 and 2019, and emission accounted for GC pollutant concentrations changes between 2020 lockdown and 2020 BAU scenario.

are used in the GC simulations for both 2019 and 2020, but with the corresponding meteorology from MERRA-2 global reanalysis product for 2019 and 2020. Natural emissions from soil and lightning are calculated for the corresponding year using mechanisms described in Hudman et al. (2012) and Murray (2016). The corresponding year’s open fire emissions from GFED4 (Werf et al., 2017) are used for 2019 and 2020. In the second case, the anthropogenic emission inventory were scaled down by the estimated emissions reduction caused by the lockdown restrictions for the 2020 lockdown period. The remaining (natural and fire) emissions are calculated in the same way as in the first case.

3 Method

The following is our methodology for estimating meteorology accounted for observed pollutant concentration changes between 2020 and 2019, similar to Balamurugan et al. (2021); Qu et al. (2021). We estimate the difference in pollutant concentrations
between 2020 and 2019 caused by changes in meteorology using GC simulated concentrations (first case). Since GC uses identical anthropogenic emission for 2020 and 2019, with the corresponding year meteorology, the difference between 2020 and 2019 GC pollutant (e.g., PM$_{2.5}$) concentrations only results from meteorology changes between 2020 and 2019. We use $\Delta$ to signify absolute concentration change, and $f$ to signify fractional (percentage) change.

$$
\Delta PM_{2.5}(GC) = PM_{2.5}(GC,2020) - PM_{2.5}(GC,2019)
$$

(1)

The observed (ground-truth measurements) pollutant concentration changes between 2020 and 2019, which includes the effects of lockdown restrictions and meteorology, is:

$$
\Delta PM_{2.5}(obs) = PM_{2.5}(obs,2020) - PM_{2.5}(obs,2019)
$$

(2)

To disentangle the meteorology contribution from the observed pollutant concentration changes, we subtract the GC pollutant concentration changes caused by meteorology from observed pollutant concentration changes between 2020 and 2019.

$$
\Delta PM_{2.5}(obs,emi) = \Delta PM_{2.5}(obs) - \Delta PM_{2.5}(GC)
$$

(3)

The fractional change in meteorology accounted for pollutant concentration between 2020 and 2019, i.e., pollutant concentration changes between 2020 and 2019 due to emission changes only, is calculated as,

$$
f PM_{2.5}(obs,emi) = \frac{\Delta PM_{2.5}(obs,emi)}{PM_{2.5}(obs,2019)}
$$

(4)

where, “obs”, “GC” and “obs,emi” refer to ground-truth measurements, GEOS-Chem simulations and meteorology accounted for ground-truth measurements, respectively.

We estimate the meteorology accounted for fractional change in other pollutant concentrations analogously. Our previous study (Balamurugan et al., 2021), using the same methodology, reported the meteorology accounted for NO$_2$ and O$_3$ concentration changes for eight German metropolitan areas. Here, we reproduce the results for NO$_2$ and O$_3$ concentrations, but for ten metropolitan areas. We use $f NO_X(emi)$ and $f CO(emi)$ to capture fractional changes in anthropogenic NO$_X$ and VOC emission ($f NO_X(emi)$ and $f VOCA(emi)$) due to lock down restrictions, respectively. Because of the scarcity of VOC measurements, CO data was used as a proxy for anthropogenic VOC (Fujita et al., 2003; Jiménez et al., 2005; Stephens et al., 2008; Yarwood et al., 2003) and NO$_2$ was used as proxy for NO$_X$. This assumption is supported by studies such as Baker et al. (2008); Von Schneidemesser et al. (2010), which show anthropogenic VOC is well correlated with CO, and Blanchard and Tanenbaum (2003), which shows comparable changes in VOC and CO between weekday and weekend. Changes in biogenic VOCs are not directly affected by lockdown measures.

$$
f NO_X(emi) \approx f NO_2(emi)
$$

(5)

$$
f VOCA(emi) \approx f CO(emi)
$$

(6)
Figure 2. Meteorology accounted for mean in-situ PM$_{2.5}$, NO$_2$, and O$_3$ concentration changes between 2020 and 2019. Results of computations according to our first case ($fX_{(obs,emi)}$) in the Sect. 3. Error bars represent the 1 σ of mean of ten metropolitan areas.

The base anthropogenic emission inventory were then scaled down by $fNOX_{(emission)}$ and $fVOC_{(emission)}$ for NO$_X$ and VOC emission, respectively, in the GC model for the 2020 lockdown period (second case), which simulates all pollutants concentrations for the lockdown emission scenario. The fractional change in emission accounted for, i.e. using scaled emission inventories, GC pollutants level during the 2020 lockdown period compared to 2020 Business As Usual (BAU), i.e., no lockdown, level is calculated as,

$$fPM_{2.5(GC,emi)} = \frac{PM_{2.5(GC,2020,lock)} - PM_{2.5(GC,2020)}}{PM_{2.5(GC,2020)}}$$  \quad (7)$$

where, “GC,emi” refers to GC simulations accounting for scaled emission and PM$_{2.5(GC,2020,lock)}$ are the PM$_{2.5}$ concentrations during the lockdown period determined via the 2020 GC simulations with down-scaled emissions. We estimate the emission accounted for concentration changes of other pollutants in the same way. Figure 1 illustrates our methodology for calculating the meteorology accounted for observed pollutant concentrations changes between 2020 and 2019, as well as emission accounted for GC pollutant concentration changes between 2020 lockdown and 2020 BAU scenario.
4 Results and discussion

4.1 Influence of lockdown restrictions on the concentrations of air pollutants

To assess the impact of lockdown restrictions on the concentration of air pollutants, we compared the 2020 lockdown period pollutant concentrations to the same period in 2019. These comparison results, however, need to take the effects of both meteorological and lockdown restrictions into account. As mentioned in Sect. 3, we used GEOS-Chem simulations to disentangle the effects of meteorology on observed pollutant concentration changes between 2020 and 2019. Studies such as Balamurugan et al. (2021) and Tai et al. (2012) have shown that GEOS-Chem can reproduce the temporal variability of observed pollutant concentrations including PM$_{2.5}$, emphasizing that GC can be used for process level analysis of PM$_{2.5}$ variability. We also compared the 2019 GC and 2019 observed in-situ PM$_{2.5}$ concentrations and found that the GC and observed in-situ PM$_{2.5}$ concentrations were in good agreement (R > 0.5 for all metropolitan areas, except Leipzig which has a R value of 0.39) (e.g., Fig. 6 (c), for Cologne metropolitan area). The GC simulations underestimate the PM$_{2.5}$ when compared to observed in-situ PM$_{2.5}$ concentrations (mean bias (GC - in-situ) ranges from -12.7 % to -37.4 %), except for the Cologne metropolitan area (+11.7 %). However, since we use the GC’s relative difference between 2020 and 2019, this bias should cancel out.

Figure 2 shows meteorology accounted for mean PM$_{2.5}$, NO$_2$ and O$_3$ concentration changes between 2020 and 2019 for ten German metropolitan areas from January 1 through May 31. Both meteorology accounted and unaccounted for mean PM$_{2.5}$, NO$_2$ and O$_3$ concentration changes between 2020 and 2019 for ten German metropolitan areas are shown in Appendix Fig. A1. The German government imposed COVID-19 lockdown restrictions on March 21, 2020 in Germany. In figures and for specific cases, the pre-lockdown period (January 1 to March 20) is divided into two sections, and the lockdown period (March 21 to May 31) is also divided into two sections (unless otherwise specified): (a) January 1 to January 31, 2020 - No lockdown restrictions, (b) February 1 to March 20, 2020 - No lockdown restrictions in the event of unusual weather conditions (occurrence of storms), (c) March 21 to April 30, 2020 (spring) - Strict lockdown measures, and (d) May 1 to May 31, 2020 (late spring) - Loose lockdown measures. Germany experienced high wind conditions due to storms in February 2020 (Matthias et al., 2021), which was used to determine the extent of meteorology’s role in pollutant concentration changes. Meteorology unaccounted for mean NO$_2$ and PM$_{2.5}$ concentrations for February 1 to March 20, 2020 period (before the implementation of lockdown) are lower than the corresponding ones in 2019 by 30 % and 42 % ($f$NO$_2$(obs) and $f$PM$_{2.5}$(obs)), respectively, due to the dilution/dispersion from the high wind conditions. However, after accounting for meteorology, the difference in mean NO$_2$ and PM$_{2.5}$ concentrations between 2020 and 2019 for the period February 1 to March 20 ($f$NO$_2$(obs,emi) and $f$PM$_{2.5}$(obs,emi)) are 8 % and 18 %, respectively. This finding is consistent with meteorology accounted for mean NO$_2$ and PM$_{2.5}$ changes between 2020 and 2019 for the period January 1 to January 31 (Fig. 2 (a,b)). This highlights the importance of accounting for meteorological impacts.

In the 2020 pre-lockdown period (January 1 to March 20), both meteorology accounted for mean NO$_2$ and PM$_{2.5}$ levels are lower by 9 % and 19 %, respectively, compared to the same period in 2019. During the 2020 lockdown period (March 21 to May 31), mean meteorology accounted for NO$_2$ concentrations dropped significantly (23 %) compared to the same period in 2019, which is greater than the drop in the 2020 pre-lockdown period compared to 2019 (9 %). Comparatively, mean meteorology
accounted for 2020 lockdown PM$_{2.5}$ concentrations show a smaller reduction (5 %) compared to the same period in 2019, while an important precursor, NO$_2$, decreased by 23 % during the same period. Furthermore, the meteorology accounted for PM$_{2.5}$ reduction during the 2020 lockdown period (5 %) is less than the meteorology accounted for PM$_{2.5}$ reduction observed during the 2020 pre-lockdown period (19 %) compared to the corresponding 2019 periods (Fig. 2). Especially in Munich and Stuttgart, meteorology accounted for PM$_{2.5}$ concentrations during the 2020 lockdown period are higher than in 2019. The meteorology accounted for mean O$_3$ concentrations in the 2020 lockdown period are increased by 6 % compared to the same period in 2019. The increase in O$_3$ concentration during the 2020 lockdown period is mainly due to being in a NO$_X$ saturated regime (Gaubert et al., 2021), in which reducing NO$_X$ emission results in an increase in O$_3$ concentrations (Sillman, 1999; Sillman et al., 1990).

The effects of lockdown restrictions on SO$_2$ concentrations are insignificant. In comparison to 2019, TROPOMI meteorology accounted for SO$_2$ levels are decreased by 1 % during the 2020 lockdown period compared to 2019 (Fig. A1). For accounting meteorological impacts on TROPOMI satellite column concentrations, GEOS-Chem diagnostics (47 vertical layers) were converted to a column, applying TROPOMI’s averaging kernel. Because of the large influence of background concentration on satellite column measurements, we also investigated in-situ SO$_2$ concentrations, but only for five metropolitan areas. Similarly, we found that the impact of lockdown restrictions on in-situ SO$_2$ concentrations is marginal (Fig. B1). The road transportation sector contributes less than 1 % of total sulfur dioxide emissions, while coal-related fuel burning (industrial and energy production) accounts for nearly 80 % of total sulfur dioxide emissions (SO2, 2021). Because the lockdown restrictions primarily reduced traffic-related emissions, we see far less effects of the lockdown on SO$_2$ concentration (slight increase or no significant decrease in other European metropolitan areas (Collivignarelli et al., 2020; Filonchyk et al., 2021; Higham et al., 2021)). We found similar effects on in-situ CO concentration changes in six metropolitan areas. The meteorology accounted for mean CO concentrations are lower by 3 % during the 2020 lockdown period compared to 2019 (Fig. B1). Stuttgart meteorology accounted for CO concentrations in 2020 were higher than 2019 at all times. Other metropolitan areas experienced minor reductions (Clark et al., 2021; Hörmann et al., 2021).

4.2 Model evidence of changes in air pollutants concentration resulting from lockdown restrictions

As mentioned in Sect. 3, we use the meteorology accounted for NO$_2$ and CO changes to adjust the anthropogenic NO$_X$ and VOC emissions in inventories due to lockdown restriction impacts. GC model simulations are then obtained with this scaled anthropogenic emission scenario (23 % reduction in NO$_X$ emission and unchanged VOC emissions) for the 2020 lockdown period. The NO$_X$ emission reduction is within the range of estimated NO$_X$ emission reductions using activity data for Europe by previous authors (Doumbia et al., 2021; Guevara et al., 2021) (25 % and 33 %, respectively). For those studies there are large differences in estimated VOC emission changes for Europe; Doumbia et al. (2021) estimated 34 % while Guevara et al. (2021) estimated 8 % reduction in VOC emissions. However, the real-time measurements at a United Kingdom station show no significant changes in many VOC concentrations during the lockdown period (Grange et al., 2020). For the NO$_X$ saturated ozone production regime, VOC emission reductions can decrease ozone levels, while NO$_X$ emission reductions increase them. Gaubert et al. (2021) conducted a sensitivity study of modelling work on ozone levels in response to the NO$_X$ or VOC
or both emission reductions for the 2020 lockdown period. The reduction in both emissions (NO\textsubscript{X} and VOC), suggested by Doumbia et al. (2021), results in slight increase in lockdown ozone levels (< 2.5 %) over only north-western Germany and slight decrease in lockdown ozone levels over other regions of Germany, compared to BAU levels. But, only reduction in NO\textsubscript{X} emission results in increased lockdown ozone levels (0-10 %) over all of Germany compared to BAU levels, which is also consistent with our results of increase in meteorology accounted for ozone levels over different metropolitan areas across Germany during 2020 lockdown period compared to 2019 levels. This implies that VOC emissions were either not reduced at all or by a much smaller percentage than NO\textsubscript{X} emissions.

The emission accounted for GC lockdown NO\textsubscript{2} concentrations decreased by 21 % ($fX_{(GC,emi)}$) while emission accounted for GC lockdown O\textsubscript{3} concentrations increased by 9 % compared to 2020 BAU (Fig. 3). This is consistent with previous studies (such as Balamurugan et al. (2021); Gaubert et al. (2021)) which show that German metropolitan areas are in a NO\textsubscript{X} saturated ozone production regime in spring. The emission accounted for GC lockdown PM concentrations show small decreases compared to 2020 BAU (Fig. 3). These results are consistent with previous studies (Gaubert et al., 2021; Hammer et al., 2021; Matthias et al., 2021; Menut et al., 2020), which used activity data to develop an emission reduction scenario and estimated small to no reduction in PM\textsubscript{2.5}, a significant drop in NO\textsubscript{2} and marginal increase in O\textsubscript{3} levels during 2020 lockdown period, compared to BAU levels, over Northern-Europe including Germany.
We investigated the GC PM$_{2.5}$ composition for the studied period to determine the role of reduced NO$_X$ emission on total PM$_{2.5}$. Major secondary PM$_{2.5}$ components are nitrate, sulfate, ammonium and organic aerosol, which, on average, correspond to 24 %, 23 %, 15 % and 30 % of total PM$_{2.5}$, respectively, during March 21 to May 31, 2019 (Fig. C1). Mean relative contribution of PM$_{2.5}$ species for 2020 (BAU) and 2020 (lockdown) are shown in Fig. D1 and E1, respectively. The emission accounted for GC PM nitrate levels during the 2020 initial lockdown period (March 21 to April 30) are 9.5 % lower than the 2020 BAU levels (fNIT$_{GC,emi}$) (Fig. 3 (a)), however, we see NO$_2$ decreased by 21 % during the same period. The decrease in emission accounted for GC PM nitrate is also less than the decrease in NO$_2$ during the second half of the lockdown (May 1 to May 31). The emission accounted for GC lockdown PM sulfate level show marginal increase (3.5 %), while emission accounted for GC lockdown PM ammonium shows marginal decrease (5.8 %), compared to 2020 BAU level. The slight increase (& decrease) in sulfate (& ammonium) was also found in the Hammer et al. (2021); Matthias et al. (2021) studies, which used activity data to adjust the COVID-19 emission scenario.

It is notable that the reduction in NO$_X$, a precursor to PM nitrate, does not directly translated into a decrease in PM nitrate formation. There are several pathways for the formation of nitric acid (HNO$_3$), which partition to PM nitrate (Allen et al., 2015; Bauer et al., 2007). The reaction of OH and NO$_2$ (homogeneous pathway) and the hydrolysis of N$_2$O$_5$ on aerosol particles (heterogeneous pathway) are the two major pathways (Chang et al., 2011, 2016; Mollner et al., 2010).

The reaction for HNO$_3$ formation via gas-phase oxidation of NO$_2$ by OH is:

$$\text{NO}_2 + \text{OH} \xrightarrow{M} \text{HNO}_3 \quad \text{(R1)}$$

The reactions resulting in HNO$_3$ formation via hydrolysis of N$_2$O$_5$ on aerosol surfaces are:

$$\text{NO}_2 + \text{O}_3 \rightarrow \text{NO}_3 + \text{O}_2 \quad \text{(R2)}$$

$$\text{NO}_3 + \text{NO}_2 \xrightarrow{M} \text{N}_2\text{O}_5 \quad \text{(R3)}$$

$$\text{N}_2\text{O}_5 + \text{H}_2\text{O}(l) \rightarrow 2\text{HNO}_3 \quad \text{(R4)}$$

The formation of HNO$_3$ from the reaction of OH and NO$_2$ dominates during the day, while hydrolysis of N$_2$O$_5$ on aerosol particles dominates at night as OH night-time concentrations are low and N$_2$O$_5$ photolyzes easily (Russell et al., 1986). At night, NO$_3$ radical can be an important precursor for PM nitrate via reactions (Eq. R3, R4) (Kang et al., 2021; Shah et al., 2020; Wang et al., 2013). The emission accounted for concentrations of OH and NO$_3$, which drive day and night-time formation of PM nitrate, increased substantially (15 % and 12 %, respectively) during the lockdown period compared to BAU (Fig. 3). The increase in OH radicals results from German metropolitan areas being in a NO$_X$ saturated regime (Shah et al., 2020). The increase in GC lockdown NO$_3$ levels is predominantly at night due to a significant increase in night-time O$_3$ (Fig. 4 (b,e)); the reaction of NO$_2$ with O$_3$ is the most important source of NO$_3$ radical (Eq. R2) (Geyer et al., 2001).
Figure 4. Diurnal cycle of emission accounted for GC NO\textsubscript{2}, O\textsubscript{3}, OH radical, HNO\textsubscript{3} production from oxidation of NO\textsubscript{2} by OH pathway, NO\textsubscript{3} radical, N\textsubscript{2}O\textsubscript{5}, HNO\textsubscript{3} production from N\textsubscript{2}O\textsubscript{5} hydrolysis pathway, PM nitrate, sulfate, ammonium concentration changes between 2020 lockdown and 2020 BAU (no lockdown) scenario ($fX_{(\text{GC,emi})}$). Error bars represent the standard error of respective hour in ten metropolitan areas.
Liu et al. (2020) have demonstrated that analyzing the diurnal cycle of total inorganic nitrate helps to identify the dominant pathway for the particulate nitrate production. The emission accounted for GC lockdown PM nitrate levels decreased significantly during the day, while night-time lockdown PM nitrate levels decreased slightly compared to BAU levels (Fig. 4 (h)). Even though GC lockdown OH levels increased, HNO$_3$ production from the OH+NO$_2$ reaction during the lockdown period is reduced due to significantly lower day-time NO$_2$ levels compared to BAU (Fig. 4 (d)); as a result, GC day-time lockdown PM nitrate levels are significantly lower compared to BAU levels. However, higher night-time NO$_3^-$ levels result in higher night-time HNO$_3$ production from N$_2$O$_5$ hydrolysis, resulting in slightly lower night-time lockdown PM nitrate compared to BAU (Fig. 4 (b,e,f,g)). This implies that the increase in NO$_3^-$ radical due to increased ozone partially offset the effect of reduced NO$_X$ on nitrate formation. Previous studies have also shown that N$_2$O$_5$ hydrolysis plays important role in nitrate formation than the gas-phase day-time pathway (NO$_2$ + OH) (Allen et al., 2015; Chan et al., 2021; Kim et al., 2014; Liu et al., 2020; Yan et al., 2019). Figure 5 illustrates the conceptual model of generalized day and night-time lockdown NO$_X$ chemistry compared to BAU scenario. The oxidation of SO$_2$ is a major source of sulfate, and the reaction with the OH radical dominates the gas-phase oxidation of SO$_2$ (Zhang et al., 2015). Therefore, the enhanced sulfate formation during the 2020 lockdown period could be due to the increased oxidizing capacity of atmosphere (OH) since we observe no significant change in emission accounted for GC SO$_2$ concentration, compared to BAU concentration (Fig. 3). Organic aerosol (OA) formation could be affected by the changes in oxidizing capacity of the atmosphere (Carlton et al., 2009), but no changes in emission accounted for GC lockdown OA were observed compared to 2020 BAU scenario. Therefore, the fact that no significant change in PM$_{2.5}$ due to lockdown restrictions is observed can be explained by a significant offset of the decreased day-time PM nitrate formation by enhanced formation of PM sulfate, while PM ammonium shows a marginal decrease.

### 4.3 Link between spring PM$_{2.5}$ pollution episodes and high NH$_3$ concentrations

It is worth noting that a significant fraction of PM$_{2.5}$ is PM nitrate. Ammonia (NH$_3$) is an important precursor for particulate nitrate formation (Ansari and Pandis, 1998; Banzhaf et al., 2013; Behera and Sharma, 2010; Wu et al., 2016). This explains the importance of monitoring and potentially regulating ammonia emissions. Therefore, the inter- and intra-annual changes in ammonia (NH$_3$) concentrations over Germany, as well as their relationship to PM$_{2.5}$ variability, are reviewed and analyzed further below. In Germany, atmospheric NH$_3$ levels follow a monthly pattern, with NH$_3$ levels peaking in April (Fig. 6 (b) and 7). NH$_3$ levels are also elevated during summer months. In Europe, major agricultural practices (fertilizer and manure applications) take place in the early spring (Petetin et al., 2016; Ramanantenasoa et al., 2018; Viatte et al., 2020). The higher atmospheric ammonia levels in April are attributable to agricultural practices such as fertilizer application. The high NH$_3$ values in summer are most likely due to warm climates (Kuttippurath et al., 2020). Monthly average NH$_3$ maps clearly show the high NH$_3$ values over North-West Germany from April to August, with particularly high values in April. It indicates that North-West Germany is a hotspot of ammonia emissions compared to the rest of the country. North-West Germany is known for its high livestock density (livestock farming (EUR, 2013; Scarlat et al., 2018)) and it is dominated by crop and grass land (ESA, 2017). Livestock farming and fertilizer application account for 75% of NH$_3$ emissions in Europe (Webb et al., 2005). NH$_3$ concentrations in Germany vary greatly from year to year (inter-annual variabilities). We consider the period between
March 21 and April 30 when a stricter lockdown was in place to illustrate the inter-annual variability of atmospheric NH$_3$ between 2018 and 2020 (Fig. 8). NH$_3$ levels are lower in 2019 than in 2018, which can be attributed to the lower temperature in 2019 compared to 2018. Meanwhile, even though strict lockdown was in place, NH$_3$ levels in 2020 are higher than in 2019 and 2018, possibly due to low precipitation. High temperatures promote NH$_3$ volatilization (increases the NH$_3$ level in the atmosphere) (Ernst and Massey, 1960), whereas high rainfall favors wet deposition (removal of atmospheric NH$_3$). Schiferl et al. (2016); Viatte et al. (2020) have also shown that meteorological parameters such as temperature and precipitation play a greater role in NH$_3$ inter-annual variability.

High PM pollution episodes are likely to occur frequently during the winter due to high residential heating demand and favorable meteorological conditions (e.g., low temperature and inversion condition). However, high concentrations of PM$_{2.5}$ are apparent in German metropolitan areas in the early spring (from the second half of March to the end of April, e.g., Fig. 6 (a) for Cologne metropolitan area). On March 21, 2020, the German government imposed COVID-19 lockdown restrictions. However, in-situ PM$_{2.5}$ concentrations during the initial lockdown period are higher than during the pre-lockdown period in 2020. High PM$_{2.5}$ levels from the second half of March to the end of April are also consistent with previous years without lockdown restrictions. It is notable that this high spring PM$_{2.5}$ episodes are associated with high NH$_3$ concentrations (Fig. 6 (b)). The high PM$_{2.5}$ events that occur in the spring have also been observed in other European cities, and they typically contain ammonium nitrate and ammonium sulfate (Fortems-Cheiney et al., 2016; Renner and Wolke, 2010; Schaap et al., 2004; Viatte et al., 2020, 2021). Above, we show the high NH$_3$ levels in early spring (April) and summer months. High PM$_{2.5}$ concentrations are evident in spring, however, we did not observe high PM$_{2.5}$ episodes in summer (Fig. 6 (a)). It is also worth noting that even
Figure 6. Annual daily mean PM$_{2.5}$ concentrations in Cologne (a). Zoom in closer to see high PM$_{2.5}$ pollution episodes with IASI NH$_3$ total columns in early spring in Cologne (b). Daily mean in-situ PM$_{2.5}$, GC simulated PM$_{2.5}$, and GC simulated NH$_4$ concentrations in Cologne (c). Statistical distribution of meteorological parameters for the cases “Simultaneous” (Sim) and “Independent” (Ind) in ten German metropolitan areas for 2018 and 2019 (d). “Simultaneous” - Simultaneous increase in NH$_3$ (IASI) and PM$_{2.5}$ (in-situ) concentrations on same day. “Independent” - Increase in NH$_3$ (IASI) concentration not corresponding to an increase in PM$_{2.5}$ (in-situ) concentration on same day. Error bars represent the 1 $\sigma$ of the mean of ten metropolitan areas (d).
Figure 7. Monthly mean IASI NH$_3$ total column at 1*1 degree resolution.
in the spring and winter PM$_{2.5}$ is not consistently high on days with high NH$_3$. This reflects the complexity of the process of gas to particle conversion. Despite high NH$_3$ concentrations, ammonia(NH$_3$)-to-ammonium(NH$_4$) conversion is mainly driven by various meteorological factors such as temperature (and relative humidity). Studies (Viatte et al., 2020; Wang et al., 2015; Watson et al., 1994) have shown that conditions such as temperature of less than 10 $^\circ$C and a high relative humidity of more than 70 % are optimal for atmospheric gas-phase NH$_3$ to transform into ammonium salts, mainly due to reversible ammonium nitrate formation, which depends on temperature and relative humidity; warm and dry conditions partition ammonia back to the gas phase (Mozurkewich, 1993). In comparison to summer, the impact of NH$_3$ on PM$_{2.5}$ formation is considerable for winter and spring over Europe (Viatte et al., 2020, 2021) and the US (Schiferl et al., 2016). Summer weather is typically warmer (and has lower relative humidity) than winter and spring, which could explain why high NH$_3$ concentrations are not associated with high PM$_{2.5}$ in summer or late spring. To further demonstrate this for German metropolitan areas, we consider two cases (“Simultaneous” and “Independent”) for 2018 and 2019 (Fig. 6 (d)).

Figure 8. Mean IASI NH$_3$ total column (daily IASI NH$_3$ measurements gridded at 0.25 degree resolution) (top), mean temperature and wind (middle) and mean precipitation (bottom).
and PM$_{2.5}$ (in-situ) concentrations on same day. “Independent” - Increase in NH$_3$ (IASI) concentration not corresponding to an increase in PM$_{2.5}$ (in-situ) concentration on same day. As an example, for the Cologne metropolitan area, the temperature and boundary layer height for the “Simultaneous” case (11.7±6.8 °C and 500.4±166.5 m, respectively) is lower than for the “Independent” case (13.4±6 °C and 628.9±274.3 m, respectively). In addition to low temperature, low boundary layer height results in higher pollutant concentrations and can thus result in more intense atmospheric chemical reactions. We found similar results for other metropolitan areas, but with different absolute values (Fig. 6 (d)). The regional differences are unsurprising, because other factors also influence the formation of PM$_{2.5}$ from NH$_3$ (e.g., other precursor concentrations such as NO$_X$ and SO$_X$). However, these findings support previous studies and imply that low temperature and low boundary layer height are most favorable for the formation of PM$_{2.5}$ during the periods of high NH$_3$. GC also simulates the high spring PM$_{2.5}$ concentrations that have been observed, with high ammonium (NH$_4$) concentrations (Fig. 6 (c)).

5 Conclusions

Our study estimates the influence of anthropogenic emission reductions on PM$_{2.5}$ concentration changes during the 2020 lockdown period in German metropolitan areas. Mean meteorology accounted for PM$_{2.5}$ concentrations decreased by 5 % during the 2020 lockdown period (spring) compared to the corresponding period in 2019. However, during the 2020 pre-lockdown period (winter), meteorology accounted for PM$_{2.5}$ concentrations are 19 % lower than in 2019. Meanwhile, meteorology accounted for NO$_2$ levels decreased 23 % during the 2020 lockdown period, which is a larger decrease than 2020 pre-lockdown period compared to 2019 (9 %). No significant change in meteorology accounted for SO$_2$ and CO concentrations were observed during the 2020 lockdown period, compared to 2019.

The GC model with the COVID-19 emission reduction scenario based on observations (23 % reduction in NO$_X$ emission with unchanged VOC and SO$_2$) supports our findings of only a marginal decrease in PM$_{2.5}$ and a significant decrease in NO$_2$ levels. Due to being in a NO$_X$ saturated ozone production regime, the GC lockdown OH and O$_3$ concentrations increased by 15 % and 9 %, respectively, compared to BAU levels. Despite an increase in OH radicals, the GC lockdown PM nitrate formation decreased significantly during the day, due to a significant decrease in NO$_2$, compared to the BAU scenario. Increased night-time ozone, however, results in increased night-time NO$_3$, despite decreased NO$_2$, in turn, resulting in slightly increased night-time N$_2$O$_5$ concentration and only a small change in night-time PM nitrate. Overall this results in a small decrease in daily PM nitrate. In addition, the increased OH concentration results in a marginal increase of sulfate formation. Nitrate, sulfate, ammonium and organic aerosol are the major secondary components of PM$_{2.5}$. The decreased day-time PM nitrate is partially offset by the enhanced PM sulfate, and there is no significant impact from slightly decreased PM ammonium and no change in organic aerosol, resulting in a marginal decrease in PM$_{2.5}$ concentrations during the lockdown period.

Based on our findings, we suggest that additional emission control measures aimed at reducing ozone pollution be implemented which should also help reduce PM. A concurrent reduction of NO$_X$ and VOCs emissions should occur. Otherwise, ozone levels will rise as NO$_X$ emissions drop, increasing oxidizing capacity, until a NO$_X$ limited ozone production regime is reached. We also addressed the annual spring PM$_{2.5}$ pollution episodes in German metropolitan areas, which are associ-
ated with high NH$_3$ concentrations. North-West Germany is a hot-spot of NH$_3$ emissions, primarily emitted from livestock farming and intensive agricultural activities (fertilizer application), with high NH$_3$ concentrations in the early spring and summer months. Winter and spring meteorological conditions are more favorable for PM$_{2.5}$ formation from NH$_3$ than summer.

Unsurprisingly, low temperature (and low boundary layer height) is shown to be a favorable meteorological condition for the formation of PM$_{2.5}$ from NH$_3$. Regulation of NH$_3$ emissions, primarily from agriculture, has the potential to reduce PM$_{2.5}$ pollution significantly in German metropolitan areas.

In this study, a COVID-19 emission reduction scenario was created using meteorology accounted for proxy pollutant concentration changes, assuming that observed proxy pollutant concentration changes are due to the combined direct effects of emission and meteorology changes. Our GC modeling study work reflects the assumed direct relationship between changes in meteorology accounted for NO$_2$ concentration and changes in NO$_X$ emission. This work also shows a direct relationship between changes in meteorology accounted for SO$_2$ (and CO) concentration and changes in SO$_X$ (and CO) emission. However, due to the non-linear feedback system in atmospheric chemistry, this assumption should be investigated further. Because of their similar sources, we use CO concentration as a proxy for anthropogenic VOC concentration. However, this is debatable because VOC is more reactive than CO. We call for further advancements in estimating the emission changes during the lockdown period, which would allow us to estimate the precise sensitivity of PM$_{2.5}$ to changes in emissions from various sources and comparison of VOC emission inventories with observations. This will help in the implementation of appropriate air quality regulation strategies in the future.

Data availability. Hourly measurements of in-situ NO$_2$, O$_3$, PM$_{2.5}$, SO$_2$ and CO data are downloaded from (https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm). The TROPOMI SO$_2$ data are obtained from https://s5phub.copernicus.eu/. The IASI NH$_3$ data are obtained from https://iasi.aeris-data.fr/catalog/. Hourly ERA5 meteorological data are available at https://cds.climate.copernicus.eu/.
Figure A1. Meteorology unaccounted for (red) and meteorology accounted for (green) mean changes in PM$_{2.5}$, NO$_2$, SO$_2$ and O$_3$ concentrations between 2020 and 2019 in ten German metropolitan areas. Error bars represent the 1 $\sigma$ of mean of ten metropolitan areas.
Figure B1. Meteorology unaccounted for (red) and meteorology accounted for (green) mean changes in in-situ SO$_2$ (Bremen, Dresden, Frankfurt, Hamburg and Leipzig) and in in-situ CO (Bremen, Frankfurt, Hamburg, Hanover, Munich and Stuttgart) between 2020 and 2019. Error bars represent the $1 \sigma$ of mean of above mentioned metropolitan areas.

$\frac{fX_{\text{obs}}}{fX_{\text{obs,est}}}$
Figure C1. Mean relative contributions of PM$_{2.5}$ species simulated by GC for 2019.
Figure D1. Mean relative contributions of PM$_{2.5}$ species simulated by GC for 2020 (no lockdown).
Figure E1. Mean relative contributions of PM$_{2.5}$ species simulated by GC for 2020 (lockdown).
Author contributions. ZQ performed the modeling work; VB, and XB obtained the measurement data; VB analyzed the data and wrote the manuscript draft; JC, and FK supervised the work and edited the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledgements. Vigneshkumar Balamurugan, Jia Chen, and Xiao Bi are 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). Frank N. Keutsch is funded and supported by the Harvard’ Solar Geoengineering Research Program.
References

EUROSAT, Livestock density by NUTS 2 regions, EU-28, 2013. https://ec.europa.eu/eurostat/statistics-explained/index.php?title=File: Livestock_density_by_NUTS_2_regions._EU-28._2013.png, 2013.

ESA, Mapping Germany’s agricultural landscape. https://www.esa.int/ESA_Multimedia/Images/2017/08/Mapping_Germany_s_agricultural_landscape, 2017.

SO2 emission (EEA), Indicator Assessment:Sulphur dioxide (SO2) emissions. https://www.eea.europa.eu/data-and-maps/indicators/eea-32-sulphur-dioxide-so2-emissions-1/assessment-3, 2021.

Allen, H. M., Draper, D. C., Ayres, B. R., Ault, A., Bondy, A., Takahama, S., Modini, R. L., Baumann, K., Edgerton, E., Knote, C., et al.: Influence of crustal dust and sea spray supermicron particle concentrations and acidity on inorganic NO3 aerosol during the 2013 Southern Oxidant and Aerosol Study, Atmospheric Chemistry and Physics, 15, 10 669–10 685, 2015.

Ansari, A. S. and Pandis, S. N.: Response of inorganic PM to precursor concentrations, Environmental Science & Technology, 32, 2706–2714, 1998.

Ayres, B., Allen, H., Draper, D., Brown, S., Wild, R., Jimenez, J., Day, D., Campuzano-Jost, P., Hu, W. d., Gouw, J. d., et al.: Organic nitrate aerosol formation via NO3+ biogenic volatile organic compounds in the southeastern United States, Atmospheric Chemistry and Physics, 15, 13 377–13 392, 2015.

Baker, A. K., Beyersdorf, A. J., Doezema, L. A., Katzenstein, A., Meinardi, S., Simpson, I. J., Blake, D. R., and Rowland, F. S.: Measurements of nonmethane hydrocarbons in 28 United States cities, Atmospheric Environment, 42, 170–182, 2008.

Balamurugan, V., Chen, J., Qu, Z., Bi, X., Gensheimer, J., Shekhar, A., Bhattacharjee, S., and Keutsch, F. N.: Tropospheric NO2 and O3 response to COVID-19 lockdown restrictions at the national and urban scales in Germany, Journal of Geophysical Research: Atmospheres, 126, 2021.

Banzhaf, S., Schaap, M., Wichink Kruit, R. J., Denier Van Der Gon, H., Stern, R., and Builjtes, P.: Impact of emission changes on secondary inorganic aerosol episodes across Germany, Atmospheric Chemistry and Physics, 13, 11 675–11 693, 2013.

Barré, J., Petetin, H., Colette, A., Guevara, M., Peuch, V.-H., Rouil, L., Engelen, R., Inness, A., Flemming, J., Pérez García-Pando, C., et al.: Estimating lockdown induced European NO2 changes, Atmospheric Chemistry and Physics Discussions, pp. 1–28, 2020.

Bauer, S., Koch, D., Unger, N., Metzger, S., Shindell, D., and Streets, D.: Nitrate aerosols today and in 2030: a global simulation including aerosols and tropospheric ozone, Atmospheric Chemistry and Physics, 7, 5043–5059, 2007.

Bauwens, M., Compernolle, S., Stavrakou, T., Müller, J.-F., Van Gent, J., Eskes, H., Levelt, P. F., van der A, R., Veefkind, J., Vlietinck, J., et al.: Impact of coronavirus outbreak on NO2 pollution assessed using TROPOMI and OMI observations, Geophysical Research Letters, 47, e2020GL087 978, 2020.

Behera, S. N. and Sharma, M.: Investigating the potential role of ammonia in ion chemistry of fine particulate matter formation for an urban environment, Science of the Total Environment, 408, 3569–3575, 2010.

Biswal, A., Singh, T., Singh, V., Ravindra, K., and Mor, S.: COVID-19 lockdown and its impact on tropospheric NO2 concentrations over India using satellite-based data, Heliyon, 6, e04 764, 2020.

Blanchard, C. L. and Tanenbaum, S. J.: Differences between weekday and weekend air pollutant levels in southern California, Journal of the Air & Waste Management Association, 53, 816–828, 2003.

Campbell, P. C., Tong, D., Tang, Y., Baker, B., Lee, P., Saylor, R., Stein, A., Ma, S., Lamsal, L., and Qu, Z.: Impacts of the COVID-19 Economic Slowdown on Ozone Pollution in the US, Atmospheric Environment, p. 118713, 2021.
Carlton, A., Wiedinmyer, C., and Kroll, J.: A review of Secondary Organic Aerosol (SOA) formation from isoprene, Atmospheric Chemistry and Physics, 9, 4987–5005, 2009.

Cesari, D., De Benedetto, G., Bonasoni, P., Busetto, M., Dinoi, A., Merico, E., Chirizzi, D., Cristofanelli, P., Donateo, A., Grasso, F., et al.: Seasonal variability of PM2.5 and PM10 composition and sources in an urban background site in Southern Italy, Science of the Total Environment, 612, 202–213, 2018.

Chan, Y.-C., Evans, M. J., He, P., Holmes, C. D., Jaeglé, L., Kasibhatla, P., Liu, X.-Y., Sherwen, T., Thornton, J. A., Wang, X., et al.: Heterogeneous nitrate production mechanisms in intense haze events in the North China Plain, Journal of Geophysical Research: Atmospheres, 126, e2021JD034688, 2021.

Chang, W. L., Bhave, P. V., Brown, S. S., Riemer, N., Stutz, J., and Dabdub, D.: Heterogeneous atmospheric chemistry, ambient measurements, and model calculations of N2O5: A review, Aerosol Science and Technology, 45, 665–695, 2011.

Chang, W. L., Brown, S. S., Stutz, J., Middlebrook, A. M., Bahreini, R., Wagner, N. L., Dubé, W. P., Pollack, I. B., Ryerson, T. B., and Riemer, N.: Evaluating N2O5 heterogeneous hydrolysis parameterizations for CalNex 2010, Journal of Geophysical Research: Atmospheres, 121, 5051–5070, 2016.

Clark, H., Bennouna, Y., Tsivlidou, M., Wolff, P., Sauvage, B., Barret, B., Le Flochmoën, E., Blot, R., Boulanger, D., Cousin, J.-M., et al.: The Effects of the COVID-19 Lockdowns on the Composition of the Troposphere as Seen by IAGOS, Atmospheric Chemistry and Physics Discussions, pp. 1–33, 2021.

Clerbaux, C., Boyard, A., Clarisse, L., George, M., Hadji-Lazaro, J., Herbin, H., Hurtmans, D., Pommier, M., Razavi, A., Turquety, S., et al.: Monitoring of atmospheric composition using the thermal infrared IASI/MetOp sounder, Atmospheric Chemistry and Physics, 9, 6041–6054, 2009.

Collivignarelli, M. C., Abbà, A., Bertanza, G., Pedrazzani, R., Ricciardi, P., and Miino, M. C.: Lockdown for CoViD-2019 in Milan: What are the effects on air quality?, Science of the total environment, 732, 139280, 2020.

Deroubaix, A., Brasseur, G., Gautbert, B., Labuhn, I., Menut, L., Siour, G., and Tuccella, P.: Response of surface ozone concentration to emission reduction and meteorology during the COVID-19 lockdown in Europe, Meteorological Applications, 28, e1990, 2021.

Dietrich, F., Chen, J., Voggenreiter, B., Aigner, P., Nachtigall, N., and Reger, B.: MUCNnet: Munich urban carbon column network, Atmospheric Measurement Techniques, 14, 1111–1126, 2021.

Doumbia, T., Granier, C., Elguindi, N., Bouarar, I., Darras, S., Brasseur, G., Gautbert, B., Liu, Y., Shi, X., Stavvakou, T., et al.: Changes in global air pollutant emissions during the COVID-19 pandemic: a dataset for atmospheric chemistry modeling, Earth System Science Data Discussions, pp. 1–26, 2021.

Erisman, J. and Schaap, M.: The need for ammonia abatement with respect to secondary PM reductions in Europe, Environmental Pollution, 129, 159–163, 2004.

Ernst, J. and Massey, H.: The effects of several factors on volatilization of ammonia formed from urea in the soil, Soil Science Society of America Journal, 24, 87–90, 1960.

Field, R. D., Hickman, J. E., Geogdzhayev, I. V., Tsigrisid, K., and Bauer, S. E.: Changes in satellite retrievals of atmospheric composition over eastern China during the 2020 COVID-19 lockdowns, Atmospheric Chemistry and Physics Discussions, pp. 1–31, 2020.

Filonchyk, M., Hurynovich, V., and Yan, H.: Impact of Covid-19 lockdown on air quality in the Poland, Eastern Europe, Environmental Research, 198, 110454, 2021.

Fisher, J. A., Jacob, D. J., Travis, K. R., Kim, P. S., Marais, E. A., Chan Miller, C., Yu, K., Zhu, L., Yantosca, R. M., Sulprizio, M. P., et al.: Organic nitrate chemistry and its implications for nitrogen budgets in an isoprene-and monoterpane-rich atmosphere: constraints from...
Fortems-Cheiney, A., Dufour, G., Hamaoui-Laguel, L., Foret, G., Siour, G., Van Damme, M., Meleux, F., Coheur, P.-F., Clerbaux, C., Clarisse, L., et al.: Unaccounted variability in NH3 agricultural sources detected by IASI contributing to European spring haze episode, Geophysical Research Letters, 43, 5475–5482, 2016.

Fujita, E. M., Campbell, D. E., Zielinska, B., Sagebiel, J. C., Bowen, J. L., Goliff, W. S., Stockwell, W. R., and Lawson, D. R.: Diurnal and weekday variations in the source contributions of ozone precursors in California’s South Coast Air Basin, Journal of the Air & Waste Management Association, 53, 844–863, 2003.

Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., Denier van der Gon, H., Facchini, M. C., Fowler, D., Koren, I., Langford, B., Lohmann, U., et al.: Particulate matter, air quality and climate: lessons learned and future needs, Atmospheric chemistry and physics, 15, 8217–8299, 2015.

Gaubert, B., Bouarar, I., Doumbia, T., Liu, Y., Stavarakou, T., Deroubaix, A., Darras, S., Elguindi, N., Granier, C., Lacey, F., et al.: Global changes in secondary atmospheric pollutants during the 2020 COVID-19 pandemic, Journal of Geophysical Research: Atmospheres, 126, e2020JD034213, 2021.

Gensheimer, J., Turner, A. J., Shekhar, A., Wenzel, A., Keutsch, F. N., and Chen, J.: What are different measures of mobility telling us about surface transportation CO2 emissions during the COVID-19 pandemic?, Journal of Geophysical Research: Atmospheres, p. e2021JD034664, 2021.

Geyer, A., Alicke, B., Konrad, S., Schmitz, T., Stutz, J., and Platt, U.: Chemistry and oxidation capacity of the nitrate radical in the continental boundary layer near Berlin, Journal of Geophysical Research: Atmospheres, 106, 8013–8025, 2001.

Goldberg, D. L., Anenberg, S. C., Griffin, D., McLinden, C. A., Lu, Z., and Streets, D. G.: Disentangling the impact of the COVID-19 lockdowns on urban NO2 from natural variability, Geophysical Research Letters, 47, eabg7670, 2020.

Grange, S. K., Lee, J. D., Drysdale, W. S., Lewis, A. C., Hueglin, C., Emmenegger, L., and Carslaw, D. C.: COVID-19 lockdowns highlight a risk of increasing ozone pollution in European urban areas, Atmospheric Chemistry and Physics Discussions, pp. 1–25, 2020.

Guevara, M., Jorba, O., Soret, A., Petetin, H., Bowdalo, D., Serradell, K., Tena, C., Denier van der Gon, H., Kuenen, J., Peuch, V.-H., et al.: Time-resolved emission reductions for atmospheric chemistry modelling in Europe during the COVID-19 lockdowns, Atmospheric Chemistry and Physics, 21, 773–797, 2021.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeyss, M., Dommen, J., Donahue, N., George, C., Goldstein, A., et al.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmospheric chemistry and physics, 9, 5155–5236, 2009.

Hammer, M. S., van Donkelaar, A., Martin, R. V., McDuffie, E. E., Lyapustin, A., Sayer, A. M., Hsu, N. C., Levy, R. C., Garay, M. J., Kalashnikova, O. V., et al.: Effects of COVID-19 lockdowns on fine particulate matter concentrations, Science Advances, 7, eabg7670, 2021.

He, C., Hong, S., Zhang, L., Mu, H., Xin, A., Zhou, Y., Liu, J., Liu, N., Su, Y., Tian, Y., et al.: Global, continental, and national variation in PM2.5, O3, and NO2 concentrations during the early 2020 COVID-19 lockdown, Atmospheric pollution research, 12, 136–145, 2021.

Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas, J., Peubey, C., Radu, R., Schepers, D., et al.: The ERA5 global reanalysis, Quarterly Journal of the Royal Meteorological Society, 146, 1999–2049, 2020.

Higham, J., Ramírez, C. A., Green, M., and Morse, A.: UK COVID-19 lockdown: 100 days of air pollution reduction?, Air Quality, Atmosphere & Health, 14, 325–332, 2021.
Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., et al.: Historical (1750–2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS), Geoscientific Model Development, 11, 369–408, 2018.

Hörmann, S., Jammoul, F., Kuenzer, T., and Stadlober, E.: Separating the impact of gradual lockdown measures on air pollutants from seasonal variability, Atmospheric pollution research, 12, 84–92, 2021.

Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., et al.: Enhanced secondary pollution offset reduction of primary emissions during COVID-19 lockdown in China, National Science Review, 8, nwaa137, 2021.

Hudman, R., Moore, N., Martin, R., Russell, A., Mebust, A., Valin, L., and Cohen, R.: A mechanistic model of global soil nitric oxide emissions: implementation and space based-constraints., Atmospheric Chemistry & Physics Discussions, 12, 2012.

Jacob, D. J.: Introduction to atmospheric chemistry, Princeton University Press, 1999.

Jacobson, M. Z.: Fundamentals of atmospheric modeling, Cambridge university press, 1999.

Jiménez, P., Parra, R., Gasso, S., and Baldasano, J. M.: Modeling the ozone weekend effect in very complex terrains: a case study in the Northeastern Iberian Peninsula, Atmospheric environment, 39, 429–444, 2005.

Kang, M., Zhang, J., Zhang, H., and Ying, Q.: On the Relevancy of Observed Ozone Increase during COVID-19 Lockdown to Summertime Ozone and PM2.5 Control Policies in China, Environmental Science & Technology Letters, 8, 289–294, 2021.

Kang, Y.-H., You, S., Bae, M., Kim, E., Son, K., Bae, C., Kim, Y., Kim, B.-U., Kim, H. C., and Kim, S.: The impacts of COVID-19, meteorology, and emission control policies on PM 2.5 drops in Northeast Asia, Scientific reports, 10, 1–8, 2020.

Keller, C. A., Evans, M. J., Knowland, K. E., Hasenkopf, C. A., Modekurty, S., Lucchesi, R. A., Oda, T., Franca, B. B., Mandarino, F. C., Diaz Suárez, M. V., et al.: Global impact of COVID-19 restrictions on the surface concentrations of nitrogen dioxide and ozone, Atmospheric Chemistry and Physics, 21, 3555–3592, 2021.

Kim, Y. J., Spak, S. N., Carmichael, G. R., Riemer, N., and Stanier, C. O.: Modeled aerosol nitrate formation pathways during wintertime in the Great Lakes region of North America, Journal of Geophysical Research: Atmospheres, 119, 12–420, 2014.

Koukouli, M.-E., Skoulidou, I., Karavias, A., Parcharidis, I., Balis, D., Manders, A., Segers, A., Eskes, H., and van Geffen, J.: Sudden changes in nitrogen dioxide emissions over Greece due to lockdown after the outbreak of COVID-19, Atmospheric Chemistry and Physics, 21, 1759–1774, 2021.

Kroll, J. H., Heald, C. L., Cappa, C. D., Farmer, D. K., Fry, J. L., Murphy, J. G., and Steiner, A. L.: The complex chemical effects of COVID-19 shutdowns on air quality, Nature Chemistry, 12, 777–779, 2020.

Kuttippurath, J., Singh, A., Dash, S., Mallick, N., Clerbaux, C., Van Damme, M., Clarisse, L., Coheur, P.-F., Raj, S., Abhishek, K., et al.: Record high levels of atmospheric ammonia over India: Spatial and temporal analyses, Science of the Total Environment, 740, 139 986, 2020.

Le Quéré, C., Jackson, R. B., Jones, M. W., Smith, A. J., Abernethy, S., Andrew, R. M., De-Gol, A. J., Willis, D. R., Shan, Y., Canadell, J. G., et al.: Temporary reduction in daily global CO2 emissions during the COVID-19 forced confinement, Nature Climate Change, pp. 1–7, 2020.

Lee, J. D., Drysdale, W. S., Finch, D. P., Wilde, S. E., and Palmer, P. I.: UK surface NO2 levels dropped by 42% during the COVID-19 lockdown: impact on surface O3, Atmospheric Chemistry and Physics, 20, 15 743–15 759, 2020.

Liu, L., Bei, N., Hu, B., Wu, J., Liu, S., Li, X., Wang, R., Liu, Z., Shen, Z., and Li, G.: Wintertime nitrate formation pathways in the north China plain: Importance of N2O5 heterogeneous hydrolysis, Environmental Pollution, 266, 115 287, 2020.
Marais, E. A., Jacob, D. J., Jimenez, J. L., Campuzano-Jost, P., Day, D. A., Hu, W., Krechmer, J., Zhu, L., Kim, P. S., Miller, C. C., et al.: Aqueous-phase mechanism for secondary organic aerosol formation from isoprene: application to the southeast United States and co-benefit of SO2 emission controls, Atmospheric Chemistry and Physics, 16, 1603–1618, 2016.

Matthias, V., Quante, M., Arndt, J. A., Badeke, R., Fink, L., Petrik, R., Feldner, J., Schwarzkopf, D., Link, E.-M., Ramacher, M. O., et al.: The role of emission reductions and the meteorological situation for air quality improvements during the COVID-19 lockdown period in Central Europe, Atmospheric Chemistry and Physics, 21, 13 931–13 971, 2021.

Mendez-Espinosa, J. F., Rojas, N. Y., Vargas, J., Pachón, J. E., Belalcazar, L. C., and Ramírez, O.: Air quality variations in Northern South America during the COVID-19 lockdown, Science of the Total Environment, 749, 141 621, 2020.

Menut, L., Bessagnet, B., Siour, G., Mailler, S., Pennel, R., and Cholakian, A.: Impact of lockdown measures to combat Covid-19 on air quality over western Europe, Science of the Total Environment, 741, 140 426, 2020.

Mollner, A. K., Valluvadasan, S., Feng, L., Sprague, M. K., Okumura, M., Milligan, D. B., Bloss, W. J., Sander, S. P., Martien, P. T., Harley, R. A., et al.: Rate of gas phase association of hydroxyl radical and nitrogen dioxide, Science, 330, 646–649, 2010.

Mozurkewich, M.: The dissociation constant of ammonium nitrate and its dependence on temperature, relative humidity and particle size, Atmospheric Environment. Part A. General Topics, 27, 261–270, 1993.

Murray, L. T.: Lightning NOx and impacts on air quality, Current Pollution Reports, 2, 115–133, 2016.

Ordóñez, C., Garrido-Perez, J. M., and García-Herrera, R.: Early spring near-surface ozone in Europe during the COVID-19 shutdown: Meteorological effects outweigh emission changes, Science of the total environment, 747, 141 322, 2020.

Pathakoti, M., Muppalla, A., Hazra, S., Dangeti, M., Shekhar, R., Jella, S., Mullahudi, S. S., Andugulapati, P., and Vijayasundaram, U.: An assessment of the impact of a nation-wide lockdown on air pollution—a remote sensing perspective over India, Atmospheric Chemistry and Physics Discussions, pp. 1–16, 2020.

Pay, M. T., Jiménez-Guerrero, P., and Baldasano, J. M.: Assessing sensitivity regimes of secondary inorganic aerosol formation in Europe with the CALIOPE-EU modeling system, Atmospheric Environment, 51, 146–164, 2012.

Petetin, H., Sciare, J., Bressi, M., Gros, V., Rosso, A., Sanchez, O., Sarda-Estève, R., Petit, J.-E., and Beekmann, M.: Assessing the ammonium nitrate formation regime in the Paris megacity and its representation in the CHIMERE model, Atmospheric Chemistry and Physics, 16, 10 419–10 440, 2016.

Petetin, H., Bowdalo, D., Soret, A., Guevara, M., Jorba, O., Serradell, K., and Pérez García-Pando, C.: Meteorology-normalized impact of the COVID-19 lockdown upon NO2 pollution in Spain, Atmospheric Chemistry and Physics, 20, 11 119–11 141, 2020.

Putaud, J.-P., Pozzoli, L., Pisoni, E., Martins Dos Santos, S., Lagler, F., Lanzani, G., Dal Santo, U., and Colette, A.: Impacts of the COVID-19 lockdown on air pollution at regional and urban background sites in northern Italy, Atmospheric Chemistry and Physics, 21, 7597–7609, 2021.

Qu, Z., Jacob, D. J., Silvern, R. F., Shah, V., Campbell, P. C., Valin, L. C., and Murray, L. T.: US COVID-19 shutdown demonstrates importance of background NO2 in inferring NOx emissions from satellite NO2 observations, Geophysical research letters, 48, e2021GL092 783, 2021.

Ramanantenasoa, M. M. J., Gilliot, J.-M., Mignolet, C., Bedos, C., Mathias, E., Eglie, T., Makowski, D., and Génermont, S.: A new framework to estimate spatio-temporal ammonia emissions due to nitrogen fertilization in France, Science of the Total Environment, 645, 205–219, 2018.

Renner, E. and Wolke, R.: Modelling the formation and atmospheric transport of secondary inorganic aerosols with special attention to regions with high ammonia emissions, Atmospheric Environment, 44, 1904–1912, 2010.
Russell, A. G., Cass, G. R., and Seinfeld, J. H.: On some aspects of nighttime atmospheric chemistry, Environmental science & technology, 20, 1167–1172, 1986.

Scarlat, N., Fahl, F., Dallemand, J.-F., Monforti, F., and Motola, V.: A spatial analysis of biogas potential from manure in Europe, Renewable and Sustainable Energy Reviews, 94, 915–930, 2018.

Schaap, M., Loon, M. v., Ten Brink, H., Dentener, F., and Builtjes, P.: Secondary inorganic aerosol simulations for Europe with special attention to nitrate, Atmospheric Chemistry and Physics, 4, 857–874, 2004.

Schiferl, L. D., Heald, C. L., Damme, M. V., Clarisse, L., Clerbaux, C., Coheur, P.-F., Nowak, J. B., Neuman, J. A., Herndon, S. C., Roscioli, J. R., et al.: Interannual variability of ammonia concentrations over the United States: sources and implications, Atmospheric Chemistry and Physics, 16, 12 305–12 328, 2016.

Schumann, U., Poll, I., Teoh, R., Koelle, R., Spinielli, E., Molloy, J., Koudis, G. S., Baumann, R., Bugliaro, L., Stettler, M., et al.: Air traffic and contrail changes over Europe during COVID-19: a model study, Atmospheric Chemistry and Physics, 21, 7429–7450, 2021.

Seinfeld, J. H. and Pankow, J. F.: Organic atmospheric particulate material, Annual review of physical chemistry, 54, 121–140, 2003.

Shah, V., Jaeglé, L., Thornton, J. A., Lopez-Hilfiker, F. D., Lee, B. H., Schroder, J. C., Campuzano-Jost, P., Jimenez, J. L., Guo, H., Sullivan, A. P., et al.: Chemical feedbacks weaken the wintertime response of particulate sulfate and nitrate to emissions reductions over the eastern United States, Proceedings of the National Academy of Sciences, 115, 8110–8115, 2018.

Shah, V., Jacob, D. J., Li, K., Silvern, R. F., Zhai, S., Liu, M., Lin, J., and Zhang, Q.: 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, 1483–1495, 2020.

Sharma, M., Kishore, S., Tripathi, S., and Behera, S.: Role of atmospheric ammonia in the formation of inorganic secondary particulate matter: a study at Kanpur, India, Journal of Atmospheric Chemistry, 58, 1–17, 2007.

Sillman, S.: The relation between ozone, NOx and hydrocarbons in urban and polluted rural environments, Atmospheric Environment, 33, 1821–1845, 1999.

Sillman, S., Logan, J. A., and Wofsy, S. C.: The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, Journal of Geophysical Research: Atmospheres, 95, 1837–1851, 1990.

Solberg, S., Walker, S.-E., Schneider, P., and Guerreiro, C.: Quantifying the Impact of the Covid-19 Lockdown Measures on Nitrogen Dioxide Levels throughout Europe, Atmosphere, 12, 131, 2021.

Souri, A. H., Chance, K., Bak, J., Nowlan, C. R., González Abad, G., Jung, Y., Wong, D. C., Mao, J., and Liu, X.: Unraveling Pathways of Elevated Ozone Induced by the 2020 Lockdown in Europe by an Observationally Constrained Regional Model: Non-Linear Joint Inversion of NO x and VOC Emissions using TROPOMI, Atmospheric Chemistry and Physics Discussions, pp. 1–45, 2021.

Squizzato, S., Masiol, M., Brunelli, A., Pistollato, S., Tarabotti, E., Rampazzo, G., and Pavoni, B.: Factors determining the formation of secondary inorganic aerosol: a case study in the Po Valley (Italy), Atmospheric chemistry and physics, 13, 1927–1939, 2013.

Steinfeld, J. I.: Atmospheric chemistry and physics: from air pollution to climate change, Environment: Science and Policy for Sustainable Development, 40, 26–26, 1998.

Steinmetz, H., Batzdorfer, V., and Bosnjak, M.: The ZPID lockdown measures dataset for Germany, 2020.

Stephens, S., Madronich, S., Wu, F., Olson, J., Ramos, R., Retama, A., and Munoz, R.: Weekly patterns of México City’s surface concentrations of CO, NO x, PM 10 and O 3 during 1986–2007, Atmospheric Chemistry and Physics, 8, 5313–5325, 2008.

Tai, A. P., Mickley, L. J., and Jacob, D. J.: Impact of 2000–2050 climate change on fine particulate matter (PM2.5) air quality inferred from a multi-model analysis of meteorological modes, Atmospheric chemistry and physics, 12, 11 329–11 337, 2012.
Theys, N., Smedt, I. D., Yu, H., Danckaert, T., Gent, J. v., Hörmann, C., Wagner, T., Hedelt, P., Bauer, H., Romahn, F., et al.: Sulfur dioxide retrievals from TROPOMI onboard Sentinel-5 Precursor: algorithm theoretical basis, Atmospheric Measurement Techniques, 10, 119–153, 2017.

Turner, A. J., Kim, J., Fitzmaurice, H., Newman, C., Worthington, K., Chan, K., Wooldridge, P. J., Köehler, P., Frankenber, C., and Cohen, R. C.: Observed Impacts of COVID-19 on Urban CO 2 Emissions, Geophysical Research Letters, 47, e2020GL090037, 2020.

Viatte, C., Wang, T., Damme, M. V., Dammers, E., Meleux, F., Clarisse, L., Shephard, M. W., Whitburn, S., Coheur, P. F., Cady-Pereira, K. E., et al.: Atmospheric ammonia variability and link with particulate matter formation: a case study over the Paris area, Atmospheric Chemistry and Physics, 20, 577–596, 2020.

Viatte, C., Petit, J.-E., Yamanouchi, S., Van Damme, M., Doucerain, C., Germain-Piaulenne, E., Gros, V., Favez, O., Clarisse, L., Coheur, P.-F., et al.: Ammonia and PM2.5 Air Pollution in Paris during the 2020 COVID Lockdown, Atmosphere, 12, 160, 2021.

Von Schneidemesser, E., Monks, P. S., and Plass-Duelmer, C.: Global comparison of VOC and CO observations in urban areas, Atmospheric Environment, 44, 5053–5064, 2010.

Wang, L., Wang, J., and Fang, C.: Assessing the Impact of Lockdown on Atmospheric Ozone Pollution Amid the First Half of 2020 in Shenyang, China, International Journal of Dammers Research and Public Health, 17, 9004, 2020.

Wang, S., Nan, J., Shi, C., Fu, Q., Gao, S., Wang, D., Cui, H., Saiz-Lopez, A., and Zhou, B.: Atmospheric ammonia and its impacts on regional air quality over the megacity of Shanghai, China, Scientific reports, 5, 1–13, 2015.

Wang, W., van der A, R., Ding, J., van Weele, M., and Cheng, T.: Spatial and temporal changes of the ozone sensitivity in China based on satellite and ground-based observations, Atmospheric Chemistry and Physics, 21, 7253–7269, 2021.

Wang, Y., Zhang, Q., He, K., Zhang, Q., and Chai, L.: Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia, Atmospheric Chemistry and Physics, 13, 2635–2652, 2013.

Watson, J. G., Chow, J. C., Lurmann, F. W., and Musarra, S. P.: Ammonium nitrate, nitric acid, and ammonia equilibrium in wintertime Phoenix, Arizona, Air & Waste, 44, 405–412, 1994.

Webb, J., Menzi, H., Pain, B., Mieselbrook, T., Dämgen, U., Hendriks, H., and Döhler, H.: Managing ammonia emissions from livestock production in Europe, Environmental pollution, 135, 399–406, 2005.

Werf, G. R., Randerson, J. T., Giglio, L., Leeuwen, T. T. v., Chen, Y., Rogers, B. M., Mu, M., Van Marle, M. J., Morton, D. C., Collatz, G. J., et al.: Global fire emissions estimates during 1997–2016, Earth System Science Data, 9, 697–720, 2017.

Wu, S.-Y., Hu, J.-L., Zhang, Y., and Aneja, V. P.: Modeling atmospheric transport and fate of ammonia in North Carolina—Part II: Effect of ammonia emissions on fine particulate matter formation, Atmospheric Environment, 42, 3437–3451, 2008.

Wu, Y., Gu, B., Erisman, J. W., Reis, S., Fang, Y., Lu, X., and Zhang, X.: PM2.5 pollution is substantially affected by ammonia emissions in China, Environmental Pollution, 218, 86–94, 2016.

Yan, C., Tham, Y. J., Zha, Q., Wang, X., Xue, L., Dai, J., Wang, Z., and Wang, T.: Fast heterogeneous loss of N2O5 leads to significant nighttime NOx removal and nitrate aerosol formation at a coastal background environment of southern China, Science of the Total Environment, 677, 637–647, 2019.

Yarwood, G., Stoeckenius, T. E., Heiken, J. G., and Dunker, A. M.: Modeling weekday/weekend ozone differences in the Los Angeles region for 1997, Journal of the Air & Waste Management Association, 53, 864–875, 2003.

Yin, H., Lu, X., Sun, Y., Li, K., Gao, M., Zheng, B., and Liu, C.: Unprecedented decline in summertime surface ozone over eastern China in 2020 comparably attributable to anthropogenic emission reductions and meteorology, Environmental Research Letters, 2021.
Zhai, S., Jacob, D. J., Wang, X., Liu, Z., Wen, T., Shah, V., Li, K., Moch, J. M., Bates, K. H., Song, S., et al.: Control of particulate nitrate air pollution in China, Nature Geoscience, pp. 1–7, 2021.

Zhang, R., Wang, G., Guo, S., Zamora, M. L., Ying, Q., Lin, Y., Wang, W., Hu, M., and Wang, Y.: Formation of urban fine particulate matter, Chemical reviews, 115, 3803–3855, 2015.