

Dear Reviewer,

We appreciate your comments and suggestions, which have helped us improve our manuscript further. We have made the necessary changes to the manuscript, which can be found in the attached file (Track Changes). The following is a response to your comments and suggestions. Corresponding changes in the revised manuscript are also made available below, if applicable, at the appropriate places.

Sincerely,

On behalf of all co-authors, Vigneshkumar Balamurugan

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**The manuscript entitled “Secondary PM decreases significantly less than NO<sub>2</sub> emission reductions during COVID lockdown in Germany” by Vigneshkumar Balamurugan et al. explored the drivers of slight decrease of PM<sub>2.5</sub> compared to NO<sub>2</sub> emission during COVID-19 lockdown in Germany. The manuscript provides valuable information for understanding PM pollution under rigorous emission reduction measures and efficiently directing PM mitigation in the future. It is recommended that this manuscript be reconsidered for publication after major revisions.**

Thank you so much for reading and reviewing our manuscript! We carefully reviewed and considered your comments/suggestions, and made improvements in the revised manuscript.

**General comments:**

**Line 54:” 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”. More cases should be given to support this sentence.**

We cited additional studies to support this statement.

Lines 55-58:	<i>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; JudaRezler et al., 2020; Samek et al., 2020; Salameh et al., 2015; Womack et al., 2019; Zhai et al., 2021).</i>
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**Line 133:” 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” This definition is misleading. According to your definition of  $\Delta PM_{2.5}(obs)$  and  $\Delta PM_{2.5}(GC)$ , the**

**$\Delta PM_{2.5}(\text{obs,emi})$  should be the change of PM<sub>2.5</sub> caused only by emission. If so, relative descriptions in the whole paper should be revised correspondingly.**

$\Delta PM_{2.5}(\text{obs,emi})$  = Absolute concentration changes ( $\mu\text{g m}^{-3}$ ) after accounting for meteorology (caused only by emission) between 2020 and 2019.

$fPM_{2.5}(\text{obs,emi})$  = Fractional concentration changes (%) after accounting for meteorology (caused only by emission) between 2020 and 2019.

We hope this clarifies your comment. We also made minor changes to the sentence and equation to make it clearer to the reader.

Lines 139-141:	<p><i>The fractional change in meteorology accounted for pollutant concentration between 2020 and 2019, i.e., fractional change (%) in pollutant concentration between 2020 and 2019 due to emission changes only, is calculated as,</i></p> $fPM_{2.5}(\text{obs,emi}) = (\Delta PM_{2.5}(\text{obs,emi}) / PM_{2.5}(\text{obs,2019})) * 100$
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**Line 170: We also compared the 2019 GC and 2019 observed in-situ PM<sub>2.5</sub> concentrations and found that the GC and observed in-situ PM<sub>2.5</sub> 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 performance of the model is the base of further analysis. Hence, more details of the statistical evaluation of the model performance for each site should be given. In addition, the agreement R is above 0.5 for most areas and is 0.39 for Leipzig. Personally, I think the R is not good enough.**

In the revised manuscript (Table A1), we now included the performance of the GC model for each metropolitan area. The R value in all cases is between 0.3 and 0.7, indicating moderate correlation (<https://link.springer.com/article/10.1057/jt.2009.5>).

Table A1. The statistical evaluation (R, RMSE and mean bias) of the GC model performance (PM<sub>2.5</sub>) for the 2019 study period (January 1 to May 31).

Metropolitan area	Correlation coefficient (R)	RMSE ( $\mu\text{g m}^{-3}$ )	Mean bias (GC – insitu / insitu) (%)
Bremen	0.6	8.7	-18.9
Cologne	0.5	11	11.7

Dresden	0.56	9.2	-18.8
Dusseldorf	0.53	10.5	-15.7
Frankfurt	0.58	9.3	-37.4
Hamburg	0.67	8	-12.7
Hanover	0.59	7.9	-13.1
Leipzig	0.39	8.4	-28.6
Munich	0.5	8.5	-18.6
Stuttgart	0.53	8.6	-16.1

**Line 273:” The increase in OH radicals results from German metropolitan areas being in a NO<sub>x</sub> saturated regime”. From BAU to lockdown period, the meteorological condition changed, which could lead to higher temperature and higher solar radiation, and this has the potential to increase OH concentration. Hence, the influence of meteorological between different period in 2020 should be considered.**

Section 4.2 shows a comparison of 2020 (lockdown) and 2020 (no lockdown). The meteorology is the same in both cases; the only difference is in the emissions. As a result, the change in OH must be due to chemistry changes caused by changes in NO<sub>x</sub> emissions. We hope this clarifies your comment.

**Line 281:”However, higher night-time NO<sub>3</sub> levels result in higher nighttime HNO<sub>3</sub> production from N<sub>2</sub>O<sub>5</sub> hydrolysis, resulting in slightly lower night-time lockdown PM nitrate compared to BAU” According to Figure 4, the change of nighttime HNO<sub>3</sub> production from N<sub>2</sub>O<sub>5</sub> hydrolysis is small compared to that during daytime. In addition, both of the production and sink of HNO<sub>3</sub> should be considered to explain its influence on PM concentration.**

Thanks for pointing this out. We modified the sentence as follow,

Lines 310-312:	<i>However, higher night-time NO<sub>3</sub> levels result in relatively unchanged night-time HNO<sub>3</sub> production from N<sub>2</sub>O<sub>5</sub> hydrolysis, resulting in slightly lower night-time lockdown PM nitrate compared to BAU (Fig. 4 (b,e,f,g)).</i>
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Our GEOS-Chem simulation diagnostics does not allow us to extract the HNO<sub>3</sub> sink information directly. Deposition (dry and wet deposition) is the primary sink of HNO<sub>3</sub> in the troposphere. Since we discuss the difference between 2020 (lockdown) and 2020 (no lockdown), which have the same meteorology with different emission, we don't expect the rate of dry deposition (calculated based on different meteorological variables ([http://wiki.seas.harvard.edu/geos-chem/index.php/Dry\\_deposition](http://wiki.seas.harvard.edu/geos-chem/index.php/Dry_deposition)) and wet deposition to differ significantly.

### Specific comments:

The use of “emission accounted”, and “meteorology accounted” makes the discussion part puzzled. The authors are suggested to use more clear phases.

We agree with the reviewer that these words make the discussion a bit confusing sometimes. We discussed this with our other colleagues and chose these words, because they are more clear in terms of methodology than other words. However, we are open to choose if a reviewer is willing to suggest new words.

Figure 1: The part of “Ground-truth measurements” is misleading, it should contain the observations data from 2019 and 2020.

Thanks for the suggestion. We modified the figure 1, as you suggested.

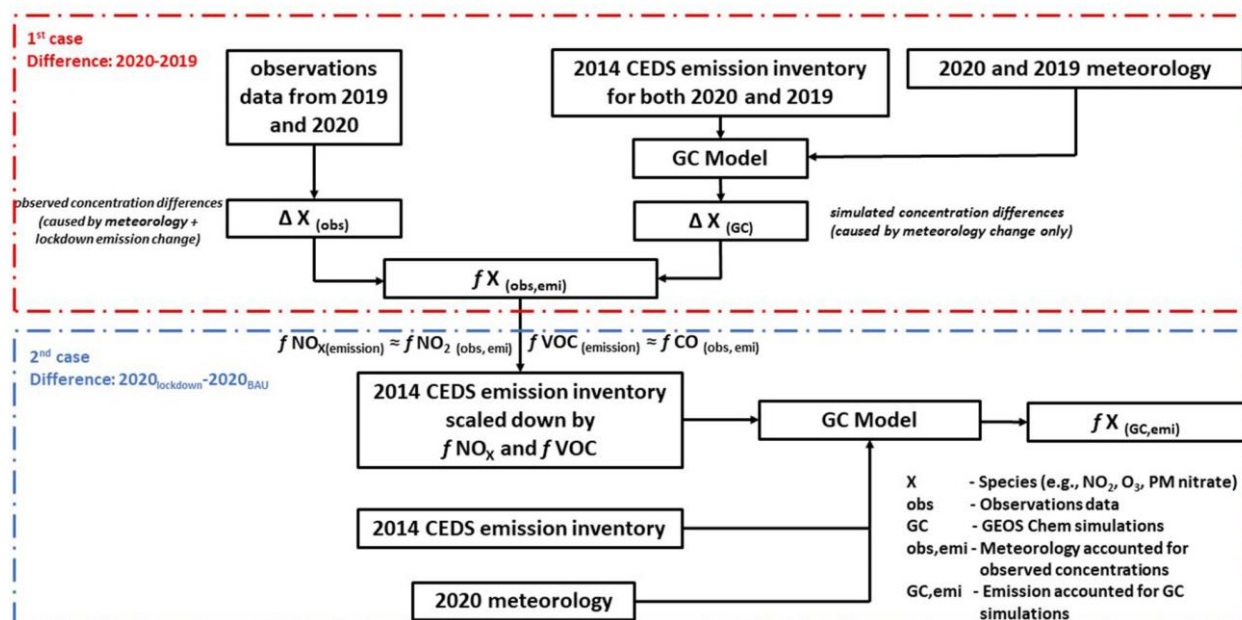


Figure 1. Schematic diagram of our methodology for calculating the meteorology accounted for observed pollutant concentrations changes between 2020 and 2019, and emission accounted for GC pollutant concentrations changes between 2020 lockdown and 2020 BAU scenario.

### References:

Cesari, D., De Benedetto, G., Bonasoni, P., Busetto, M., Dinoi, A., Merico, E., Chirizzi, D., Cristofanelli, P., Donato, A., Grasso, F., et al.: Seasonal variability of PM<sub>2.5</sub> and PM<sub>10</sub> composition and sources in an urban background site in Southern Italy, *Science of the Total Environment*, 612, 202–213, 2018.

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Salameh, D., Detournay, A., Pey, J., Pérez, N., Liguori, F., Saraga, D., Bove, M. C., Brotto, P., Cassola, F., Massabò, D., et al.: PM<sub>2.5</sub> chemical composition in five European Mediterranean cities: a 1-year study, *Atmospheric Research*, 155, 102–117, 2015.

Womack, C., McDuffie, E., Edwards, P., Bares, R., de Gouw, J., Docherty, K., Dubé, W., Fibiger, D., Franchin, A., Gilman, J., et al.: An odd oxygen framework for wintertime ammonium nitrate aerosol pollution in urban areas: NO<sub>x</sub> and VOC control as mitigation strategies, *Geophysical Research Letters*, 46, 4971–4979, 2019.

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# Secondary PM<sub>2.5</sub> decreases significantly less than NO<sub>2</sub> emission reductions during COVID lockdown in Germany

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**Abstract.** This study estimates the influence of anthropogenic emission reductions on the concentration of particulate matter with a diameter smaller than 2.5  $\mu\text{m}$  (PM<sub>2.5</sub>) during the 2020 lockdown period in German metropolitan areas. After accounting for meteorological effects, PM<sub>2.5</sub> 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 PM<sub>2.5</sub> concentrations were 19 % lower than in 2019. Meanwhile, meteorology accounted for NO<sub>2</sub> 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 SO<sub>2</sub> 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 anthropogenic NO<sub>x</sub> emission with unchanged anthropogenic VOC and SO<sub>2</sub>) are consistent with the small reductions of PM<sub>2.5</sub> during the lockdown and are used to identify the underlying drivers for this. Due to being in a NO<sub>x</sub> saturated ozone production regime, GC OH radical and O<sub>3</sub> concentrations increased (15 and 9 %, respectively) during the lockdown compared to a Business As Usual (no lockdown) scenario. O<sub>x</sub> (=NO<sub>2</sub>+O<sub>3</sub>) analysis implies that the increase in ozone at night-time is solely due to reduced NO titration. The increased O<sub>3</sub> results in increased NO<sub>3</sub> radical concentrations, primarily during the night, despite the large reductions in NO<sub>2</sub>. Thus, the oxidative capacity of the atmosphere is increased in all three important oxidants, OH, O<sub>3</sub>, and NO<sub>3</sub>. PM nitrate formation from gas-phase nitric acid (HNO<sub>3</sub>) is decreased during the lockdown as the increased OH concentration cannot compensate for the strong reductions in NO<sub>2</sub> resulting in decreased day-time HNO<sub>3</sub> formation from the OH + NO<sub>2</sub> reaction. However, night-time formation of PM nitrate from N<sub>2</sub>O<sub>5</sub> hydrolysis is relatively unchanged. This results from the fact that increased night-time O<sub>3</sub> results in significantly increased NO<sub>3</sub> which roughly balances the effect of the strong NO<sub>2</sub> reductions on N<sub>2</sub>O<sub>5</sub> formation. Ultimately, the only small observed decrease in lockdown PM<sub>2.5</sub> 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 PM<sub>2.5</sub> 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 NH<sub>3</sub> emissions, primarily emitted from livestock farming and intensive agricultural activities (fertilizer application), with high NH<sub>3</sub> concentrations in the early spring and summer months. Based on our findings, we suggest that appropriate NO<sub>x</sub> and VOC emission controls are required to limit ozone, and that should also

help reduce PM<sub>2.5</sub>. Regulation of NH<sub>3</sub> emissions, primarily from agricultural sectors, could result in significant reductions in PM<sub>2.5</sub> 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<sub>3</sub> and PM<sub>2.5</sub>), 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; Sourì 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  $\mu\text{m}$  are referred to as PM<sub>10</sub>, while particles smaller than 2.5  $\mu\text{m}$  AD are referred to as PM<sub>2.5</sub>. 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<sub>x</sub> (nitrogen oxides), SO<sub>2</sub> (sulfur dioxide), NH<sub>3</sub> (ammonia), VOCs (Volatile Organic Compounds) 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](#) (Cesari et al., 2018; Juda-Rezler et al., 2020; Samek et al., 2020; Salameh et al., 2015; Womack et al., 2019; Zhai et al., 2021).

In this study, we mainly focus on the response of urban surface  $\text{PM}_{2.5}$  to COVID-19 lockdown restrictions in Germany. Because major anthropogenic emissions are reduced, this unplanned intervention can test the understanding of the contribution of secondary  $\text{PM}_{2.5}$  sources, as well as the processes important in secondary  $\text{PM}_{2.5}$  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 ( $\text{NH}_3$ ) emissions (agricultural sources) are a significant source of  $\text{PM}_{2.5}$  in Germany in the spring (Fortems-Cheiney et al., 2016), when lockdown restrictions are implemented. Secondary inorganic aerosols such as ammonium sulfate and ammonium nitrate are the largest contributors to  $\text{PM}_{2.5}$  in Europe (Pay et al., 2012; Petetin et al., 2016). In comparison to sulfate formation, nitrate formation is more dependent on  $\text{NH}_3$  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  $\text{NH}_3$  in  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  are directly proportional to primary emission but secondary components of  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  concentrations. To this end, we used ground and space-based measurements of  $\text{PM}_{2.5}$ ,  $\text{NO}_2$ ,  $\text{O}_3$ ,  $\text{SO}_2$ ,  $\text{CO}$  and  $\text{NH}_3$  in conjunction with GEOS-Chem simulations to investigate the influence of lockdown restrictions on  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  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 ( $\text{NO}_x$ ) on  $\text{PM}_{2.5}$  formation (Sect. 4.2), as well as the role of ammonia ( $\text{NH}_3$ ) emissions in  $\text{PM}_{2.5}$  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 ( $\text{PM}_{2.5}$ ,  $\text{NO}_2$ ,  $\text{O}_3$ ) for all of these while  $\text{SO}_2$  data was only available for five of these areas (Bremen, Dresden, Frankfurt, Hamburg and Leipzig) and  $\text{CO}$  data was limited to six metropolitan areas (Bremen, Frankfurt, Hamburg, Hanover, Munich



**Table 1.** Data sets used in this study.

Data source	Data	Temporal resolution	Spatial resolution	Data availability
Governmental in-situ measurements	NO <sub>2</sub> , O <sub>3</sub> , PM <sub>2.5</sub>	1 h	-	Bremen, Cologne, Dresden, Dusseldorf, Frankfurt, Hamburg, Hanover, Leipzig, Munich and Stuttgart metropolitan areas
	SO <sub>2</sub>	1 h	-	Bremen, Dresden, Frankfurt, Hamburg and Leipzig metropolitan areas
	CO	1 h	-	Bremen, Frankfurt, Hamburg, Hanover, Munich and Stuttgart metropolitan areas
TROPOMI satellite measurements	SO <sub>2</sub>	daily	7*3.5 km (5.5*3.5 km, after August 6, 2019)	All of Germany
IASI satellite measurements	NH <sub>3</sub>	twice a day	12 km diameter	All of Germany
		monthly	1 degree	All of Germany
ERA 5 (ECMWF reanalysis)	Temperature, relative humidity, boundary layer height and wind speed	1 h	0.25 degree	All of Germany
		Precipitation	daily	1 degree
GEOS-Chem (GC) chemical transport model	All species	1 h	0.5 * 0.625 degree	All of Germany

and Stuttgart). We use data for 2019 and 2020 in this work (data-obtained from <https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm>).

TROPOMI tropospheric SO<sub>2</sub> (Theys et al., 2017) column products are also used (offline products-obtained from <https://s5phub.copernicus.eu>). The TROPOMI SO<sub>2</sub> product provides the total SO<sub>2</sub> 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<sub>2</sub> is calculated by averaging these values within 0.5-degree radius of the urban center.

The daily atmospheric NH<sub>3</sub> variability in Germany was studied using the “near-real time daily IASI/Metop-B ammonia (NH<sub>3</sub>) total column (ANNI-NH<sub>3</sub>-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<sub>3</sub> variability in Germany was studied using the “standard monthly IASI/Metop-B ULB-LATMOS ammonia (NH<sub>3</sub>) L3 product (total column)” dataset. This product contains a monthly averaged NH<sub>3</sub> 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  $0.5^{\circ} \times 0.625^{\circ}$  with dynamic boundary conditions generated from a global simulation with  $4^{\circ} \times 5^{\circ}$  resolution. We ran the GC simulation for two cases. In the first case, anthropogenic emissions from the 2014 CEDS inventory (Hoesly et al., 2018), the most recent version of which is 2014, 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 2014 CEDS 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. Even though the 2014 CEDS anthropogenic emission inventory is used in GC simulations, the effects of anthropogenic emission changes between 2014 and 2019 or 2020 will be canceled out because we use the difference between two years (e.g., 2020 - 2019) or two cases (e.g., 2020<sub>lockdown</sub> - 2020<sub>no lockdown</sub>) in our study.

### 125 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.

$$\Delta PM_{2.5(GC)} = PM_{2.5(GC,2020)} - PM_{2.5(GC,2019)} \quad (1)$$

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

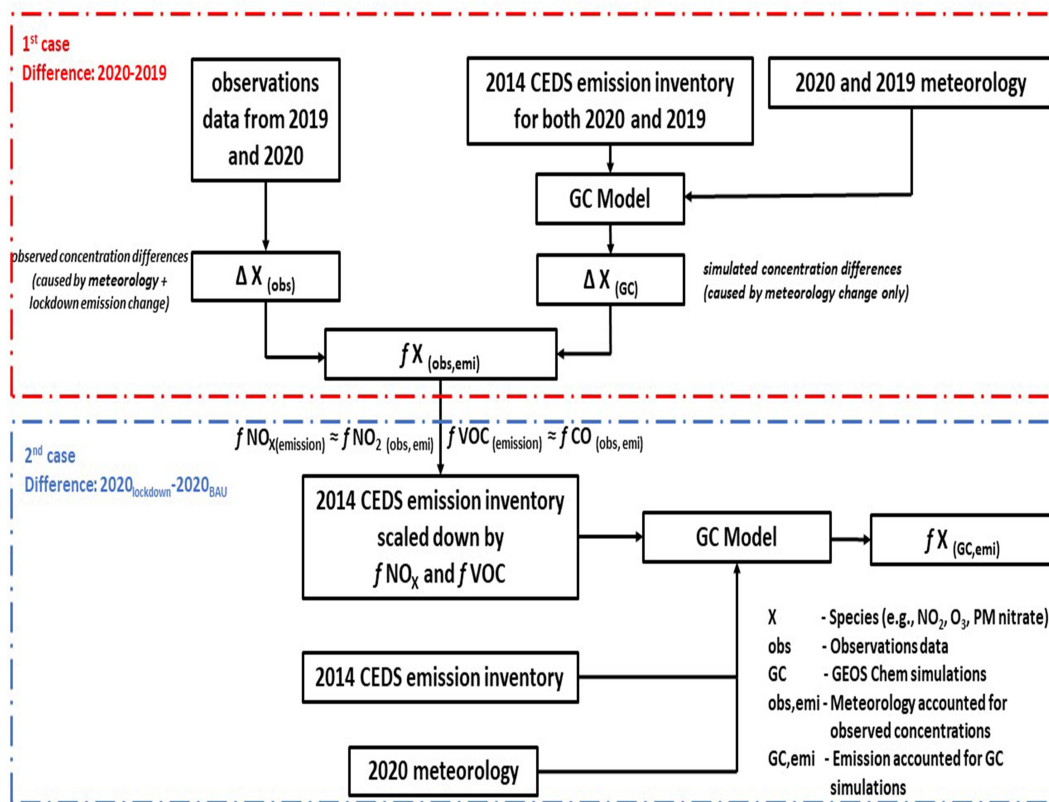
$$135 \quad \Delta PM_{2.5(obs)} = PM_{2.5(obs,2020)} - PM_{2.5(obs,2019)} \quad (2)$$

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.

$$\Delta PM_{2.5(obs,emi)} = \Delta PM_{2.5(obs)} - \Delta PM_{2.5(GC)} \quad (3)$$

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 fractional change (%) in pollutant concentration between 2020 and 2019 due to emission changes only, is calculated as,

$$140 \quad fPM_{2.5(obs,emi)} = \frac{\Delta PM_{2.5(obs,emi)}}{PM_{2.5(obs,2019)}} * 100 \quad (4)$$



**Figure 1.** Schematic diagram of our methodology for calculating the meteorology accounted for observed pollutant concentrations changes between 2020 and 2019, and emission accounted for GC pollutant concentrations changes between 2020 lockdown and 2020 BAU scenario.

where, “obs”, “GC” and “obs,emi” refer to ground-truth measurements ([observations data](#)), GEOS-Chem simulations and meteorology accounted for ground-truth measurements, respectively.

145 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<sub>2</sub> and O<sub>3</sub> concentration changes for eight German metropolitan areas. Here, we reproduce the results for NO<sub>2</sub> and O<sub>3</sub> concentrations, but for ten metropolitan areas. We use  $fNO_2_{(obs,emi)}$  and  $fCO_{(obs,emi)}$  to capture fractional changes in anthropogenic NO<sub>X</sub> and VOC emission ( $fNO_{X(emission)}$  and  $fVOC_{(emission)}$ ) due to lock down restrictions, respectively. Because of the scarcity of VOC

150 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<sub>2</sub> was used as proxy for NO<sub>X</sub>. This assumption is supported by studies such as Baker et al. (2008); Von Schneidmesser 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.

$$fNO_X(emission) \approx fNO_2(obs,emi) \quad (5)$$

$$fVOC(emission) \approx fCO(obs,emi) \quad (6)$$

The base anthropogenic emission inventory were then scaled down by  $fNO_X(emission)$  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,

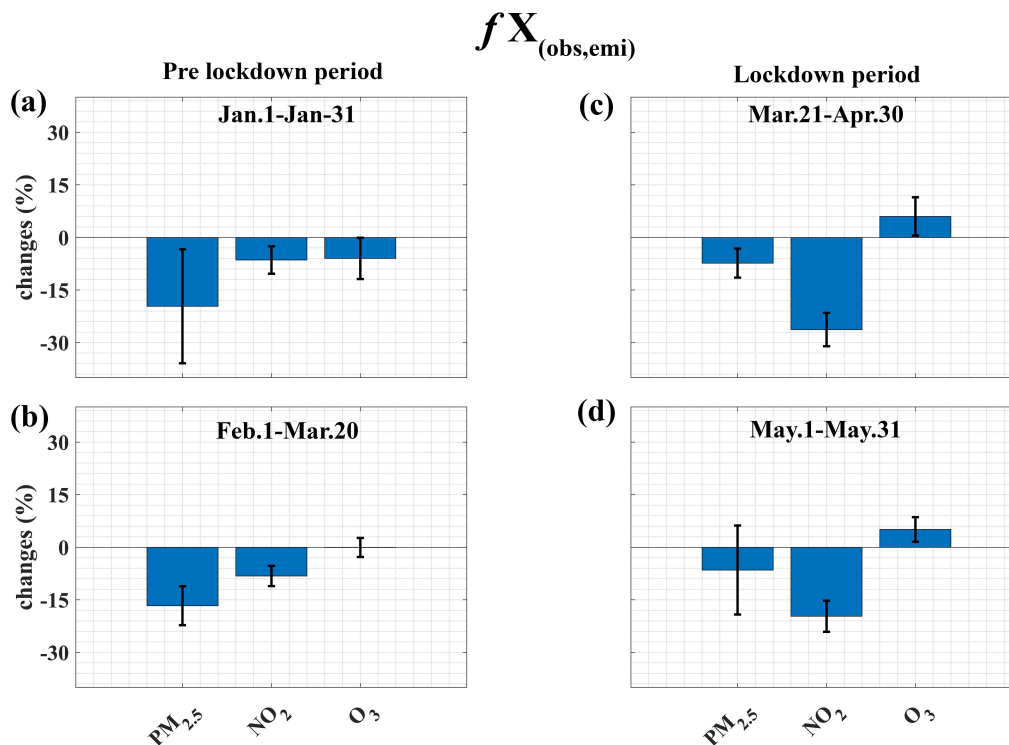
$$fPM_{2.5}(GC,emi) = \frac{PM_{2.5}(GC,2020,lock) - PM_{2.5}(GC,2020)}{PM_{2.5}(GC,2020)} * 100 \quad (7)$$

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). Table A1 shows the statistical evaluation (R, RMSE and mean bias) of the GC model performance for each 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)/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. We also compared the 2019 GC simulated nitrate and ammonium concentration for the urban measurement station in Germany (14.33°E,



**Figure 2.** Meteorology accounted for mean in-situ PM<sub>2.5</sub>, NO<sub>2</sub>, and O<sub>3</sub> 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  $1\sigma$  of mean of ten metropolitan areas.

51.75°N). The statistical evaluation (R, RMSE and mean bias) of the GC model performance for these species is given in Table

185 B1.

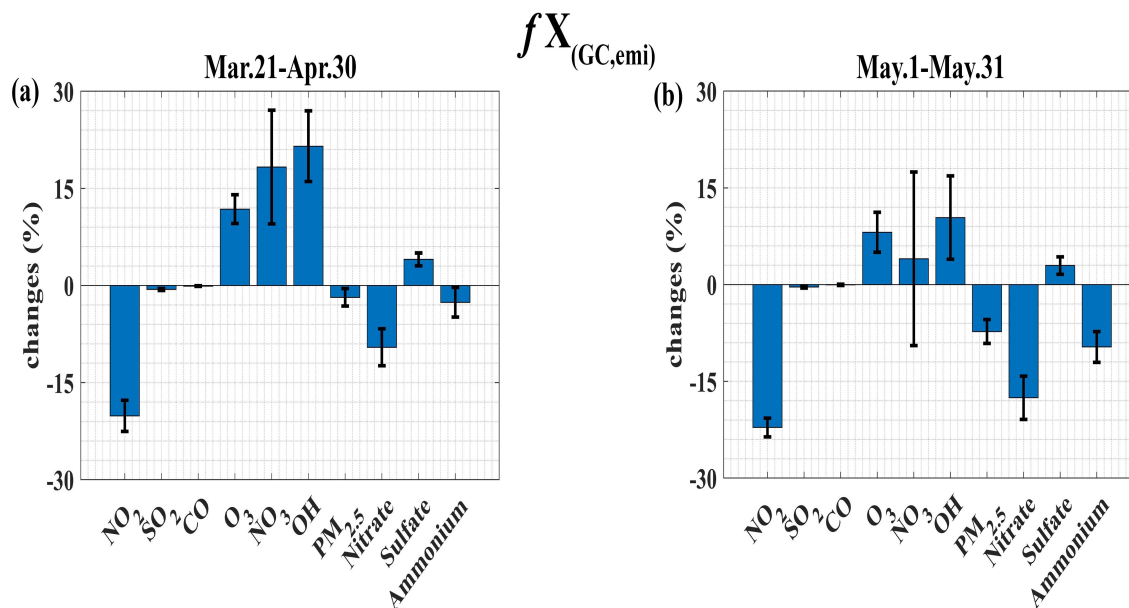
Figure 2 shows meteorology accounted for mean PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub> concentration changes between 2020 and 2019 for ten German metropolitan areas from January 1 through May 31. Both meteorology accounted and unaccounted for mean PM<sub>2.5</sub>, NO<sub>2</sub> and O<sub>3</sub> 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<sub>2</sub> and PM<sub>2.5</sub> 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 % ( $fNO_{2(obs)}$  and  $fPM_{2.5(obs)}$ ), respectively, due to

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the dilution/dispersion from the high wind conditions. However, after accounting for meteorology, the difference in mean  $\text{NO}_2$  and  $\text{PM}_{2.5}$  concentrations between 2020 and 2019 for the period February 1 to March 20 ( $f\text{NO}_{2(\text{obs},\text{emi})}$  and  $f\text{PM}_{2.5(\text{obs},\text{emi})}$ ) are 8 % and 18 %, respectively. This finding is consistent with meteorology accounted for mean  $\text{NO}_2$  and  $\text{PM}_{2.5}$  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  $\text{NO}_2$  and  $\text{PM}_{2.5}$  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  $\text{NO}_2$  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  $\text{PM}_{2.5}$  concentrations show a smaller reduction (5 %) compared to the same period in 2019, while an important precursor,  $\text{NO}_2$ , decreased by 23 % during the same period. Furthermore, the meteorology accounted for  $\text{PM}_{2.5}$  reduction during the 2020 lockdown period (5 %) is less than the meteorology accounted for  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  concentrations during the 2020 lockdown period are higher than in 2019. The meteorology accounted for mean  $\text{O}_3$  concentrations in the 2020 lockdown period are increased by 6 % compared to the same period in 2019. The increase in  $\text{O}_3$  concentration during the 2020 lockdown period is mainly due to being in a  $\text{NO}_X$  saturated regime (Gaubert et al., 2021), in which reducing  $\text{NO}_X$  emission results in an increase in  $\text{O}_3$  concentrations (Sillman, 1999; Sillman et al., 1990). It is also possible that the increase in ozone is due to less ozone destruction via lower NO titration, in addition to an increase in ozone formation efficiency through  $\text{NO}_X$  saturated regime chemistry. The meteorology accounted for mean  $\text{O}_X$  ( $= \text{NO}_2 + \text{O}_3$ ) concentrations in the 2020 lockdown period is 2 % higher than in 2019 (Fig. C1(a)), implies that the reduced NO titration effect partly contributed to the increased ozone.  $\text{O}_X$  analysis also implies that the decrease in  $\text{NO}_2$  was offset by an increase in  $\text{O}_3$ , and ozone production is overwhelmingly  $\text{NO}_X$  saturated in Germany.

The effects of lockdown restrictions on  $\text{SO}_2$  concentrations are insignificant. In comparison to 2019, TROPOMI meteorology accounted for  $\text{SO}_2$  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  $\text{SO}_2$  concentrations, but only for five metropolitan areas. Similarly, we found that the impact of lockdown restrictions on in-situ  $\text{SO}_2$  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 (SO<sub>2</sub>, 2021). Because the lockdown restrictions primarily reduced traffic-related emissions, we see far less effects of the lockdown on  $\text{SO}_2$  concentration (slight increase or no significant decrease in other European metropolitan areas (Collivignarelli et al., 2020; Filonchuk 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



**Figure 3.** The emission accounted for GC NO<sub>2</sub>, SO<sub>2</sub>, CO, O<sub>3</sub>, NO<sub>3</sub> radical, OH radical, PM<sub>2.5</sub>, inorganic nitrate, sulfate, ammonium concentration changes between 2020 lockdown and 2020 BAU (no lockdown) scenario ( $fX_{(GC,emi)}$ ). Error bars represent the 1  $\sigma$  of mean of ten metropolitan areas.

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

235 As mentioned in Sect. 3, we use the meteorology accounted for NO<sub>2</sub> and CO changes to adjust the anthropogenic NO<sub>X</sub> 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 anthropogenic NO<sub>X</sub> emission and unchanged anthropogenic VOC emis-  
 sions) for the 2020 lockdown period. The NO<sub>X</sub> emission reduction is within the range of estimated anthropogenic NO<sub>X</sub>  
 emission reductions using activity data for Europe by previous authors (Doumbia et al., 2021; Guevara et al., 2021) (25 % and  
 240 33 %, respectively). For those studies there are large differences in estimated anthropogenic VOC emission changes for Europe;  
 Doumbia et al. (2021) estimated 34 % while Guevara et al. (2021) estimated 8 % reduction in anthropogenic VOC emissions,  
compared to the BAU scenario. 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<sub>X</sub> saturated ozone production regime,  
 VOC emission reductions can decrease ozone levels, while NO<sub>X</sub> emission reductions increase them. Gaubert et al.  
 245 (2021) conducted a sensitivity study of modelling work on ozone levels in response to the NO<sub>X</sub> or VOC or both emission re-  
 ductions for the 2020 lockdown period. The reduction in both emissions (NO<sub>X</sub> 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  $\text{NO}_x$  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 anthropogenic VOC emissions were either not reduced at all or by a much smaller percentage than anthropogenic  $\text{NO}_x$  emissions, compared to the BAU scenario. According to the European Environment Agency (EEA) (<https://www.eea.europa.eu/data-and-maps/indicators/eea-32-non-methane-volatile-1/assessment-4>), the road transport sector accounts for 14.6 % of total NMVOC emissions, while the road transport sector accounts for 40.5 % of total  $\text{NO}_x$  emissions (<https://www.eea.europa.eu/data-and-maps/indicators/eea-32-nitrogen-oxides-nox-emissions-1/assessment.2010-08-19.0140149032-3>: :text=EEA%2D33%20emissions%20of%20nitrogen,households%20(13%25)%20sectors). According to Guevara et al. (2021), the transportation sector accounts for nearly 90 % of the reduction in total anthropogenic  $\text{NO}_x$  and VOC emissions during lockdown. As we noted that  $\text{NO}_x$  emission decreased by 23 %, and the lockdown restrictions primarily reduced traffic-related emissions, we can directly extrapolate this to a reduction in road transportation-related emissions; approximately 43 % ( $23 \cdot 40.5 / 40.5$ ). This finding also corresponds to a 40 % decrease in traffic vehicle count (Gensheimer et al., 2021). Therefore, the decrease in VOC emission from transport sector should be 6 % ( $14.6 \cdot 0.43$ ). However, due to a significant decline in the transport sector's VOC emission in recent years, this reduction in VOC emission from the transport sector, calculated based on the EEA's 2015 data, should be even less than 6 %. There is also no evidence that lockdown measures affect the major source of VOC emissions, which are use of volatile chemical products such as cleaning agents and personal care products, as well as biogenic emissions.

The emission accounted for GC lockdown  $\text{NO}_2$  concentrations decreased by 21 % ( $f_{\text{NO}_2(\text{GC},\text{emi})}$ ) while emission accounted for GC lockdown  $\text{O}_3$  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  $\text{NO}_x$  saturated ozone production regime in spring. However, the diurnal cycle of GC  $\text{O}_3$  changes between 2020 lockdown and BAU suggests that night-time ozone increases are solely due to a decrease in NO titration effects (Fig. C1(b)). 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  $\text{PM}_{2.5}$ , a significant drop in  $\text{NO}_2$  and marginal increase in  $\text{O}_3$  levels during 2020 lockdown period, compared to BAU levels, over Northern-Europe including Germany.

We investigated the GC  $\text{PM}_{2.5}$  composition for the studied period to determine the role of reduced  $\text{NO}_x$  emission on total  $\text{PM}_{2.5}$ . Major secondary  $\text{PM}_{2.5}$  components are nitrate, sulfate, ammonium and organic aerosol, which, on average, correspond to 24 %, 23 %, 15 % and 30 % of total  $\text{PM}_{2.5}$ , respectively, during March 21 to May 31, 2019 (Fig. D1). Mean relative contribution of  $\text{PM}_{2.5}$  species for 2020 (BAU) and 2020 (lockdown) are shown in Fig. E1 and F1, 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 ( $f_{\text{NIT}(\text{GC},\text{emi})}$ ) (Fig. 3 (a)), however, we see  $\text{NO}_2$  decreased by 21 % during the same period.



The decrease in emission accounted for GC PM nitrate is also less than the decrease in NO<sub>2</sub> 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. 285 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<sub>x</sub>, 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<sub>3</sub>), which partition to PM nitrate (Allen et al., 2015; Bauer et al., 2007). The reaction of OH and NO<sub>2</sub> (homogeneous pathway) and the hydrolysis of N<sub>2</sub>O<sub>5</sub> on aerosol 290 particles (heterogeneous pathway) are the two major pathways (Chang et al., 2011, 2016; Mollner et al., 2010).

The reaction for HNO<sub>3</sub> formation via gas-phase oxidation of NO<sub>2</sub> by OH is:



The reactions resulting in HNO<sub>3</sub> formation via hydrolysis of N<sub>2</sub>O<sub>5</sub> on aerosol surfaces are:



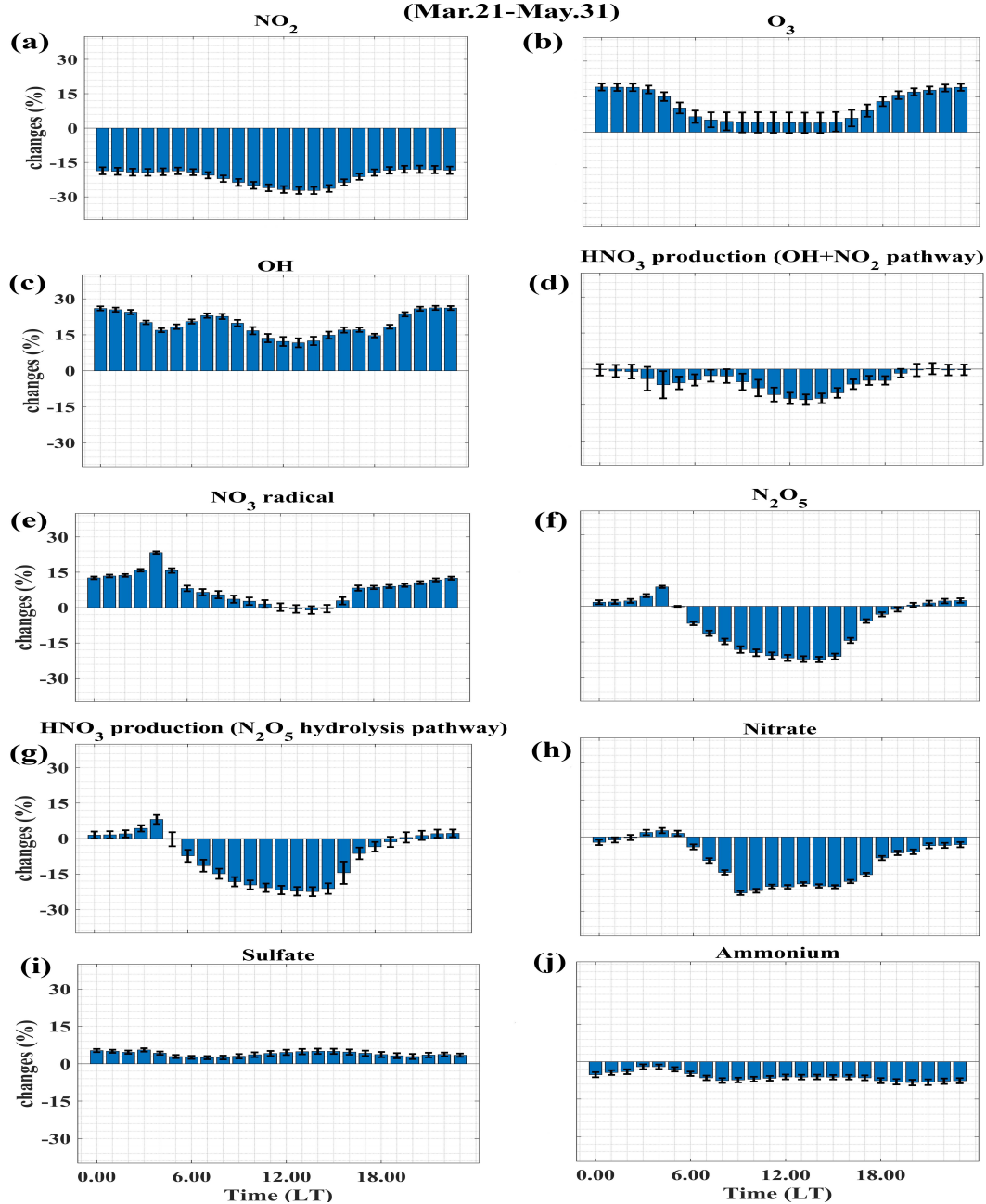
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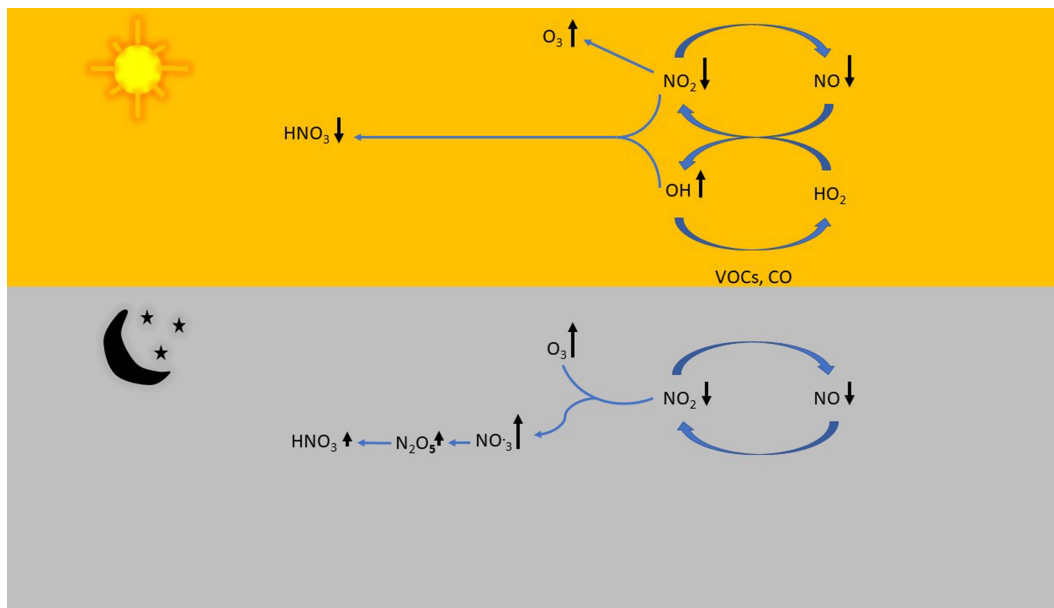
The formation of HNO<sub>3</sub> from the reaction of OH and NO<sub>2</sub> dominates during the day, while hydrolysis of N<sub>2</sub>O<sub>5</sub> on aerosol 300 particles dominates at night as OH night-time concentrations are low and N<sub>2</sub>O<sub>5</sub> photolyzes easily (Russell et al., 1986). At night, NO<sub>3</sub> 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<sub>3</sub>, 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<sub>x</sub> saturated regime (Shah et al., 2020). The 305 increase in GC lockdown NO<sub>3</sub> levels is predominantly at night due to a significant increase in night-time O<sub>3</sub> (Fig. 4 (b,e)); the reaction of NO<sub>2</sub> with O<sub>3</sub> is the most important source of NO<sub>3</sub> radical (Eq. R2) (Geyer et al., 2001).

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. 310 4 (h)). Even though GC lockdown OH levels increased, HNO<sub>3</sub> production from the OH+NO<sub>2</sub> reaction during the lockdown period is reduced due to significantly lower day-time NO<sub>2</sub> 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<sub>3</sub> levels result in higher night-time

$$fX_{(GC,emi)}$$



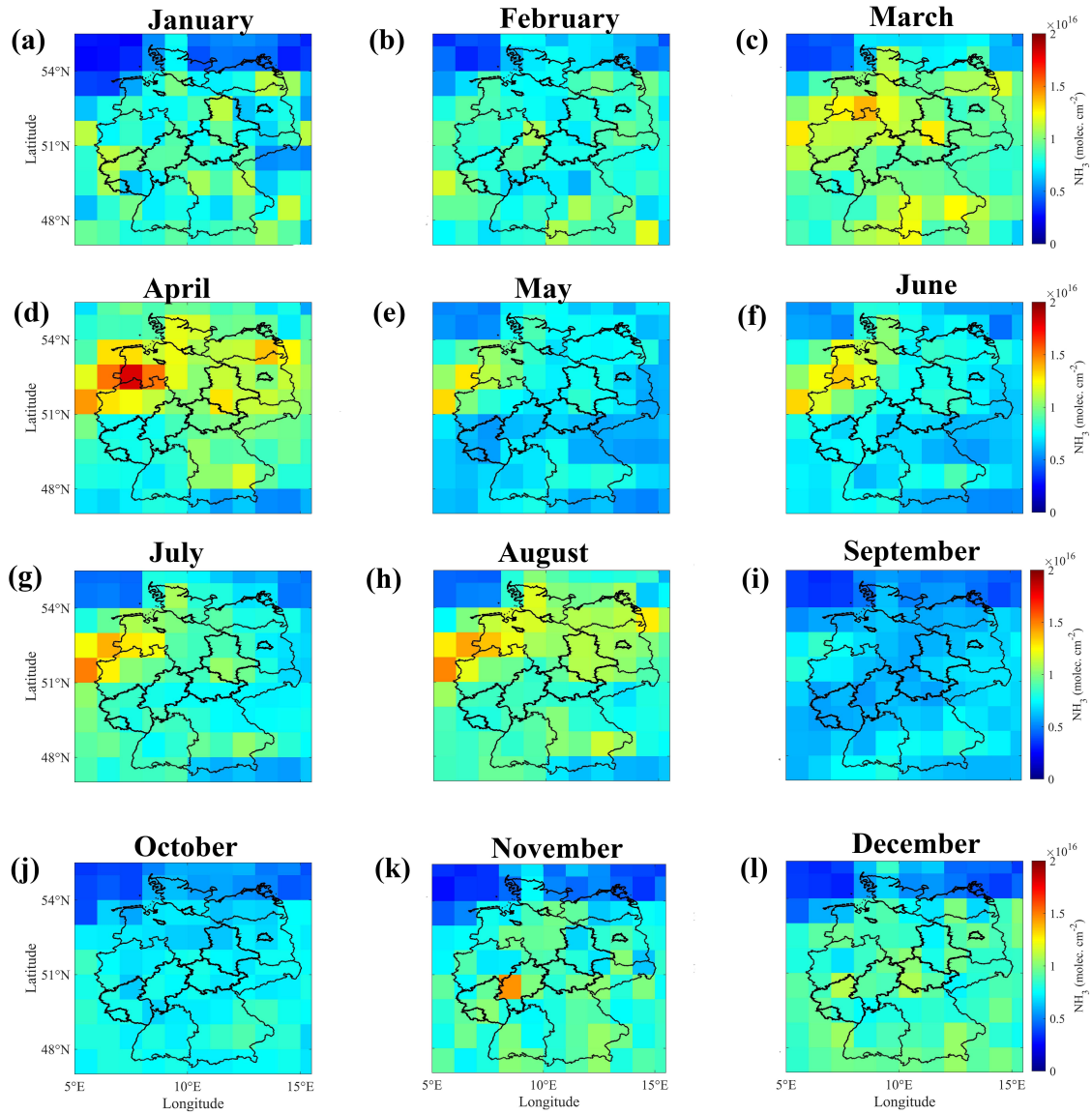
**Figure 4.** Diurnal cycle of emission accounted for GC NO<sub>2</sub>, O<sub>3</sub>, OH radical, HNO<sub>3</sub> production from oxidation of NO<sub>2</sub> by OH pathway, NO<sub>3</sub> radical, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub> production from N<sub>2</sub>O<sub>5</sub> hydrolysis pathway, PM nitrate, sulfate, ammonium concentration changes between 2020 lockdown and 2020 BAU (no lockdown) scenario ( $fX_{(GC,emi)}$ ). Error bars represent the standard error of respective hour in ten metropolitan areas.



**Figure 5.** Generalized schematic diagram of day and night-time lockdown  $\text{NO}_X$  chemistry compared to BAU scenario.

~~$\text{HNO}_3$  production from  $\text{N}_2\text{O}_5$  hydrolysis, resulting in slightly lower night-time lockdown PM nitrate compared to BAU (Fig. 4 (b,e,f,g))~~ However, higher night-time  $\text{NO}_3$  levels result in relatively unchanged night-time  $\text{HNO}_3$  production from  $\text{N}_2\text{O}_5$  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  $\text{NO}_3$  radical due to increased ozone partially offset the effect of reduced  $\text{NO}_X$  on nitrate formation. Previous studies have also shown that  $\text{N}_2\text{O}_5$  hydrolysis plays important role in nitrate formation than the gas-phase day-time pathway ( $\text{NO}_2 + \text{OH}$ ) (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  $\text{NO}_X$  chemistry compared to BAU scenario. The oxidation of  $\text{SO}_2$  is a major source of sulfate, and the reaction with the OH radical dominates the gas-phase oxidation of  $\text{SO}_2$  (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  $\text{SO}_2$  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  $\text{PM}_{2.5}$  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.

### IASI NH<sub>3</sub> total column (2014-2019)

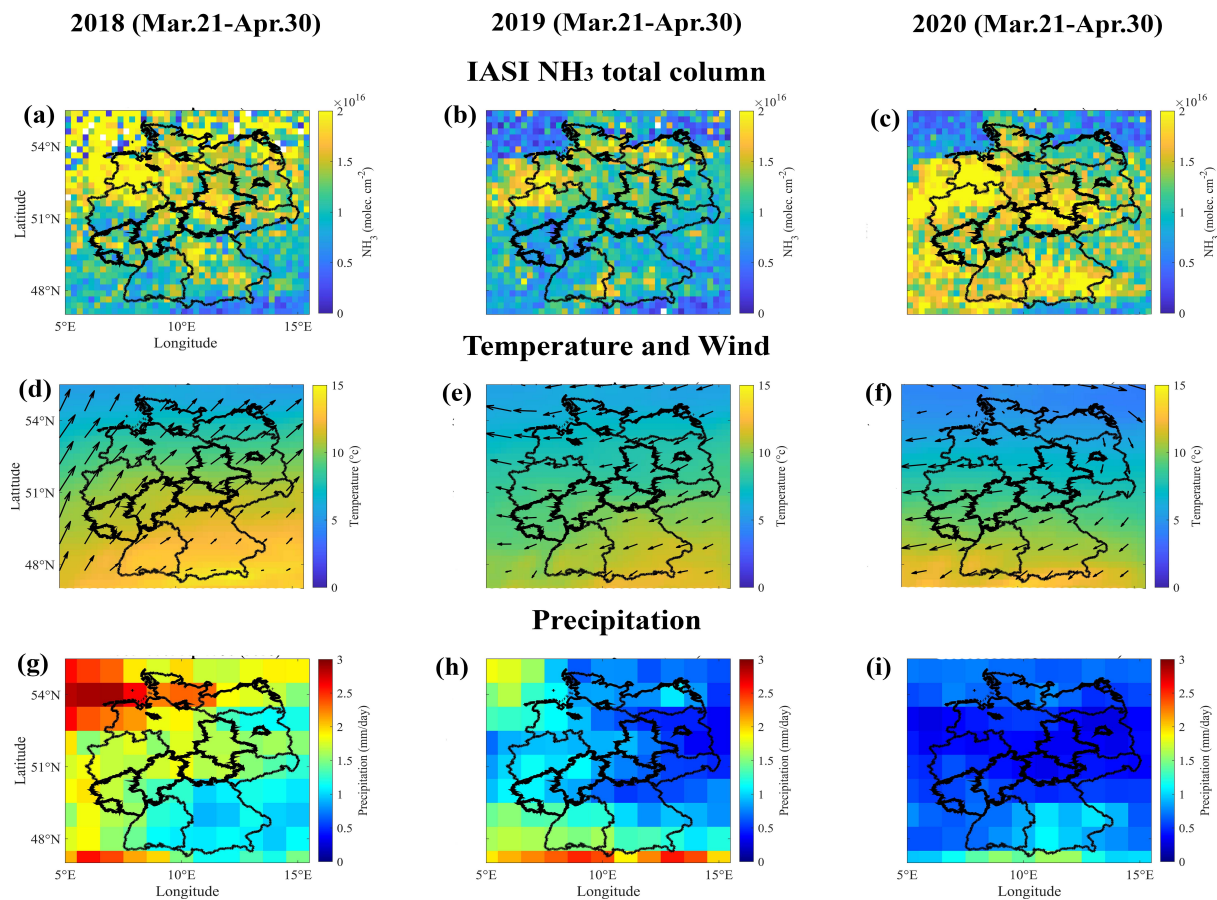


**Figure 6.** Monthly mean IASI NH<sub>3</sub> total column at 1\*1 degree resolution.

### 4.3 Link between spring PM<sub>2.5</sub> pollution episodes and high NH<sub>3</sub> concentrations

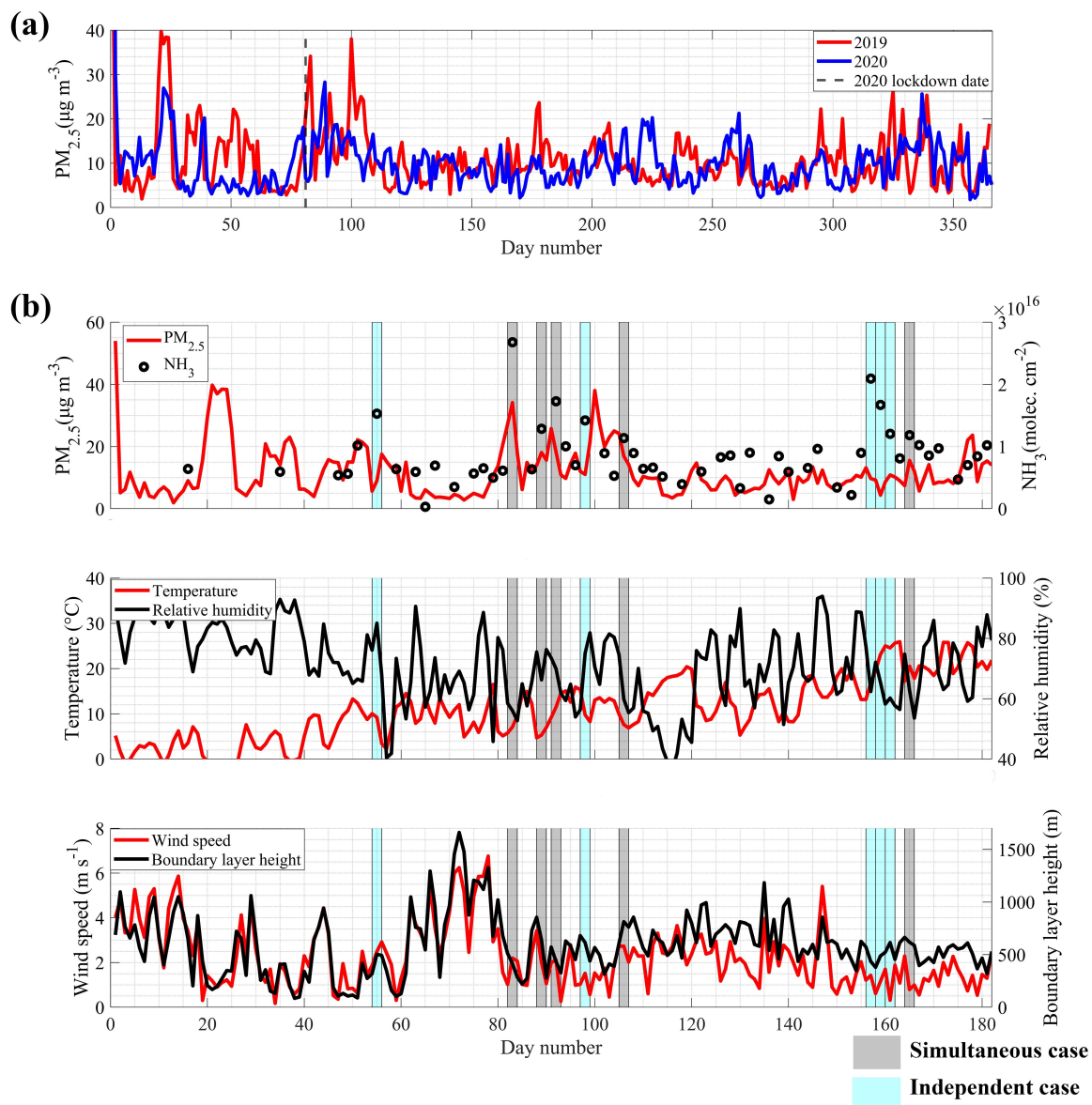
It is worth noting that a significant fraction of PM<sub>2.5</sub> is PM nitrate. Ammonia (NH<sub>3</sub>) is an important precursor for particulate nitrate formation (Ansari and Pandis, 1998; Banzhaf et al., 2013; Behera and Sharma, 2010; Wu et al., 2016). This explains the importance of monitoring and potentially regulating ammonia emissions. Therefore, the inter- and intra-annual changes in ammonia (NH<sub>3</sub>) concentrations over Germany, as well as their relationship to PM<sub>2.5</sub> variability, are reviewed and analyzed further below. In Germany, atmospheric NH<sub>3</sub> levels follow a monthly pattern, with NH<sub>3</sub> levels peaking in April (Fig. 6). NH<sub>3</sub> 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<sub>3</sub> values in summer are most likely due to warm climates (Kuttippurath et al., 2020). Monthly average NH<sub>3</sub> maps clearly show the high NH<sub>3</sub> 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<sub>3</sub> emissions in Europe (Webb et al., 2005). NH<sub>3</sub> 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<sub>3</sub> between 2018 and 2020 (Fig. 7). NH<sub>3</sub> 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<sub>3</sub> levels in 2020 are higher than in 2019 and 2018, possibly due to low precipitation. High temperatures promote NH<sub>3</sub> volatilization (increases the NH<sub>3</sub> level in the atmosphere) (Ernst and Massey, 1960), whereas high rainfall favors wet deposition (removal of atmospheric NH<sub>3</sub>). 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<sub>3</sub> 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<sub>2.5</sub> are apparent in German metropolitan areas in the early spring (from the second half of March to the end of April, e.g., Fig. 8 (a) for [Cologne](#)[Munich](#) metropolitan area). On March 21, 2020, the German government imposed COVID-19 lockdown restrictions. However, in-situ PM<sub>2.5</sub> concentrations during the initial lockdown period are higher than during the pre-lockdown period in 2020. High PM<sub>2.5</sub> 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<sub>2.5</sub> episodes are associated with high NH<sub>3</sub> concentrations (Fig. 6 (b)).~~ The high PM<sub>2.5</sub> 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<sub>3</sub> levels in early spring (April) and summer months. High PM<sub>2.5</sub> concentrations are evident in spring, however, we did not observe high PM<sub>2.5</sub> episodes in summer (Fig. 8 (a)). It is also worth noting that even in the spring and winter PM<sub>2.5</sub> is not consistently high on days with high NH<sub>3</sub>. This reflects the complexity of the process of gas to particle



**Figure 7.** Mean IASI NH<sub>3</sub> total column (daily IASI NH<sub>3</sub> measurements gridded at 0.25 degree resolution) (top), mean temperature and wind (middle) and mean precipitation (bottom).

conversion. Despite high NH<sub>3</sub> concentrations, ammonia(NH<sub>3</sub>)-to-ammonium(NH<sub>4</sub>) 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 °C and a high relative humidity of more than 70 % are optimal for atmospheric gas-phase NH<sub>3</sub> 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<sub>3</sub> on PM<sub>2.5</sub> 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<sub>3</sub> concentrations are not associated with high PM<sub>2.5</sub> in summer or late spring. Furthermore, it is important to note that PM<sub>2.5</sub> anthropogenic precursor emissions (NO<sub>x</sub>, SO<sub>2</sub>, VOCs) have a seasonal cycle, with higher emissions in winter than summer; however, biogenic VOC emissions dominate in the summer. To further demonstrate this for German metropolitan areas To further demonstrate the relationship between PM<sub>2.5</sub> and NH<sub>3</sub> for



**Figure 8.** 2019 and 2020 annual daily mean in-situ  $PM_{2.5}$  concentrations in Munich (a). In figure panel (a), the vertical dashed line denotes the start of 2020 lockdown. 2019 daily mean in-situ  $PM_{2.5}$  and column  $NH_3$  from IASI satellite (b, top). 2019 daily mean temperature and relative humidity (b, middle). 2019 daily mean wind speed and boundary layer height (b, bottom). The corresponding days for the cases “Simultaneous” are shaded with gray color and for the cases “Independent” are shaded with cyan color. “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.

German metropolitan areas, we consider two cases ("Simultaneous" and "Independent") for 2018 and 2019 (e.g., Fig. 8 (b) for Munich metropolitan area). "Simultaneous" - Simultaneous increase in  $\text{NH}_3$  (IASI) and  $\text{PM}_{2.5}$  (in-situ) concentrations on same day. "Independent" - Increase in  $\text{NH}_3$  (IASI) concentration not corresponding to an increase in  $\text{PM}_{2.5}$  (in-situ) concentration on same day. As an example, for the CologneMunich metropolitan area, the temperature and boundary layer height for the "Simultaneous" case ( $11.7 \pm 6.8 \text{ }^\circ\text{C}$  and  $500.4 \pm 166.5 \text{ m}$ , respectively) ( $14.8 \pm 8.3 \text{ }^\circ\text{C}$  and  $557.9 \pm 193.4 \text{ m}$ , respectively) is lower than for the "Independent" case ( $13.4 \pm 6 \text{ }^\circ\text{C}$  and  $628.9 \pm 274.3 \text{ m}$ , respectively) ( $15.5 \pm 5.4 \text{ }^\circ\text{C}$  and  $599.8 \pm 196.3 \text{ m}$ , 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))(Table C1). The regional differences are unsurprising, because other factors also influence the formation of  $\text{PM}_{2.5}$  from  $\text{NH}_3$  (e.g., other precursor concentrations such as  $\text{NO}_X$  and  $\text{SO}_X$ ). However, these findings support previous studies and imply that low temperature and low boundary layer height are most favorable for the formation of  $\text{PM}_{2.5}$  during the periods of high  $\text{NH}_3$ . GC also simulates the high spring  $\text{PM}_{2.5}$  concentrations that have been observed, with high ammonium ( $\text{NH}_4$ ) concentrations (Fig. 6 (e)).

## 385 5 Conclusions

Our study estimates the influence of anthropogenic emission reductions on  $\text{PM}_{2.5}$  concentration changes during the 2020 lockdown period in German metropolitan areas. Mean meteorology accounted for  $\text{PM}_{2.5}$  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  $\text{PM}_{2.5}$  concentrations are 19 % lower than in 2019. Meanwhile, meteorology accounted for  $\text{NO}_2$  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  $\text{SO}_2$  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  $\text{NO}_X$  emission with unchanged VOC and  $\text{SO}_2$ ) supports our findings of only a marginal decrease in  $\text{PM}_{2.5}$  and a significant decrease in  $\text{NO}_2$  levels. Due to being in a  $\text{NO}_X$  saturated ozone production regime, the GC lockdown OH and  $\text{O}_3$  concentrations increased by 15 % and 9 %, respectively, compared to BAU levels. However,  $\text{O}_X$  analysis suggest that the only increase in ozone during the day-time is due to increased ozone production efficiency via  $\text{NO}_X$  saturated regime chemistry, whereas the increase at night-time is due to decreased NO titration. Despite an increase in OH radicals, the GC lockdown PM nitrate formation decreased significantly during the day, due to a significant decrease in  $\text{NO}_2$ , compared to the BAU scenario. Increased night-time ozone, however, results in increased night-time  $\text{NO}_3$ , despite decreased  $\text{NO}_2$ , in turn, resulting in slightly increased night-time  $\text{N}_2\text{O}_5$  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  $\text{PM}_{2.5}$ . 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  $\text{PM}_{2.5}$  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  $\text{NO}_X$  and VOCs emissions should occur. Otherwise, ozone levels will rise as  $\text{NO}_X$  emissions drop, increasing oxidizing capacity, until a  $\text{NO}_X$  limited ozone production regime is reached. We also addressed the annual spring  $\text{PM}_{2.5}$  pollution episodes in German metropolitan areas, which are associated with high  $\text{NH}_3$  concentrations. North-West Germany is a hot-spot of  $\text{NH}_3$  emissions, primarily emitted from livestock farming and intensive agricultural activities (fertilizer application), with high  $\text{NH}_3$  concentrations in the early spring and summer months. Winter and spring meteorological conditions are more favorable for  $\text{PM}_{2.5}$  formation from  $\text{NH}_3$  than summer. Unsurprisingly, low temperature (and low boundary layer height) is shown to be a favorable meteorological condition for the formation of  $\text{PM}_{2.5}$  from  $\text{NH}_3$ . Regulation of  $\text{NH}_3$  emissions, primarily from agriculture, has the potential to reduce  $\text{PM}_{2.5}$  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  $\text{NO}_2$  concentration and changes in  $\text{NO}_X$  emission. This work also shows a direct relationship between changes in meteorology accounted for  $\text{SO}_2$  (and CO) concentration and changes in  $\text{SO}_X$  (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  $\text{PM}_{2.5}$  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. Organic aerosol accounts for nearly 30 % of total  $\text{PM}_{2.5}$ , which could be influenced by both primary/secondary biogenic and anthropogenic sources. However, our study is limited to examining the effects of  $\text{NO}_X$  emission changes on  $\text{PM}_{2.5}$  formation. Therefore, future studies on VOC emission changes on OA formation during high PM pollution episodes, particularly in the spring, will be more important in mitigating PM pollution.

*Data availability.* Hourly measurements of in-situ  $\text{NO}_2$ ,  $\text{O}_3$ ,  $\text{PM}_{2.5}$ ,  $\text{SO}_2$  and CO data are downloaded from (<https://discomap.eea.europa.eu/map/fme/AirQualityExport.htm>). The TROPOMI  $\text{SO}_2$  data are obtained from <https://s5phub.copernicus.eu/>. The IASI  $\text{NH}_3$  data are obtained from <https://iasi.aeris-data.fr/catalog/>. Hourly ERA5 meteorological data are available at <https://cds.climate.copernicus.eu/>.

**Table A1.** The statistical evaluation (R, RMSE and mean bias) of the GC model performance (PM<sub>2.5</sub>) for the 2019 study period (January 1 to May 31).

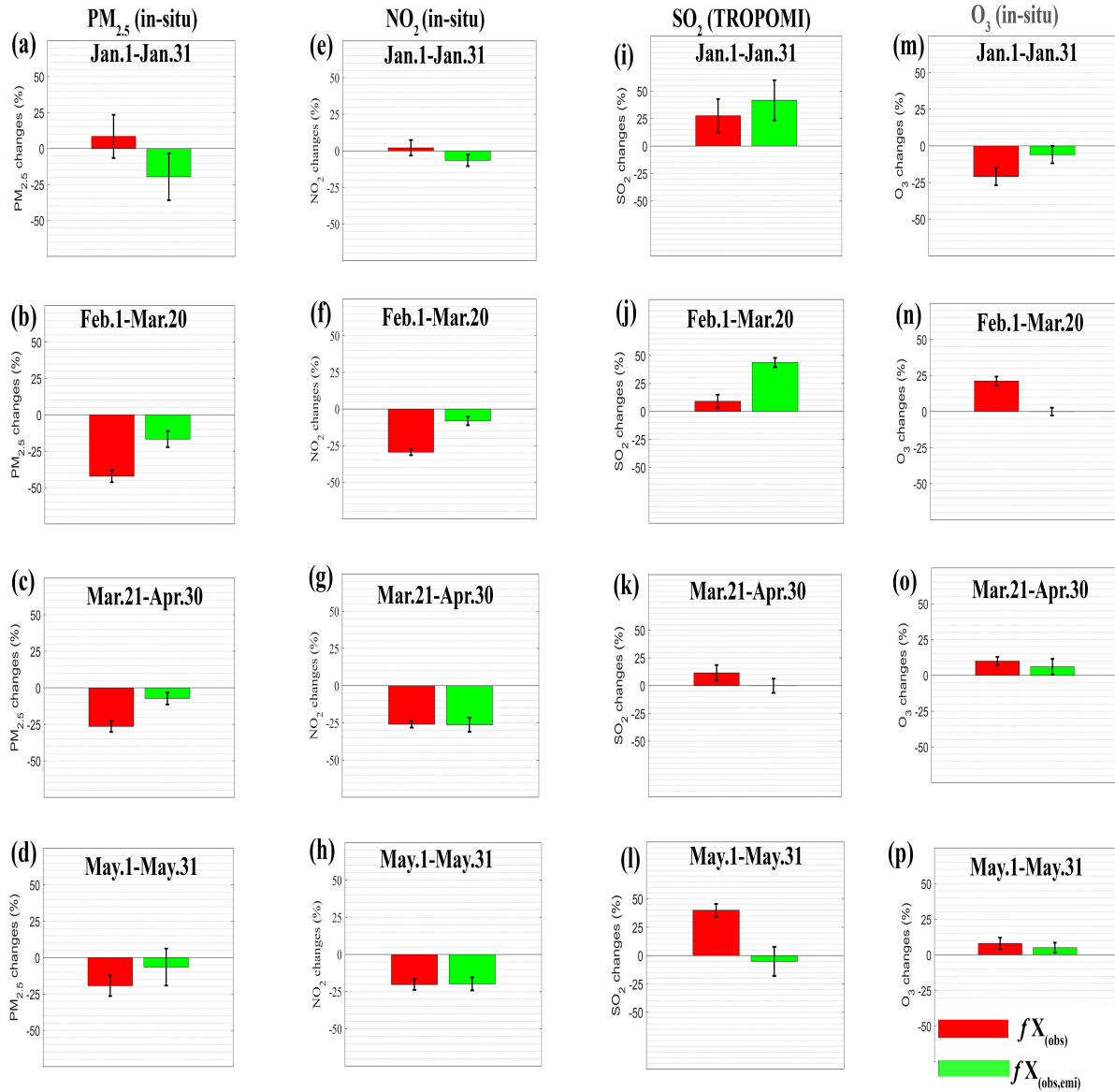
Metropolitan area	Correlation coefficient (R)	RMSE ( $\mu\text{g m}^{-3}$ )	Mean bias (GC – insitu / insitu) (%)
Bremen	0.6	8.7	-18.9
Cologne	0.5	11	11.7
Dresden	0.56	9.2	-18.8
Dusseldorf	0.53	10.5	-15.7
Frankfurt	0.58	9.3	-37.4
Hamburg	0.67	8	-12.7
Hanover	0.59	7.9	-13.1
Leipzig	0.39	8.4	-28.6
Munich	0.5	8.5	-18.6
Stuttgart	0.53	8.6	-16.1

**Table B1.** The statistical evaluation (R, RMSE and mean bias) of the GC model performance (nitrate and ammonium in PM<sub>2.5</sub>) for the 2019 study period (January 1 to May 31). For this comparison, data from the urban measurement station (14.33°E, 51.75°N) in Germany is used.

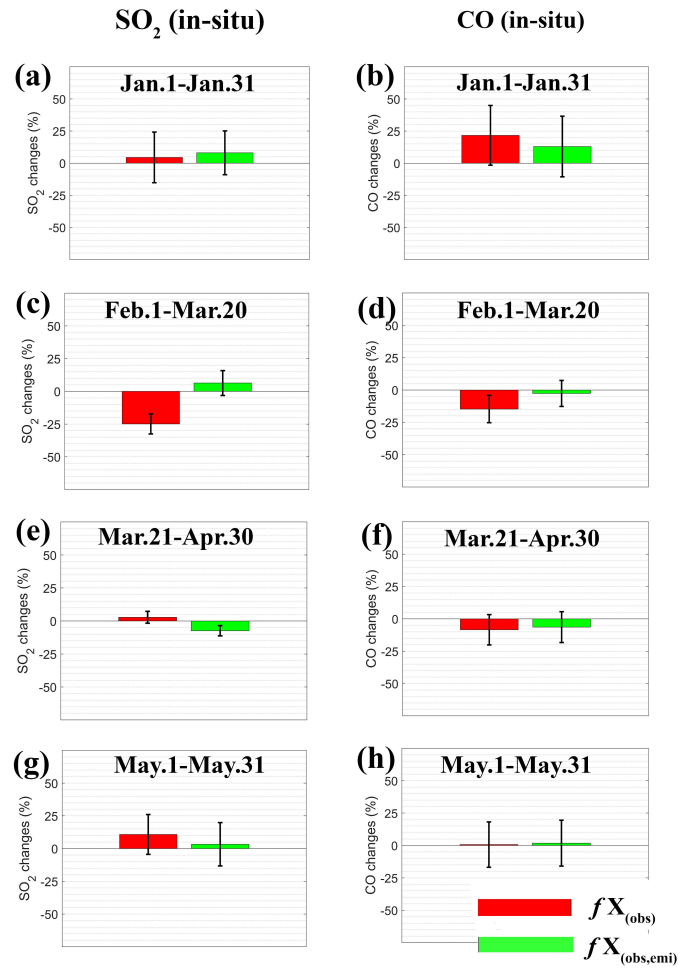
Species	Correlation coefficient (R)	RMSE ( $\mu\text{g m}^{-3}$ )	Mean bias (GC – insitu / insitu) (%)
Nitrate	0.51	2.33	-32.1
Ammonium	0.45	1.34	37

**Table C1.** The Statistical distribution of meteorological parameters for the cases “Independent” (each row top) and “Simultaneous” (each row bottom) in ten German metropolitan areas for 2018 and 2019.

Metropolitan area	Number of days	Wind speed (m/s)	Temperature (° C)	RH (%)	PBL height (m)
Bremen	17	4.3 ± 2.1	13.6 ± 5.8	62.3 ± 14.1	625.5 ± 211.1
	27	4.5 ± 2	11.5 ± 7	67.3 ± 16	541 ± 212.5
Cologne	16	3±2.2	13.4±6.1	74.3±11.4	628.9±274.31
	24	3.2±1.7	11.7±6.8	65.3±14.4	500.4±166.4
Dresden	24	1.9±1.1	14.9±6.9	68.6±12.8	578.9±220.7
	20	2.4±0.8	11.1±7.4	66.3±11	592.1±208.8
Dusseldorf	10	3.4±2.1	13.2±4.8	69±11.3	732.1±311.8
	30	3.4±1.8	13.5±5.6	66.2±13.5	494±168
Frankfurt	18	3.2±1.8	13.1±6.3	64.9±13.2	695.2±284.1
	21	2.2±1.1	13.1±6.6	63.6±13.6	442.8±194.5
Hamburg	14	5.4±2.5	13.7±6.5	57.5±11.8	705.3±249.2
	27	5.2±2.3	11.1±3.3	67.7±15	674.1±262
Hannover	14	3.2±2	14.2±7.8	62.5±10.4	697.5±210.2
	24	3.8±1.9	9.3±7.6	67.6±13.1	557.5±176.3
Leipzig	18	2.9±1.4	14.9±8	63.7±12.7	674.6±206.3
	30	3.4±1.6	11.2±7.1	61.9±10.8	532.3±227.3
Munich	26	2±1.1	15.5±5.4	71.5±12.3	599.8±196.3
	17	1.6±0.8	14.8±8.3	65.4±9.8	557.9±193.4
Stuttgart	22	1.9±0.9	13.8±6.4	71.7±11	600.7±234.9
	22	1.5±0.6	13.7±6.3	67.3±12.9	449±191.1

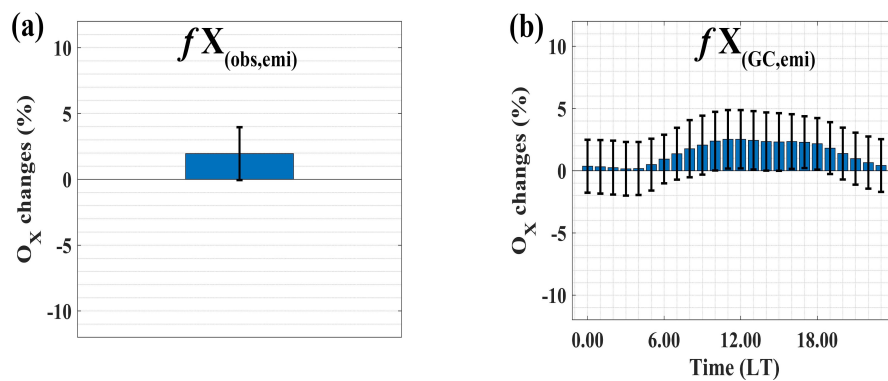


**Figure A1.** Meteorology unaccounted for (red) and meteorology accounted for (green) mean changes in PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub> and O<sub>3</sub> concentrations between 2020 and 2019 in ten German metropolitan areas. Error bars represent the 1  $\sigma$  of mean of ten metropolitan areas.



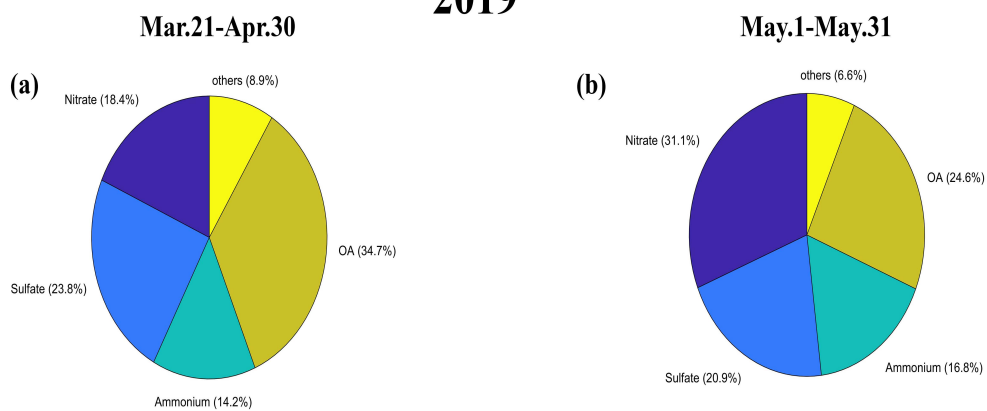
**Figure B1.** Meteorology unaccounted for (red) and meteorology accounted for (green) mean changes in in-situ SO<sub>2</sub> (Bremen, Dresden, Frankfurt, Hamburg and Leipzig) and in in-situ CO (Bremen, Frankfurt, Hamburg, Hanover, Munich and Stuttgart) between 2020 and 2019. Error bars represent the 1  $\sigma$  of mean of above mentioned metropolitan areas.

### Lockdown period (Mar.21-May.31)



**Figure C1.** Meteorology accounted for mean changes in in-situ  $O_X$  between 2020 and 2019 (left). Diurnal cycle of emission accounted for GC  $O_X$  concentration changes between 2020 lockdown and 2020 BAU (no lockdown) scenario (right). Error bars represent the 1  $\sigma$  of mean of ten metropolitan areas.

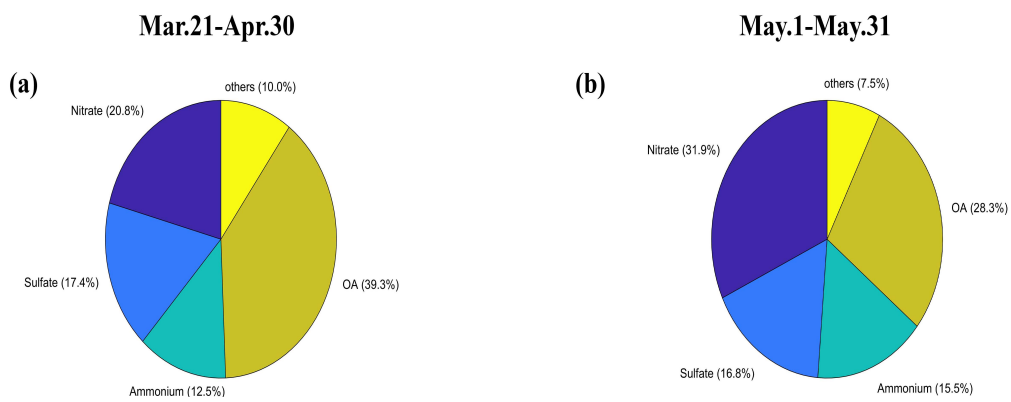
2019



**Figure D1.** Mean relative contributions of PM<sub>2.5</sub> species simulated by GC for 2019.

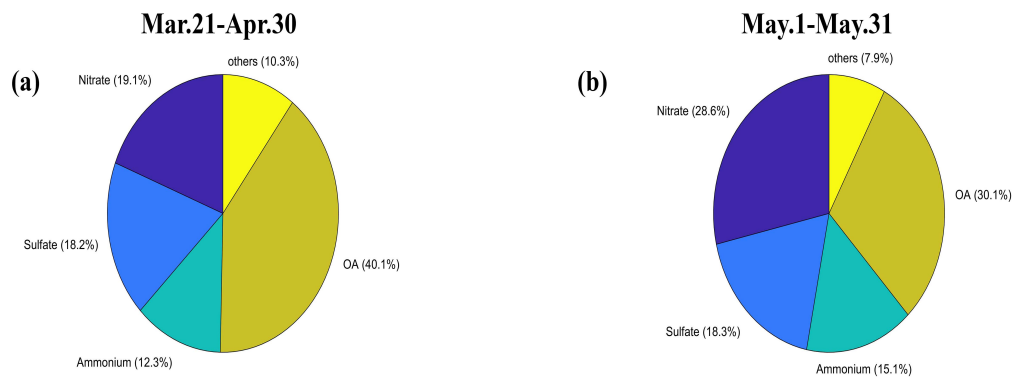


## 2020 (BAU)



**Figure E1.** Mean relative contributions of PM<sub>2.5</sub> species simulated by GC for 2020 (no lockdown).

## 2020 (lockdown)



**Figure F1.** Mean relative contributions of PM<sub>2.5</sub> species simulated by GC for 2020 (lockdown).

*Author contributions.* VB, and XB obtained the measurement data; ZQ performed the modeling work; VB analyzed the data and wrote the manuscript draft; JC, and FK supervised the work and edited the manuscript.

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

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