Aggravated surface O₃ pollution primarily driven by meteorological variation in China during the early COVID-19 pandemic lockdown period

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Abstract
Due to the lockdown during the COVID-19 pandemic in China from late January to early April in 2020, a significant reduction of primary air pollutants has been identified by satellite and ground observations. However, this reduction is in contrast with the increase of surface O\textsubscript{3} concentration in many parts of China during the same period. The reasons for this contrast are studied here from two perspectives: emission changes and inter-annual meteorological variations. Based on top-down constraints of NOx emissions from TROPOMI measurements and GEOS-Chem model simulations, our analysis reveals that NOx and volatile organic compound (VOC) emission reductions as well as meteorological variations lead to 8\%, -3\%, and 1\% changes in O\textsubscript{3} over North China, respectively. In South China, however, we find that meteorological variations cause ~30\% increases in O\textsubscript{3}, which is much larger than -1\% and 2\% changes due to VOC and NOx emission reductions, respectively, and the overall O\textsubscript{3} increase is consistent with the surface observations. The higher temperature is the main reason that leads to the surface O\textsubscript{3} increase in South China. Overall, inter-annual meteorological variations have a larger impact than emission reductions on the aggravated surface O\textsubscript{3} pollution in China during the early lockdown period of COVID-19 pandemic.

1. Introduction
Surface ozone (O\textsubscript{3}), an important air pollutant that is harmful to human health (Jerrett et al., 2009) and stomatal conductance of green vegetations (Gong et al., 2020), is produced by photochemical reactions of nitrogen oxides (NOx) and volatile organic compounds (VOC) (Liu et al., 1987; Sillman et al., 1990). In addition to emissions, meteorological conditions, such as temperature, solar radiation and relative humidity, also have large impacts on surface O\textsubscript{3} formation (Lu et al., 2019).

Ground observations show that surface O\textsubscript{3} increased dramatically during the COVID-19 lockdown period in China by around 40\% on average (Tong et al., 2023) and even larger than 100\% (Shi and Brasseur, 2020; Liu et al., 2021) depending on the time period and region. The reduction of economic activities during the lockdown period led to a significant decrease of several primary air pollutants emissions. The NO\textsubscript{2} vertical column density (VCD) from satellite measurements and surface NO\textsubscript{2} concentration from ground measurements were reduced by 40\% - 60\% in China during the lockdown period (Bauwens et al., 2020; Shi and Brasseur, 2020; Liu et al., 2020a; Zhang...
et al., 2020). A lower but discernible reduction of sulfur dioxide (SO$_2$), carbon monoxide (CO), and formaldehyde (HCHO) have also been identified by satellite or ground-based observations in China (Shi and Brasseur, 2020; Levelt et al., 2022; Ghahremanloo et al., 2021). However, during this period surface O$_3$ concentrations increased, and the respective roles of meteorological factor and emission reduction for the aggravated surface O$_3$ pollution during the lockdown in China need to be further quantified.

This study provides a quantitative analysis of the causes for the unexpectedly aggravated surface O$_3$ pollution in China during the early lockdown period of the pandemic from two perspectives using GEOS-Chem model. One is anthropogenic emission reduction of NOx and VOC in response to the lockdown possibly under a VOC-limiting chemical regime of surface O$_3$ production (Guo et al., 2023), while the other is the impact of natural variability of meteorological conditions. Previous studies have reported the enhanced surface O$_3$ due to NOx emission decline during the lockdown period in North China using chemical transport model (CTM) simulations without controlling for the impacts of meteorological variability (Zhang et al., 2021; Huang et al., 2020; Miyazaki et al., 2020). Other studies quantified or excluded the meteorological impacts on the surface O$_3$ using statistical analysis instead of CTM that account for the physical and chemical processes (Venter et al., 2020; Bi et al., 2022; Tong et al., 2023). Although a few studies have investigated the contributions from both emission reduction and meteorological variability to surface O$_3$ increase using CTMs, most of their results have uncertainties due to the limitations of their analysis. For example, some of them keep the emissions unchanged (Zhao et al., 2020) or assume an arbitrarily uniform emission reduction instead of constraining the emission based on observations (Le et al., 2020; Liu et al., 2021). In cases where the emissions were constrained by the observations, the focus was limited to several cities in China (Liu et al., 2020b). Furthermore, in the past studies, the surface O$_3$ increase during the lockdown period of 2-4 weeks is quantified in reference to the time period right before the lockdown instead of the same period in previous years; such comparisons by design cannot exclude the possibility that the seasonal variation of meteorology from early January to early April may have dominated the cause for the surface O$_3$ increase. A comprehensive analysis of the contributions from emission reductions and meteorological variations to the surface O$_3$ increase during the first round of the lockdown period with respect to the same time period in previous years in China is therefore overdue.
Here, we apply a top-down method to update NOx and VOC emission in February and March in 2020 based on the TROPOMI NO$_2$ and formaldehyde (HCHO) product. A set of GEOS-Chem model simulations with NOx and VOC emissions and meteorological fields in different time periods are then conducted. Based on the difference in surface O$_3$ concentration in different modeling sensitivity experiments, we quantitatively assess the respective roles of emission and meteorology in regulating surface O$_3$ concentration in continental China. The ground observations of surface O$_3$ and NO$_2$ concentration are compared with the model simulations to verify our analysis. Section 2 introduces the satellite and ground-based measurements, NOx emission update scheme, and the configurations of GEOS-Chem simulation experiments. Section 3 provides an evaluation of the constrained NOx emission and surface O$_3$ simulations. The analysis of the mechanism of the aggravated surface O$_3$ pollution is presented in Section 4, followed by the summary and conclusions in Section 5.

2. Datasets and Methods

2.1 TROPOMI NO$_2$ and HCHO product

We used tropospheric NO$_2$ and HCHO level 2 VCD product provided by the Tropospheric Monitoring Instrument (TROPOMI) onboard the Sentinel-5 Precursor (S5P) satellite (Veefkind et al., 2012). S5P is a sun-synchronous polar orbit satellite launched on 13 October 2017, which covers the near-global domain in a single day. TROPOMI provides NO$_2$ and HCHO retrievals at an approximately 7 km x 3.5 km spatial resolution (5.5 km x 3.5 km since 6 August 2019) from the ascending orbit with an equatorial crossing time of ~13:30 local time (Van Geffen et al., 2020; De Smedt et al., 2018). The datasets were obtained from the NASA Goddard Earth Sciences Data and Information Services Center (https://daac.gsfc.nasa.gov). A quality control procedure similar to Bauwens et al. (2020) but with slightly stricter criteria is adopted for TROPOMI NO$_2$ and HCHO data. The TROPOMI retrievals under one or more than one of the following conditions are screened out for data quality control. (1) Quality assurance value is no larger than 0.5; (2) cloud radiance fraction within NO$_2$ or HCHO retrieval window is larger than 0.3; (3) solar zenith angle is larger than 70°; and (4) viewing zenith angle is larger than 70°.

2.2 Ground O$_3$ and NO$_2$ measurements
Surface measurements of O\textsubscript{3} and NO\textsubscript{2} were collected from ~1600 operational air quality monitoring stations over the mainland China managed by the China National Environmental Monitoring Center (http://www.cnemc.cn/en/). We calculated daily maximum 8-hour average (MDA8) O\textsubscript{3} concentration from hourly in situ measurements. Surface O\textsubscript{3} are measured by ultraviolet photometric method and Indigo disulfonate spectrophotometry, following the national environmental standards of HJ 590-2010 and HJ 504-2009. Surface NO\textsubscript{2} concentrations are measured by the chemiluminescence method (Zhang and Cao, 2015), which can cause a positive bias in the NO\textsubscript{2} measurements (Steinbacher et al., 2007). The true NO\textsubscript{2} concentrations only account for 43%-76% and 70%-83% of measured values for rural and urban sites (Steinbacher et al., 2007). Following Wang et al. (2020b), we also applied a correction factor but with a lower value of 0.75 to the measured NO\textsubscript{2}, considering that we included both rural and urban sites. The sampling ports are placed at 3 to 15 meters above the ground following the national environmental monitoring method standard HJ 664-2013. The measured data are reported in the unit of \(\mu g\ m^{-3}\) under standard temperature (273.15 K) and pressure (101.325 kPa) according to national environmental standards GB 3095-2012.

2.3 GEOS-Chem model and its adjoint

The global 3-D chemical transport model GEOS-Chem (Bey et al., 2001) version 12.7.2 is used here. We apply the nested-grid version of GEOS-Chem (Chen et al., 2009; Wang et al., 2004) with the horizontal resolution of 0.25\(^\circ\times\)0.3125\(^\circ\) and 47 vertical hybrid-sigma levels over East Asia (70\(^\circ\)E-140\(^\circ\)E, 15\(^\circ\)N-55\(^\circ\)N). The boundary conditions are obtained from the 2\(^\circ\)×2.5\(^\circ\) global simulation. The model is driven by the GEOS-FP meteorological field provided by NASA Global Modeling and Assimilation Office (GMAO). A detailed O\textsubscript{3}-NO\textsubscript{x}-hydrocarbon chemistry (Mao et al., 2010; Mao et al., 2013; Travis et al., 2016) is included in the GEOS-Chem model. The altitude of the surface O\textsubscript{3} output from GEOS-Chem is specified at 9 meters above the ground to match the in-situ measurements (Travis et al., 2017; Zhang et al., 2012). Through our sensitivity test using GEOS-Chem, the variation of surface O\textsubscript{3} from 3 to 9 meters above the surface is generally less than 0.723 ppb (75th percentile), and the median bias is 0.283 ppb. Travis et al. (2017) reported from 60 m to 10 m above the ground, the MDA8 O\textsubscript{3} could decrease by ~3 ppb. Therefore, when comparing GEOS-Chem surface O\textsubscript{3} with in-situ measurements, the differences caused by inconsistent reported altitudes (9 m versus 3-15 m) can be ignored.
The global anthropogenic emission used in GEOS-Chem model is the Community Emissions Data System (CEDS) inventory (Hoesly et al., 2018), which is replaced by the MIX inventory (Li et al., 2017) over the Asian region. Biogenic emissions for VOCs follows the Model of Emissions of Gases and Aerosols from Nature (MEGAN) inventory (Guenther et al., 2012). Natural NOx emissions includes biomass burning from GFED4 inventory (Van Der Werf et al., 2017), soil NOx emissions (Hudman et al., 2012) and lightning sources (Murray et al., 2012; Ott et al., 2010).

The adjoint of the GEOS-Chem model (Henze et al., 2007; Henze et al., 2009) is a component of the 4D-Var inversion method that can efficiently optimize spatially disaggregated aerosol and gas emissions. This is done through iterative minimization of a cost function using the model adjoint to calculate the gradient of the cost function with respect to a large number of model parameters (such as anthropogenic NOx emissions in each grid box) simultaneously. The cost function is the sum of the error weighted difference between forward model outputs and observations and the divergence of posterior model parameters from the prior estimate (Section 2.4). We developed and validated the observation operator for TROPOMI NO\textsubscript{2} in the GEOS-Chem adjoint model version 35n similar to Wang et al. (2020a) and used it to optimize the anthropogenic NOx emission during the lockdown period in China. The NOx emission optimization is implemented using the 4D-Var method with GEOS-Chem adjoint at the nested grid with the resolution of 0.25°×0.3125°. The prior anthropogenic NOx emission used in the GEOS-Chem adjoint is HTAP version 2 (Janssens-Maenhout et al., 2015), which is equivalent to the MIX inventory in East Asia (Li et al., 2017).

2.4 NOx and VOC emission updates

Two approaches are used to update the emissions during the lockdown period in 2020. The first is a simple mass balance approach (Leue et al., 2001; Martin et al., 2003; Vinken et al., 2014) for updating the NOx emission by assuming a constant NOx lifetime and NOx/NO\textsubscript{2} ratio. In the period from 2010 to 2019, the anthropogenic NOx emissions have declined significantly as a result of the clean air actions of Chinese government (Zheng et al., 2018). We scale the anthropogenic NOx emission from year 2010 to 2019 using the spatially gridded ratio of mean TROPOMI tropospheric NO\textsubscript{2} VCD in Feb.-Mar. 2019 to GEOS-Chem simulated NO\textsubscript{2} column with default MIX 2010 emission (Appendix A), to obtain the baseline anthropogenic NOx emission in 2019,
which is denoted as MIX 2019. To derive anthropogenic NOx emissions in 2020 in China during the COVID-19 lockdown (MIX 2020), the spatially gridded ratio of mean TROPOMI tropospheric NO2 VCD in 2020 Feb.-Mar. to that in 2019 Feb.-Mar. is taken as a scaling factor for the updated baseline anthropogenic NOx emission in 2019 (MIX 2019). The two-month mean of TROPOMI NO2 VCD in 2019 and 2020 are calculated with the physical oversampling procedure (Sun et al., 2018). Scaling factors in regions where mean TROPOMI tropospheric NO2 VCD in 2019 Feb.-Mar. is less than 0.1 Dobson unit (DU) are set to 1 for emission updates in both 2020 and 2019, assuming that the lockdown only affects the populated areas (that have high NO2 in 2019).

The second method for updating NOx emission is 4D-Var via the GEOS-Chem adjoint model. The anthropogenic NOx emissions in 2020 lockdown period derived from the GEOS-Chem adjoint is denoted as 2020 Adjoint. Following Wang et al. (2020a), the cost function \( J \) for optimizing the NOx emission is defined as

\[
J = \frac{1}{2} \sum_{c \in \Omega} [H(c) - s]^T S_{\text{obs}}^{-1} [H(c) - s] + \frac{1}{2} \gamma (\sigma - \sigma_a)^T S_a^{-1} (\sigma - \sigma_a)
\]

where \( s \) is the tropospheric slant column density of TROPOMI NO2, which is the product of TROPOMI NO2 VCD and air mass factor. \( H \) is the TROPOMI NO2 observation operator that maps the modeled NO2 concentrations \( c \) to the observations in time and space and calculates the corresponding slant column density to make an apple-to-apple comparison of the model to TROPOMI. \( \Omega \) is the spatial and temporal domain where both model simulations and observations are available. \( \sigma \) is the scaling factor of anthropogenic NOx emissions to be optimized, and \( \sigma_a \) is the prior emission scaling factors, which equals 1. \( S_{\text{obs}} \) and \( S_a \) are observational and prior error covariance matrices, respectively. \( \gamma \) is the regularization factor that balances the weights of the observational term and prior term. We assumed \( S_{\text{obs}} \) to be diagonal following Wang et al. (2020a) with the diagonal values calculated as the square of the standard error of tropospheric NO2 slant column density from the TROPOMI product. The prior error of the NOx emissions is assumed to be 100%. The spatial correlation of NOx emissions is considered in this study, and the off-diagonal elements of \( S_a \) are computed by assuming an exponentially decaying error correlation with a fixed decaying distance of 150 km following Qu et al. (2017). The \( \gamma \) value was determined as 500 via the total error minimization and L-curve test (Henze et al., 2009; Qu et al., 2017).

We developed the observation operator for TROPOMI NO2 product in the GEOS-Chem adjoint model with GEOS-Chem NO2 vertical profiles and TROPOMI NO2 averaging kernel...
applied to minimize the discrepancies between the assumptions in TROPOMI NO\textsubscript{2} retrieval and GEOS-Chem model simulation. See Appendix B for additional details. The observation operator has been validated using the finite difference method (Appendix C).

For anthropogenic VOC emissions update, we only applied the mass balance method based on the TROPOMI HCHO data. The default anthropogenic VOC emissions used in the GEOS-Chem is also MIX 2010 (Li et al., 2017). We ignore the change of anthropogenic VOC emissions from 2010 to 2019. The baseline VOC emission in 2019 (MIX 2019) is identical to that of MIX 2010. The updated anthropogenic VOC emissions during the lockdown period is denoted as MIX 2020. HCHO is one species of VOC and may not be able to represent other VOC species. Different from NO\textsubscript{x}, biogenic sources, meteorological impacts, and large retrieval uncertainty of HCHO due to its low optical depth prevent accurately quantifying the emission decline due to lockdown from satellite retrievals (Levelt et al., 2022). To optimize the signal, we spatially aggregate the ratio of TROPOMI HCHO during the lockdown period to that before the lockdown to the resolution of 0.5\textdegree, which are used as the scaling factors for updating the anthropogenic VOC emissions during the lockdown period. The aggregation is based on the oversampling of TROPOMI HCHO at 0.01\textdegree resolution, and the ratio is computed as the mean of the lowest 25th percentile of all ratios at 0.01\textdegree resolution in each 0.5\textdegree×0.5\textdegree grid box, which ensures that only statistically significant changes are considered. We assumed the change of anthropogenic VOC emissions over sparsely populated areas (TROPOMI NO\textsubscript{2} in 2019 Feb.-Mar. less than 0.1 DU) is insignificant and assigned the ratio values as one. To further evaluate the uncertainties associated with this approach, we also conducted sensitivity study by using different threshold in the aggregation.

We assess the results from model experiments (as described in Section 2.5) adopting the updated NO\textsubscript{x} emission by comparing mean tropospheric NO\textsubscript{2} VCD from GEOS-Chem and from TROPOMI observations in Feb.-Mar. of 2019 and 2020. The averaging kernel of TROPOMI NO\textsubscript{2} is applied to modeled NO\textsubscript{2} column for this comparison, following Sha et al. (2021). Further quantitative evaluation of the model results also used the surface observation of \textit{O}3 and NO\textsubscript{2}.

### 2.5 GEOS-Chem model experiments

A series of sensitivity experiments is conducted over China with different NO\textsubscript{x} and VOC emissions and GEOS-FP meteorological fields in different years using GEOS-Chem (v12.7.2) model. All simulations are conducted from Jan. 15 to Mar. 31. The 17 days before Feb. 1 are used...
for spin up, and the model output for Feb. and Mar. are used for the analysis. The configurations of different simulations are listed in Table 1.

Table 1. Configurations of model sensitivity experiments.

<table>
<thead>
<tr>
<th>Experiments</th>
<th>Abbreviation</th>
<th>Meteorology</th>
<th>NOx Emission</th>
<th>VOC Emission</th>
</tr>
</thead>
<tbody>
<tr>
<td>2020 Default</td>
<td>2020D</td>
<td>GEOS-FP 2020</td>
<td>MIX 2019</td>
<td>MIX 2019</td>
</tr>
<tr>
<td>2020 NOx</td>
<td>2020N</td>
<td>GEOS-FP 2020</td>
<td>MIX 2020</td>
<td>MIX 2019</td>
</tr>
<tr>
<td>2020 VOC</td>
<td>2020V</td>
<td>GEOS-FP 2020</td>
<td>MIX 2019</td>
<td>MIX 2020</td>
</tr>
<tr>
<td>2020 Lockdown</td>
<td>2020L</td>
<td>GEOS-FP 2020</td>
<td>MIX 2020</td>
<td>MIX 2020</td>
</tr>
<tr>
<td>2020 Adjoint</td>
<td>2020A</td>
<td>GEOS-FP 2020</td>
<td>Adjoint 2020</td>
<td>MIX 2020</td>
</tr>
</tbody>
</table>

We use the following equations to quantify the contributions from NOx and VOC emission reduction due to COVID-19 and meteorological variation to the increase of surface O3.

\[
\Delta O^\text{NOx}_3 = \frac{O_3^{2020A} - O_3^{2020V}}{O_3^{2019B}} \times 100\% \tag{2}
\]

\[
\Delta O^\text{VOC}_3 = \frac{O_3^{2020L} - O_3^{2020N}}{O_3^{2019B}} \times 100\% \tag{3}
\]

\[
\Delta O^\text{ems}_3 = \frac{O_3^{2020A} - O_3^{2020D}}{O_3^{2019B}} \times 100\% \tag{4}
\]

\[
\Delta O^\text{met}_3 = \frac{O_3^{2020D} - O_3^{2019B}}{O_3^{2019B}} \times 100\% \tag{5}
\]

Where \(\Delta O^\text{NOx}_3\), \(\Delta O^\text{VOC}_3\) and \(\Delta O^\text{ems}_3\) are the relative differences in surface O3 concentration caused by emission decline of NOx, VOC, and both NOx and VOC resulting from COVID-19. \(\Delta O^\text{met}_3\) represents the relative contribution to the surface O3 change from the meteorological variation between 2 years. \(O_3^{2019B}\), \(O_3^{2020D}\), \(O_3^{2020N}\), \(O_3^{2020V}\), \(O_3^{2020L}\) and \(O_3^{2020A}\) are mean MDA8 surface O3 concentration simulated by modeling experiments Baseline (2019), 2020 Default, 2020 NOx, 2020 VOC, 2020 Lockdown and 2020 Adjoint, respectively (Table 1).

The difference in simulated surface O3 between 2020 and 2019, is the result of both emission reductions and meteorological variations and is denoted as \(\Delta O^\text{all}_3\). It is calculated as
follows and is evaluated against the observed relative difference of mean MDA8 O3 in Feb. to Mar. between 2019 and 2020 at all ground sites:

\[
\Delta O_3^{all} = \frac{O_3^{2020A} - O_3^{2019B}}{O_3^{2019B}} \times 100\% 
\] (6)

3. Results of model development, emissions, and validation

3.1 Changes of NOx and VOC emissions during COVID

We updated the anthropogenic NOx emissions during the COVID lockdown using both 4D-Var and mass balance methods (Fig. 1 and 2). The NOx emissions from the 4D-Var inversion share a similar spatial pattern and magnitude with those found using the mass balance method (Fig. 1). However, the NOx emissions from the 4D-Var inversion are lower overall than those from the mass balance method over North China by ~10% and larger over central China by ~40%. Fig. 2(a-b) shows that the 4D-Var NOx emission reduction is more severe over urban regions and displays a smoother spatial pattern than that from the mass balance approach, which is caused by the arbitrary cut off with 0.1 DU of NO2 VCD in the latter. Furthermore, the 4D-Var inversion captured the NOx emission decline in Northeast China where the mass balance approach did not because of the low NO2 VCD. During Feb.-Mar. 2020, the anthropogenic NOx emissions in East China decreased by ~30% compared to those in the same period in 2019. We also scale the anthropogenic VOC emissions based on the TROPOMI HCHO data (Fig. 2(c)). The VOC emissions decrease by ~20%-30% in East and South Asia. The anthropogenic VOC emission changes in sparsely populated areas over Northwest China are neglected.
Figure 1. Updated anthropogenic NOx emission during Feb.-Mar. 2020 from (a) mass balance method, (b) 4D-Var method and (c) their relative difference.

Figure 2. Scaling factors for anthropogenic NOx emission in Feb.-Mar. from 2019 to 2020 as derived from (a) 4D-Var, (b) mass balance. Scaling factors for anthropogenic VOC emissions from the mass balance are in (c).

3.2 Validation of NO2 simulations

We further assess our updated anthropogenic NOx emissions by comparing the NO2 VCD from TROPOMI with that from GEOS-Chem with the anthropogenic NOx emissions before and after the scaling (Fig. 3 and 4). Before updating the NOx emissions, the 2020 Default (Fig. 3(b)) simulation significantly overestimates the NO2 VCD compared to the TROPOMI NO2 observations (Fig. 3(a)). With the NOx emissions updated, the simulations 2020 NOx (Fig. 3(c))
and 2020 Adjoint (Fig. 3(d)) exhibit a much better agreement with TROPOMI NO\textsubscript{2} observation than 2020 Default. However, Fig. 3(c) shows the GEOS-Chem simulation with the NOx emissions from mass balance approach overestimated the NO\textsubscript{2} VCD over Beijing and southwest of Hebei Province (pink and black circles in Fig. 3) compared with TROPOMI data. The reason is that scaling factors are applied only to anthropogenic NOx emissions, not total NOx emissions, so it is expected that the model may still overestimate the NO\textsubscript{2} column after scaling part of the total NOx emission. With the anthropogenic NOx emissions optimized by the 4D-Var method, the overestimation of NO\textsubscript{2} VCD over Beijing and southwest of Hebei Province (pink and black circles in Fig. 3) is mitigated compared with the NOx emissions from mass balance approach.

Fig. 4 further displays the statistics for the comparison between the TROPOMI NO\textsubscript{2} and GEOS-Chem simulations via the scatterplot. The Baseline (2019) simulation captures the magnitude of NO\textsubscript{2} VCD observations in 2019 well (Fig. 4(a)). The root-mean-square-error (RMSE) and mean bias error (MBE) for the simulation with 2020 NOx emission derived from mass balance method (Fig. 4(b)) decreased by 0.050 DU and 0.057 DU as compared to the 2020 Default (Fig. 4(c)). Compared with the result from GEOS-Chem simulation 2020 NOx, emissions from 2020 Adjoint (Fig. 4(d)) further led to the reduction of the MBE of the NO\textsubscript{2} VCD by 0.006 DU and improve the correlation coefficient by 0.003. The significant overestimation of several pixels with TROPOMI NO\textsubscript{2} VCD larger than 0.4 DU by the simulation 2020 NOx is also mitigated by 2020 Adjoint. The MBE between GEOS-Chem and TROPOMI for Baseline (2019), 2020 NOx and 2020 Adjoint are -0.004, 0.015 and 0.009 DU, respectively. The corresponding relative bias are 1.9%, 10% and 6.0%, which are all less than the relative uncertainty of \(\sim 30\%\) for TROPOMI tropospheric NO\textsubscript{2} VCD over East China (Van Geffen et al., 2022). The improved agreement between the simulation with updated NOx emission and TROPOMI NO\textsubscript{2} provides a basis for further analyzing the mechanism of aggravated surface O\textsubscript{3} pollution.
Figure 3. Comparison of tropospheric NO$_2$ VCD from (a) TROPOMI product in 2020 Feb.-Mar with that from GEOS-Chem simulations (b) 2020 Default, (c) 2020 NOx and (d) 2020 Adjoint. The pink and black circles mark the areas where NOx emissions from 4D-Var mitigated the NO$_2$ overestimation by mass balance method. The emissions and meteorology configurations for GEOS-Chem simulations 2020 Default, 2020 NOx and 2020 Adjoint are listed in Table 1.
Figure 4. Scatter plot of TROPOMI NO$_2$ VCD versus the GEOS-Chem simulations for (a) Baseline (2019), (b) 2020 Default, (c) 2020 NOx and (d) 2020 Adjoint, respectively. TROPOMI data in Feb.-Mar. of 2019 was used in (a), and that of 2020 was used in (b-d). The emissions and meteorology configurations for GEOS-Chem simulations are listed in Table 1. Only pixels with TROPOMI NO$_2$ VCD in 2019 Feb.-Mar. larger than 0.1 DU are included in all comparisons.
Fig. 5 and Fig. 6 show the comparison of surface NO$_2$ between ground measurements and GEOS-Chem simulations. The GEOS-Chem simulations Baseline (2019) (Fig. 5(b)) and 2020 Adjoint (Fig. 5(d)) both capture the spatial pattern and magnitude of surface NO$_2$ measurements in Feb.-Mar. of 2019 (Fig. 5(a)) and 2020 (Fig. 5(c)) well, respectively. Fig. 6 further displays the good agreements of surface NO$_2$ from Baseline (2019) (Fig. 6(a)) and 2020 Adjoint (Fig. 6(b)) to the in-situ measurements via scatter plots. Table 2 displays the evaluation statistics, including the correlation coefficient (R), MBE, RMSE and the slope and intercept of the linear regression, for the simulated surface NO$_2$ from various simulation experiments compared with the in-situ measurements. The correlation coefficient, MBE and RMSE between the simulation Baseline (2019) and ground measurements in 2019 Feb.-Mar. are 0.724, 1.572 µg m$^{-3}$ and 8.49 µg m$^{-3}$, respectively. Without updating the NOx emissions in 2020, the simulation 2020 Default overestimate the ground measurements of surface NO$_2$ in 2020 Feb.-Mar (Table 2). The slope for the linear regression is 1.19, and the MBE and RMSE are 6.021 µg m$^{-3}$ and 10.43 µg m$^{-3}$, respectively (Table 2). After updating the NOx emissions, the GEOS-Chem simulations 2020 NOx and 2020 Adjoint have good agreements with the in-situ measurements in 2020 Feb.-Mar. The correlation coefficient between the simulation 2020 Adjoint versus the in-situ measurements is 0.651, higher than that of 0.608 for the simulation 2020 NOx versus the ground measurements (Table 2). The MBE and RMSE of 2020 Adjoint (0.683 µg m$^{-3}$ and 6.68 µg m$^{-3}$) are lower than those of 2020 NOx (1.726 µg m$^{-3}$ and 7.74 µg m$^{-3}$) (Table 2). This result further indicates the superiority of 4D-Var for optimizing NOx emissions compared with the mass balance method (Cooper et al., 2017; Streets et al., 2013).
Figure 5. Comparison of surface NO$_2$ concentrations from ground measurements for (a) 2019 Feb.-Mar. and (c) 2020 Feb.-Mar. versus those from GEOS-Chem simulations (b) Baseline (2019) and (d) 2020 Adjoint. Grey color means no data is presented.
Figure 6. Scatter plots for comparing the surface NO$_2$ concentrations from GEOS-Chem simulations and ground measurements. (a) GEOS-Chem simulation Baseline (2019) versus ground measurements in 2019 Feb.-Mar. (b) GEOS-Chem simulation 2020 Adjoint versus ground measurements in 2020 Feb.-Mar. Note: the number of ground sites differ in these two years.

Table 2. Evaluation statistics for modeled surface NO$_2$ compared with the in-situ measurements*.

<table>
<thead>
<tr>
<th>Experiments</th>
<th>R</th>
<th>MBE (µg m$^{-3}$)</th>
<th>RMSE (µg m$^{-3}$)</th>
<th>Slope</th>
<th>Intercept (µg m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Baseline (2019)</td>
<td>0.724</td>
<td>1.572</td>
<td>8.49</td>
<td>1.01</td>
<td>1.43</td>
</tr>
<tr>
<td>2020 Default</td>
<td>0.661</td>
<td>6.021</td>
<td>10.43</td>
<td>1.19</td>
<td>2.95</td>
</tr>
<tr>
<td>2020 NOx</td>
<td>0.608</td>
<td>1.726</td>
<td>7.74</td>
<td>0.92</td>
<td>3.03</td>
</tr>
<tr>
<td>2020 Adjoint</td>
<td>0.651</td>
<td>0.683</td>
<td>6.68</td>
<td>0.91</td>
<td>2.22</td>
</tr>
</tbody>
</table>

* The simulation experiment Baseline (2019) is compared with the ground measurements in 2019 Feb.-Mar. Other three experiments are compared with the ground measurements in 2020 Feb.-Mar.

Fig. 7 (a) is the Taylor diagram for evaluating the GEOS-Chem simulations of surface NO$_2$ concentrations from 2020 Default, 2020 NOx and 2020 Adjoint using the in-situ measurements.
The simulation 2020 Adjoint (inverted triangle in Fig. 7(a)) has the best performance among these three simulations with the lowest relative bias and lowest normalized centered RMSE. Without updating the NOx emission, 2020 Default features a relative bias of ~37%. After updating the NOx emissions, 2020 NOx reduces the relative bias, normalized centered RMSE and normalized standard deviation from around 37%, 1.38 and 1.80 to around 10%, 1.20 and 1.51 compared with 2020 Default, but the correlation coefficient also decreases. By using 4D-Var method, 2020 Adjoint further reduces the relative bias, normalized centered RMSE and normalized standard deviation and increases the correlation coefficient compared with 2020 NOx.

Figure 7. Taylor diagram for evaluating the GEOS-Chem simulations of (a) surface NO$_2$ and (b) surface O$_3$ during lockdown period (2020 Feb.-Mar.) using ground observations for different simulation experiments listed in Table 1. The evaluation of surface O$_3$ only includes the areas where the NOx emissions optimized by 4D-Var reduced by more than 10%.

3.3 Evaluation of surface O$_3$ simulations

We evaluated the GEOS-Chem simulations of MDA8 surface O$_3$ from different simulation experiments listed in Table 1 using ground measurements. Fig. 7(b) is the Taylor diagram for comparing the surface O$_3$ concentrations during 2020 Feb.-Mar. from ground measurements and
GEOS-Chem simulations. We focused on areas with significant NOx emissions reduction to better assess the role of updated NOx emissions in improving surface O$_3$ simulations. The ground sites where the NOx emissions from 4D-Var decline by less than 10% are excluded. The correlation coefficient between the simulation Baseline (2019) and ground observations is ~0.53 and the relative bias is around -25%. By applying 2020 meteorological fields and scaling the VOC emissions, the correlation coefficients decreased to ~0.40 for model simulations 2020 Default and 2020 VOC, with little reduction in the relative bias. By updating the NOx emissions, the relative bias reduced to around -10% while the correlation coefficients remain at ~0.50 for model simulations 2020 NOx, 2020 Lockdown and 2020 Adjoint. This indicates the NOx emission updates significantly improve the surface O$_3$ simulations. Comparing the simulations 2020 Default and 2020 VOC, or 2020 NOx and 2020 Lockdown, the results show that scaling VOC emissions does not improve the surface O$_3$ simulations significantly over the continental China, but over South China, VOC emissions update reduces the relative bias by 3%. Among all simulations, 2020 Adjoint exhibits the best performance with the lowest normalized centered RMSE, largest correlation coefficient and a low relative bias of ~10%. This result further confirms the superiority of the 4D-Var with respect to the mass balance method for optimizing NOx emissions. Therefore, we used the 2020 Adjoint to evaluate the impacts of NOx emission on surface O$_3$ in the following analysis.

Fig. 8 compares the modeled surface O$_3$ in Feb.-Mar. of 2019 (Fig. 8(a)) and 2020 (Fig. 8(b)) and the relative difference (Fig. 8(c)) computed from Equation (6) with the in-situ measurements (Fig. 8(d-f)). The ground observations show that the highest level of surface O$_3$ pollution occurs in North China and southwest of China. The average MDA8 O$_3$ in two months can reach up to ~110 µg m$^{-3}$ at STP (~51.4 ppbv), which is higher than the China National Ambient Air Quality Standard daily maximum 8-hour Grade I standard of 100 µg m$^{-3}$. GEOS-Chem model underestimates the surface O$_3$ over North China for both years compared with ground observations, which could be a result of the out-of-date VOC emissions, but it captures the magnitude and spatial distribution of surface O$_3$ and the increasing trend in South China well. In South China, the measured surface O$_3$ in 2020 Feb.-Mar. increases by 30-50%, while over North China, it increases generally by less than 20% even decreases in some regions. The relative differences of simulated surface O$_3$ between two years is comparable to the ground observations over South China (green box in Fig. 8(c, f)). Over North China (pink box in Fig. 8(c, f)), the average relative difference
between two years from the model and observation are 4.27% and -3.01%, respectively, both of which are much smaller than their counterparts in South China. While the relative difference from model simulations has different signs as compared to that of observations on average, both the change of O₃ is indeed small and the model is able to capture the part of O₃ decrease in the southwest part of the North China domain (Fig. 8(c)). We note that some previous studies showed large increase of O₃ in North China, but such increase is in comparison with the O₃ in the month right before the lockdown (not the same time in 2019; (Shi & Brasseur, 2020; Y. M. Liu et al., 2021).

**Figure 8.** Comparison of MDA8 surface O₃ in 2019 and 2020 Feb.-Mar. and the relative difference between two years from GEOS-Chem model (a-c) versus ground observations (d-f). GEOS-Chem mean MDA8 O₃ at 9 m above the surface under standard temperature and pressure (STP; 273.15
K, 101.325 kPa) from (a) Baseline (2019) and (b) 2020 Adjoint simulation (Table 1) together with (c) their relative difference. Ground observed mean MDA8 surface O3 under STP in (d) 2019 Feb.-Mar.; (e) 2020 Feb.-Mar. and (f) their relative difference. The pink and green boxes in (c) and (f) define the North China and South China domain.

4. Mechanism of aggravated surface O3 pollution

4.1 Relative contribution from declining emissions and meteorological variations

From equations (2)-(5) we can analyze the mechanism of surface O3 increase in China during the COVID-19 pandemic (Fig. 9). NOx emission reduction as a result of COVID-19 lockdown leads to a ~8% increase in the mean MDA8 surface O3 over North China (pink boxes in Fig. 9) between 2019 and 2020 Feb.-Mar. (Fig. 9(a)), while the VOC emission decline causes ~3% of O3 decrease (Fig. 9(b)). The average contribution of the meteorological variations to the surface O3 change is less than 1% in North China (Fig. 9(d)). However, in South China, the inter-annual meteorological variations dominate the surface O3 increases, causing a ~30% increase (Fig. 9(d)), while the NOx and VOC emission reduction has little impacts. The overall magnitude of emissions contribution to the surface O3 change over North China is ~5%, similar to that of the meteorological effects, but meteorological variations lead to both O3 increases and decreases in different regions. Over South China, the meteorological effect is much larger than the net effects of declining emissions. Overall, the impact of inter-annual meteorological variations between 2019 and 2020 is almost 30 times larger than the overall emissions impacts on the aggregated surface O3 pollution in China. Our results are consistent with the conclusion from Zhao et al. (2020) that meteorological variation has larger impacts than emissions reduction on surface O3 in the southern city of Guangzhou, but in Beijing, emission reduction has a larger impact during 23-29 January. Liu et al. (2020b) reported that the surface O3 increase in the major cites of the Yangtze River Delta region were driven by both emission reduction and meteorological variations to a similar degree from pre-lockdown period (Jan. 1-22, 2020) to lockdown period (Jan. 23-Feb. 29, 2020). However, Zhao et al. (2020) and Liu et al. (2020b) only focused on the lockdown period of one week in reference to the time period right before the lockdown instead of the same period in previous years, which cannot exclude the effects of seasonal variation of meteorology and did not provide a comprehensive analysis over the whole lockdown period. Moreover, Liu et al. (2020b)
only analyzed four representative cities instead of showing the analysis at a national scale. Further, Zhao et al. (2020) did not update the anthropogenic emissions during the lockdown period, which brings significant uncertainties to their analysis.

Figure 9. Relative difference in simulated surface O$_3$ caused by (a) NOx emission reduction, (b) VOC emission reduction, (c) overall emission reduction and (d) meteorological variations due to COVID-19 lockdown. The pink and green boxes in each panel define the North China and South China domain.
4.2 Higher temperature leading to aggravated surface O₃ pollution in South China

The positive correlation between the surface O₃ and temperature is widely observed and reported in the literature (Pusede et al., 2015). Higher temperature leads to higher concentrations of surface O₃ because it improves the O₃ production rate by affecting the organic reactivity, production of HOx radicals, formation and decomposition of peroxy nitrates and alkyl nitrates (Pusede et al., 2015). We calculated the daily difference in Feb.-Mar. between 2020 and 2019 (excluding Feb. 29, 2020) for the daily mean of MDA8 O₃ from ground measurements and 2-meter air temperature from GEOS-FP data used in our GEOS-Chem simulations for the South China (106°E - 118°E, 22°N - 31°N, green box in Fig. 10(a)) and North China (111°E - 123°E, 33°N - 40°N, pink box in Fig. 10(a)). Fig. 10 displays the difference of 2-month mean 2-meter air temperature in Feb.-Mar. between 2020 and 2019 (Fig. 10(a)) and the scatter plot between the daily difference of measured surface O₃ concentration and 2-meter air temperature over both South China (green dots in Fig. 10(b)) and North China (pink dots in Fig. 10(b)). We found the 2-meter air temperature increased by ~2.3°C in South China, and the daily difference of surface O₃ concentration and 2-meter air temperature are well correlated with a positive correlation coefficient of 0.612. Therefore, the significant aggravated surface O₃ pollution in South China could be attributed to the temperature increase. The reason for the temperature increase is the lower cloud fraction. Via analyzing the GEOS-FP data, we found the cloud fraction decreases by ~5%, and the downward visible direct flux at surface increased by 5 W m⁻² over South China. The lower cloud fraction increases the downward solar radiation at the surface during the lockdown period, leading to higher surface air temperature. The enhanced solar radiation at the surface could also promote the production of O₃ via photochemical reactions. In North China, 2-meter air temperature also increased by 1.8°C, but the measured surface MDA8 O₃ decreased by 3% (Fig. 8(f)). Fig. 10(b) shows the daily difference of MDA8 O₃ and 2-meter air temperature over North China also has a high correlation coefficient of 0.731. However, the intercept of the linear regression line is negative, so that the surface O₃ could decrease even though the temperature increases. The predicted average change of surface MDA8 O₃ in South China and North China are marked by the green and pink open squares respectively in Fig. 10(b) based on the linear regression. Because of the different intercepts, the predicted MDA8 O₃ in South China increases by ~9.0 µg m⁻³, while it decrease by 2.2 µg m⁻³ in North China, although the average temperature increased in both South and North China.
Figure 10. (a) The change of 2-meter air temperature from 2019 Feb.-Mar. to 2020 Feb.-Mar. (b) The scatter plots between the daily difference of surface $O_3$ measurements and 2-meter air temperature in Feb.-Mar. between 2020 and 2019 over South China (green dots) and North China (pink dots). The green and pink open squares mark the predicted average change of surface MDA8 $O_3$ in South China (green open square) and North China (pink open square), respectively, based on the linear regression against temperature change.

5. Summary

A significant reduction in primary air pollutants has been identified by surface and satellite observations during the COVID-19 pandemic in China (Bauwens et al., 2020; Miyazaki et al., 2020), which is in contrast to the increase of surface $O_3$. In this study, we analyzed the reasons for the enhanced surface $O_3$ pollution from two perspectives: anthropogenic emissions reduction and inter-annual meteorological variations. We constrain the NOx emissions based on the TROPOMI NO$_2$ product using both the mass balance and 4D-Var methods. The VOC emission were also updated based on the TROPOMI HCHO product via the mass balance approach. We analyzed the contributions from emissions reduction and meteorological variations to surface $O_3$ increases through a series of sensitivity simulations using the GEOS-Chem model.
The updated NOx emissions from the 4D-Var and mass balance approaches share a similar spatial pattern. However, the NOx emissions from 4D-Var are lower than those from the mass balance method over North China by ~10% but larger over central China by ~40%. The evaluation of the simulations with the updated emissions against the TROPOMI NO2, in-situ measurements of surface NO2 and O3 indicate that the NOx emissions from the 4D-Var inversion leads to better model performance than that from the mass balance approach.

The anthropogenic NOx emission decreased by ~30% over East China during 2020 Feb.-Mar. compared to the same period in 2019. Over North China, NOx emission reduction leads to a ~8% increase in the mean MDA8 surface O3, while the VOC emissions decline causes O3 to decrease by ~3%. The average contribution of meteorological variations to the surface O3 change is less than 1% in North China. However, in South China, the inter-annual meteorological variation dominates the surface O3 increase, causing a ~30% increase, while the NOx and VOC emission reduction has nearly no impacts on O3. Overall, the impact of inter-annual meteorological variations between 2019 and 2020 is almost 30 times larger than the impact of emissions on the enhanced surface O3 pollution in China.

The significant increase of surface O3 in South China could be attributed to the higher temperature during the lockdown period, which is caused by the lower cloud fraction. The lower cloud fraction increases the downward solar radiation at the surface during the lockdown period, leading to higher surface air temperature. The enhanced solar radiation at the surface could also promote the production of O3 via photochemical reactions.

Appendix A: NOx emission reduction in China from 2010 to 2019

The default anthropogenic NOx emission over East Asia in GEOS-Chem is MIX 2010 (Li et al., 2017). To generate the anthropogenic NOx emission in 2019, we calculated the ratio of mean TROPOMI tropospheric NO2 VCD in Feb.-Mar. 2019 to GEOS-Chem simulated NO2 VCD with the default MIX 2010 emission as the scaling factor (Fig. A1). The scaling factors in regions where mean TROPOMI tropospheric NO2 VCD in 2019 Feb.-Mar. less than 0.1 DU are set to 1. From 2010 to 2019, the anthropogenic NOx emission has declined significantly as a result of the clean air actions of Chinese government (Zheng et al., 2018).
Figure A1. The scaling factor of anthropogenic NOx emission from year 2010 to 2019.

Appendix B: Applying the TROPOMI NO\textsubscript{2} averaging kernel in the observation operator

To optimize the NOx emissions and minimize the cost function (Equation (1)) with the 4D-Var method, GEOS-Chem adjoint needs to compute the derivative of the cost function with respect to the model parameters to be optimized, which is the scaling factors of the anthropogenic NOx emissions in this study. An essential step is to calculate the adjoint forcing $\mathbf{F}$, which is the derivative of the cost function with respect to the modeled NO\textsubscript{2} concentration shown as Equation (B1).

$$\mathbf{F} = \frac{\partial J}{\partial \mathbf{c}} = \mathbf{S}_{\text{obs}}^{-1} [H(\mathbf{c}) - \mathbf{s}] \frac{\partial H(\mathbf{c})}{\partial \mathbf{c}}$$

(B1)

For each single TROPOMI NO\textsubscript{2} observation, the adjoint forcing component $f$ and cost function component $j$ are computed as Equation (B2) and Equation (B3).

$$f = \frac{M_{\text{gc}} v_{\text{gc}} - M_{\text{obs}} v_{\text{obs}}}{e_{\text{obs}} M_{\text{obs}}} M_{\text{gc}}$$

(B2)

$$j = 0.5 f \left( \frac{M_{\text{gc}} v_{\text{gc}} - M_{\text{obs}} v_{\text{obs}}}{M_{\text{gc}}} \right)$$

(B3)
Here $M_{gc}$ is GEOS-Chem air mass factor applying the GEOS-Chem NO$_2$ vertical profiles and TROPOMI NO$_2$ averaging kernel. $M_{obs}$ is TROPOMI air mass factor. $v_{gc}$ and $v_{obs}$ are the tropospheric NO$_2$ VCD from GEOS-Chem model and TROPOMI observation, respectively. The product of air mass factor and NO$_2$ VCD is NO$_2$ slant column density. $e_{obs}$ is the standard error of TROPOMI tropospheric NO$_2$ VCD.

We calculated the GEOS-Chem air mass factor $M_{gc}$ as Equation (B4) following Qu et al. (2019).

$$M_{gc} = \frac{\sum_{i \in \text{trop}} c_{i}^{gc} \Delta p_{i}^{gc} w_{i}^{gc}}{\sum_{i \in \text{trop}} c_{i}^{gc} \Delta p_{i}^{gc}}$$

Here $c_{i}^{gc}$ is GEOS-Chem NO$_2$ mixing ratio at vertical layer $i$, $\Delta p_{i}^{gc}$ is the pressure difference between the GEOS-Chem vertical layer $i$ and $i+1$. $w_{i}^{gc}$ is the scattering weight at the GEOS-Chem vertical layer $i$, which is calculated by the linear interpolation of the scattering weights at the vertical coordinate of the model TM5 used for TROPOMI NO$_2$ retrieval. The scattering weight at the TM5 vertical layer $l$ ($w_{l}^{TM5}$) is computed as the product of TROPOMI air mass factor and the TROPOMI averaging kernel at the TM5 vertical layer $l$ ($A_{l}^{TM5}$) using Equation (B5) (Eskes and Boersma, 2003).

$$w_{l}^{TM5} = M_{obs} A_{l}^{TM5}$$

**Appendix C: Validation of the TROPOMI NO$_2$ observation operator**

We validated the observation operator by comparing the sensitivity of the cost function with respect to the emission scaling factor from GEOS-Chem adjoint and a finite difference estimation as shown in Equation (C1). We shut down the transport and exclude a priori term from the cost function for the validation, so that the gradient of cost function component in each grid cell to the local emission scaling factor equals to the gradient of total cost function to the emission scaling factor in the same grid cell.

$$\frac{\partial J(\ln \sigma)}{\partial \ln \sigma} \approx \frac{J(\ln (\sigma + 0.05)) - J(\ln (\sigma - 0.05))}{\ln (\sigma + 0.05) - \ln (\sigma - 0.05)}$$

Fig. C1 compared the cost function sensitivities calculated from GEOS-Chem adjoint and the finite difference method for the nested grids with the spatial resolution of 0.25°×0.3125°. The spatial pattern and magnitude of the cost function sensitivities from the two methods match with
each other with a correlation coefficient of 0.97. The statistics show that the agreement of the
adjoint sensitivities and finite difference sensitivities in this study is comparable to that in Wang
et al. (2020a) although we constrain the NOx emission at a much finer resolution of 0.25°×0.3125°
than in their study (2°×2.5°).

![Comparison of adjoint sensitivities and finite difference sensitivities. (a) Scatter plot of the adjoint sensitivity of the cost function with respect to the logarithm of NOx emission scaling factor versus the finite difference sensitivities. The color scheme for panel (a) encodes the number of samples (the legend on the right of panel (a)). (b) Map of finite difference sensitivity. (c) Map of adjoint sensitivity.](image)

**Figure C1.** Comparison of adjoint sensitivities and finite difference sensitivities. (a) Scatter plot of the adjoint sensitivity of the cost function with respect to the logarithm of NOx emission scaling factor versus the finite difference sensitivities. The color scheme for panel (a) encodes the number of samples (the legend on the right of panel (a)). (b) Map of finite difference sensitivity. (c) Map of adjoint sensitivity.

**Data availability**

The TROPOMI NO₂ and HCHO product are available at the NASA Goddard Earth Sciences Data and Information Services Center ([https://daac.gsfc.nasa.gov](https://daac.gsfc.nasa.gov)). The ground O₃ and NO₂ measurements are available at the China National Environmental Monitoring Center ([http://www.cnemc.cn/en/](http://www.cnemc.cn/en/)).

**Author contribution**

ZL and JW designed the research, and ZL conducted the research. YW, DKH and XC contributed to the research design. ZL and JW wrote the manuscript, and XC and DKH contributed to the writing. YW and TS developed the codes for comparing the tropospheric NO₂ VCD from model and TROPOMI data. KS developed the codes for oversampling.
Competing interests

The authors declare that they have no conflict of interest.

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