

**Aggravated surface O₃ pollution primarily driven by meteorological variation
in China during the 2020 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, as compared to the same time period in 2019, has been identified by satellite and ground observations. However, this reduction is in contrast with the increase of surface O_3 concentration in many parts of China during the same period from 2019 to 2020. The reasons for this contrast are studied here from two perspectives: emission changes and inter-annual meteorological variations. Based on top-down constraints of NO_x emissions from TROPOMI measurements and GEOS-Chem model simulations, our analysis reveals that NO_x and volatile organic compound (VOC) emission reductions as well as meteorological variations lead to 8%, -3%, and 1% changes in O_3 over North China, respectively. In South China, however, we find that meteorological variations cause ~30% increases in O_3 , which is much larger than -1% and 2% changes due to VOC and NO_x emission reductions, respectively, and the overall O_3 increase in the simulations is consistent with the surface observations. The higher temperature associated with increase of solar radiation and declined relative humidity are the main reasons that lead to the surface O_3 increase in South China. Collectively, inter-annual meteorological variations have a larger impact than emission reductions on the aggravated surface O_3 pollution in China during the lockdown period of COVID-19 pandemic.

1. Introduction

Surface ozone (O_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 (NO_x) 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_3 formation (Lu et al., 2019).

Ground observations show that surface O_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_2 vertical column density (VCD) from satellite measurements and

surface NO₂ 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 (SO₂), 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 O₃ concentrations increased, and the respective roles of meteorological factor and emission reduction for the aggravated surface O₃ 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 O₃ pollution in China during the lockdown period of the pandemic from two perspectives using GEOS-Chem model. One is anthropogenic emission reduction of NO_x and VOC in response to the lockdown possibly under a VOC-limiting chemical regime of surface O₃ production (Guo et al., 2023), while the other is the impact of natural variability of meteorological conditions. Previous studies have reported the enhanced surface O₃ due to NO_x 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 O₃ 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 O₃ 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 O₃ 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 O₃ increase. A comprehensive analysis of the contributions from emission reductions and

meteorological variations to the surface O₃ 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 NO_x and VOC emission in February and March in 2020 based on the TROPOMI NO₂ and formaldehyde (HCHO) product. A set of GEOS-Chem model simulations with NO_x and VOC emissions and meteorological fields in different time periods are then conducted. Based on the difference in surface O₃ concentration in different modeling sensitivity experiments, we quantitatively assess the respective roles of emission and meteorology in regulating surface O₃ concentration in continental China. The ground observations of surface O₃ and NO₂ concentration are compared with the model simulations to verify our analysis. Section 2 introduces the satellite and ground-based measurements, NO_x emission update scheme, and the configurations of GEOS-Chem simulation experiments. Section 3 provides an evaluation of the constrained NO_x emission and surface O₃ simulations. The analysis of the mechanism of the aggravated surface O₃ pollution is presented in Section 4, followed by the summary and conclusions in Section 5.

2. Datasets and Methods

2.1 TROPOMI NO₂ and HCHO product

We used tropospheric NO₂ 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 NO₂ and HCHO retrievals at an approximately 7 km x 3.5 km spatial resolution (5.5km 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 NO₂ 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 NO₂ 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 O₃ and NO₂ measurements

Surface measurements of O₃ and NO₂ 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) O₃ concentration from hourly in situ measurements. Surface O₃ are measured by ultraviolet photometric method and Indigo disulfonate spectrophotometry, following the national environmental standards of HJ 590-2010 and HJ 504-2009. Surface NO₂ concentrations are measured by the chemiluminescence method (Zhang and Cao, 2015) that quantifies the NO₂ concentrations by measuring the NO decomposed from NO₂, which can cause a positive bias in the NO₂ measurements (Steinbacher et al., 2007) because NO_x (compounds produced from the atmospheric oxidation of NO_x) can also be decomposed to NO. The true NO₂ 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 NO₂, 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 $\mu\text{g m}^{-3}$ 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°E - 140°E , 15°N - 55°N). The boundary conditions are obtained from the $2^\circ \times 2.5^\circ$ global simulation. The model is driven by the GEOS-FP meteorological field provided by NASA Global Modeling and Assimilation Office (GMAO). A detailed O₃-NO_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 O₃ 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 O₃ 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 O₃ could decrease by ~3 ppb. Therefore, when comparing GEOS-Chem surface O₃ 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 NO_x emissions includes biomass burning from GFED4 inventory (Van Der Werf et al., 2017), soil NO_x 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 NO_x 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 NO₂ in the GEOS-Chem adjoint model version 35n similar to Wang et al. (2020a) and used it to optimize the anthropogenic NO_x emission during the lockdown period in China. The monthly NO_x 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° via assimilating the daily TROPOMI NO₂ measurements. The prior anthropogenic NO_x 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 NO_x 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 NO_x emission by assuming a constant NO_x lifetime and NO_x/NO₂ ratio. In the period from 2010 to 2019, the anthropogenic NO_x 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 NO₂ VCD in Feb.-Mar. 2019 to GEOS-Chem simulated NO₂ 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

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

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. Ω is the spatial and temporal domain where both model simulations and observations are available. σ is the scaling factor of anthropogenic NOx emissions to be optimized, and σ_a is the prior emission scaling factors, which equals 1. S_{obs} and S_a are observational and prior error covariance matrices, respectively. γ is the regularization factor that balances the weights of the observational term and prior term. We assumed S_{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 γ 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 NO₂ 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 (Appendix D). 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_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). Vigouroux et al. (2020) reported that TROPOMI HCHO tends to be overestimated by ~26% for HCHO column lower than 2.5×10^{15} molecules cm⁻² and underestimated by ~31% for HCHO column higher than 8.0×10^{15} molecules cm⁻². To optimize the signal, we spatially aggregate the ratio of TROPOMI HCHO in 2020 Feb.-Mar. to that in 2019 Feb.-Mar. to the resolution of 0.5°, 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° resolution, and the ratio is computed as the mean of the lowest 25th percentile of all ratios at 0.01° resolution in each 0.5°×0.5° grid box, which ensures that only statistically significant changes are considered. We assumed the change of anthropogenic VOC emissions over sparsely populated areas (TROPOMI NO₂ 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_x emission by comparing mean tropospheric NO₂ VCD from GEOS-Chem and from TROPOMI observations in Feb.-Mar. of 2019 and 2020. The averaging kernel of TROPOMI NO₂

is applied to modeled NO₂ column for this comparison, following Sha et al. (2021). Further quantitative evaluation of the model results also used the TROPOMI measurements of HCHO and surface observation of O₃ and NO₂.

2.5 GEOS-Chem model experiments

A series of sensitivity experiments is conducted over China with different NO_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.

Experiments	Abbreviation	Meteorology	NO _x Emission	VOC Emission
Baseline (2019)	2019B	GEOS-FP 2019	MIX 2019	MIX 2019
2020 Default	2020D	GEOS-FP 2020	MIX 2019	MIX 2019
2020 NO_x	2020N	GEOS-FP 2020	MIX 2020	MIX 2019
2020 VOC	2020V	GEOS-FP 2020	MIX 2019	MIX 2020
2020 Lockdown	2020L	GEOS-FP 2020	MIX 2020	MIX 2020
2020 Adjoint	2020A	GEOS-FP 2020	Adjoint 2020	MIX 2020

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

$$\Delta O_3^{\text{NO}_x} = \frac{O_3^{2020A} - O_3^{2020V}}{O_3^{2019B}} \times 100\% \quad (2)$$

$$\Delta O_3^{\text{VOC}} = \frac{O_3^{2020L} - O_3^{2020N}}{O_3^{2019B}} \times 100\% \quad (3)$$

$$\Delta O_3^{\text{ems}} = \frac{O_3^{2020A} - O_3^{2020D}}{O_3^{2019B}} \times 100\% \quad (4)$$

$$\Delta O_3^{\text{met}} = \frac{O_3^{2020D} - O_3^{2019B}}{O_3^{2019B}} \times 100\% \quad (5)$$

Where ΔO_3^{NOx} , ΔO_3^{VOC} and ΔO_3^{ems} are the relative differences in surface O_3 concentration caused by emission decline of NOx, VOC, and both NOx and VOC resulting from COVID-19. ΔO_3^{met} represents the relative contribution to the surface O_3 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 O_3 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 O_3 between 2020 and 2019, is the result of both emission reductions and meteorological variations and is denoted as ΔO_3^{all} . It is calculated as follows and is evaluated against the observed relative difference of mean MDA8 O_3 in Feb. to Mar. between 2019 and 2020 at all ground sites:

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

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. TROPOMI HCHO data cannot distinguish the anthropogenic emissions from biogenic and biomass burning sources for the Indochinese Peninsula in Southeast Asia because of the dense

vegetation in this region. However, this study investigated the O₃ pollution in China, the Southeast Asia with the dense vegetation is out of our study domain. The impact of VOC emission bias in Southeast Asia on the surface O₃ pollution in China is negligible considering the lifetime of biogenic VOC is generally short (Atkinson, 2000). For the populated urban regions in China, where the surface O₃ pollution exerts more significant health impacts, the anthropogenic source dominates the VOC emissions (Williams and Koppmann, 2007).

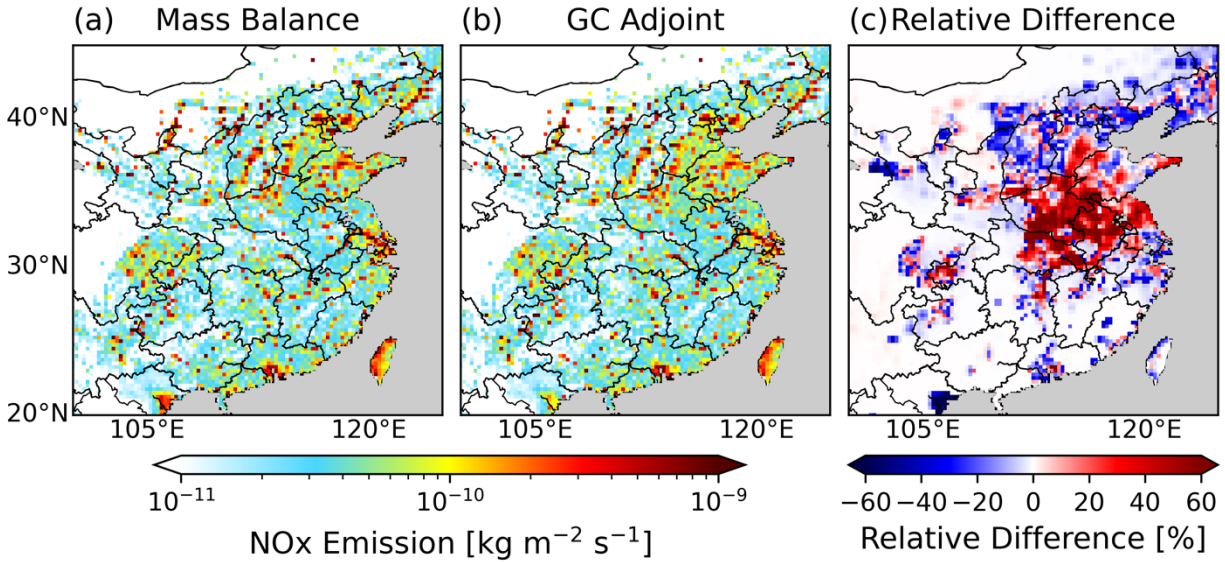


Figure 1. Updated anthropogenic NOx emission during Feb.-Mar. 2020 from (a) mass balance method, (b) 4D-Var method and (c) their relative difference. The relative difference is calculated as dividing the difference of (b) minus (a) by (a).

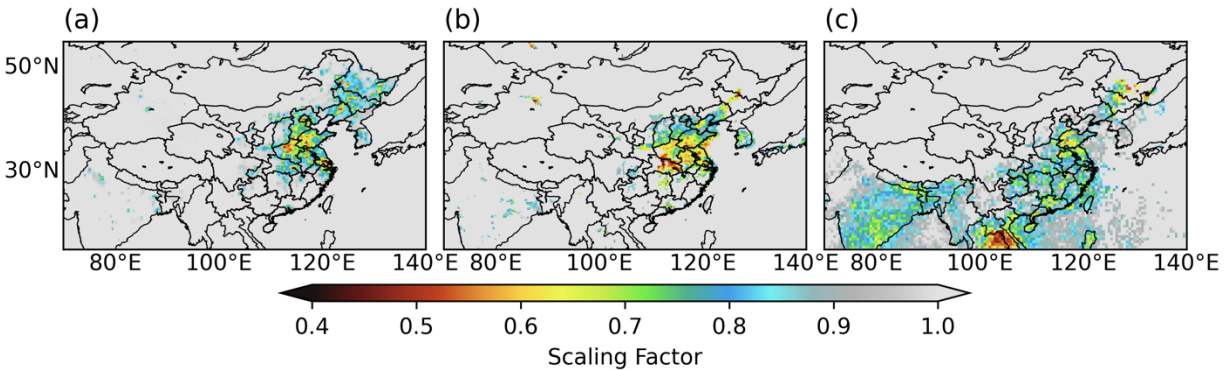


Figure 2. Scaling factors for anthropogenic NOx emission in Feb.-Mar. from 2019 to 2020 as derived from (a) 4D-Var, (b) mass balance. Scaling factors for anthropogenic VOC emissions from the mass balance are in (c).

3.2 Validation of NO₂ simulations

We further assess our updated anthropogenic NO_x emissions by comparing the NO₂ VCD from TROPOMI with that from GEOS-Chem with the anthropogenic NO_x emissions before and after the scaling (Fig. 3 and 4). Before updating the NO_x emissions, the 2020 Default (Fig. 3(b)) simulation significantly overestimates the NO₂ VCD compared to the TROPOMI NO₂ observations (Fig. 3(a)). With the NO_x emissions updated, the simulations 2020 NO_x (Fig. 3(c)) and 2020 Adjoint (Fig. 3(d)) exhibit a much better agreement with TROPOMI NO₂ observation than 2020 Default. However, Fig. 3(c) shows the GEOS-Chem simulation with the NO_x emissions from mass balance approach overestimated the NO₂ 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 NO_x emissions, not total NO_x emissions, so it is expected that the model may still overestimate the NO₂ column after scaling part of the total NO_x emission. With the anthropogenic NO_x emissions optimized by the 4D-Var method, the overestimation of NO₂ VCD over Beijing and southwest of Hebei Province (pink and black circles in Fig. 3) is mitigated compared with the NO_x emissions from mass balance approach.

Fig. 4 further displays the statistics for the comparison between the TROPOMI NO₂ and GEOS-Chem simulations via the scatterplot. The Baseline (2019) simulation captures the magnitude of NO₂ VCD observations in 2019 well (Fig. 4(a)). The root-mean-square-error (RMSE) and mean bias error (MBE) for the simulation with 2020 NO_x 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 NO_x, emissions from 2020 Adjoint (Fig. 4(d)) further led to the reduction of the MBE of the NO₂ VCD by 0.006 DU and improve the correlation coefficient by 0.003. The significant overestimation of several pixels with TROPOMI NO₂ VCD larger than 0.4 DU by the simulation 2020 NO_x is also mitigated by 2020 Adjoint. The MBE between GEOS-Chem and TROPOMI for Baseline (2019), 2020 NO_x 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 ~30% for TROPOMI tropospheric NO₂ VCD over East China (Van Geffen et al., 2022). The improved agreement between the simulation with updated NO_x emission and TROPOMI NO₂ provides a basis for further analyzing the mechanism of aggravated surface O₃ pollution.

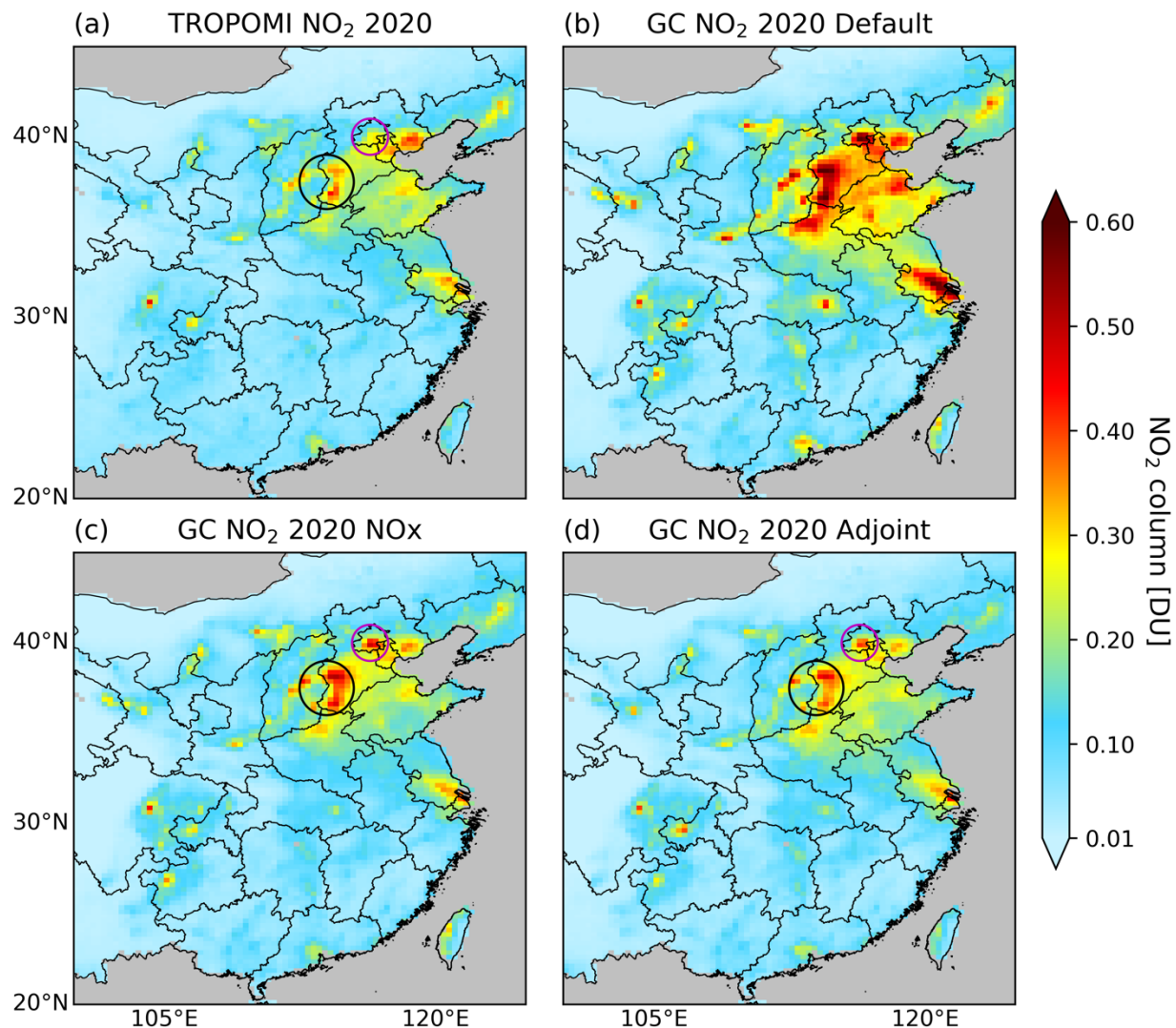


Figure 3. Comparison of tropospheric NO₂ VCD from (a) TROPOMI product in 2020 Feb.-Mar with that from GEOS-Chem simulations (b) 2020 Default, (c) 2020 NOx and (d) 2020 Adjoint. The pink and black circles mark the areas where NOx emissions from 4D-Var mitigated the NO₂ overestimation by mass balance method. The emissions and meteorology configurations for GEOS-Chem simulations 2020 Default, 2020 NOx and 2020 Adjoint are listed in Table 1.

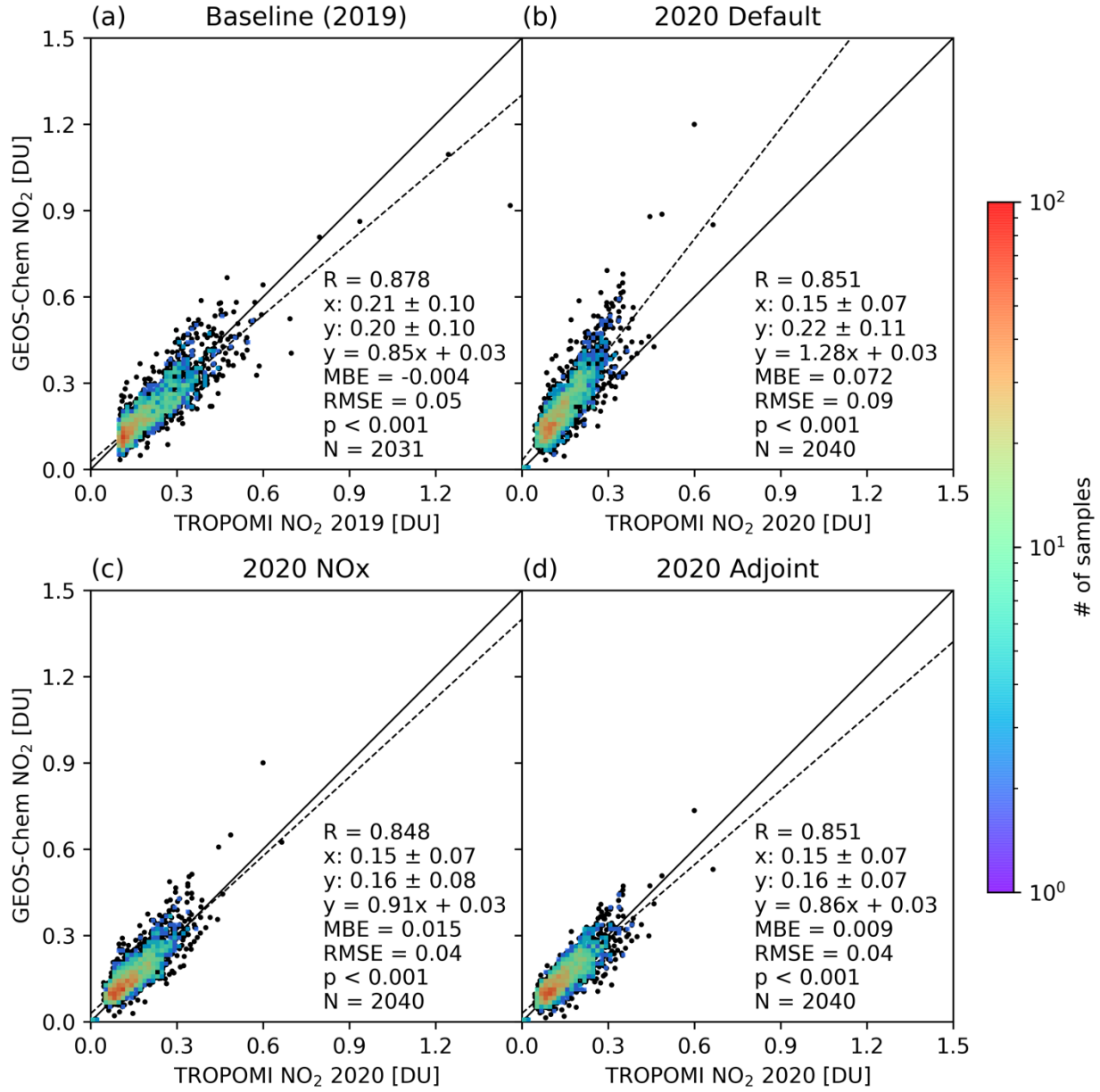


Figure 4. Scatter plot of TROPOMI NO₂ VCD versus the GEOS-Chem simulations for (a) Baseline (2019), (b) 2020 Default, (c) 2020 NO_x and (d) 2020 Adjoint, respectively. TROPOMI data in Feb.-Mar. of 2019 was used in (a), and that of 2020 was used in (b-d). The emissions and meteorology configurations for GEOS-Chem simulations are listed in Table 1. Only pixels with TROPOMI NO₂ 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 NO₂ 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 NO₂ 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 NO₂ 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 NO₂ 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 $\mu\text{g m}^{-3}$ and 8.49 $\mu\text{g m}^{-3}$, respectively. Without updating the NO_x emissions in 2020, the simulation 2020 Default overestimate the ground measurements of surface NO₂ in 2020 Feb.-Mar (Table 2). The slope for the linear regression is 1.19, and the MBE and RMSE are 6.021 $\mu\text{g m}^{-3}$ and 10.43 $\mu\text{g m}^{-3}$, respectively (Table 2). After updating the NO_x emissions, the GEOS-Chem simulations 2020 NO_x 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 NO_x versus the ground measurements (Table 2). The MBE and RMSE of 2020 Adjoint (0.683 $\mu\text{g m}^{-3}$ and 6.68 $\mu\text{g m}^{-3}$) are lower than those of 2020 NO_x (1.726 $\mu\text{g m}^{-3}$ and 7.74 $\mu\text{g m}^{-3}$) (Table 2). This result further indicates the superiority of 4D-Var for optimizing NO_x emissions compared with the mass balance method (Cooper et al., 2017; Streets et al., 2013).

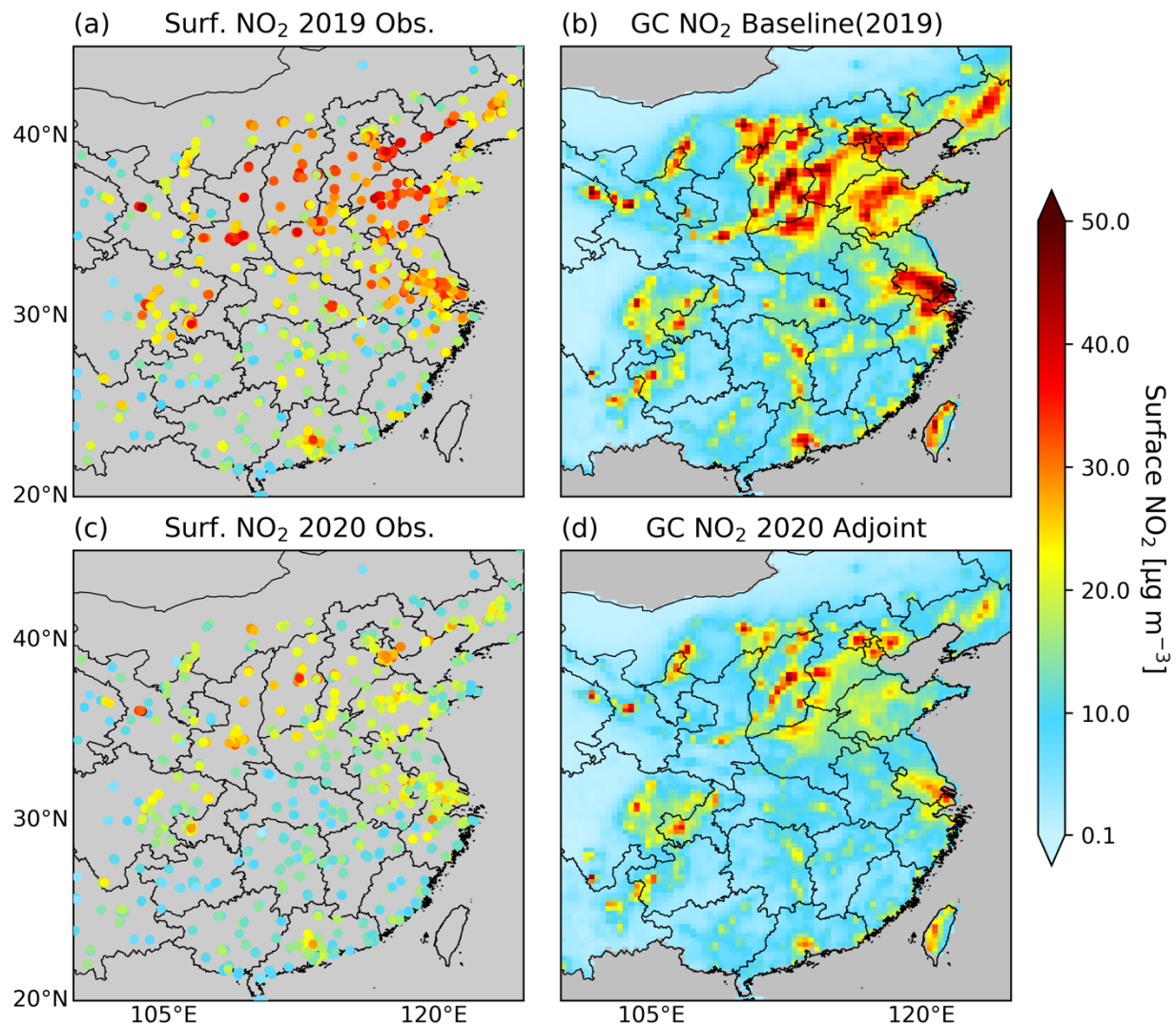


Figure 5. Comparison of surface NO_2 concentrations from ground measurements for (a) 2019 Feb.-Mar. and (c) 2020 Feb.-Mar. versus those from GEOS-Chem simulations (b) Baseline (2019) and (d) 2020 Adjoint. Grey color means no data is presented.

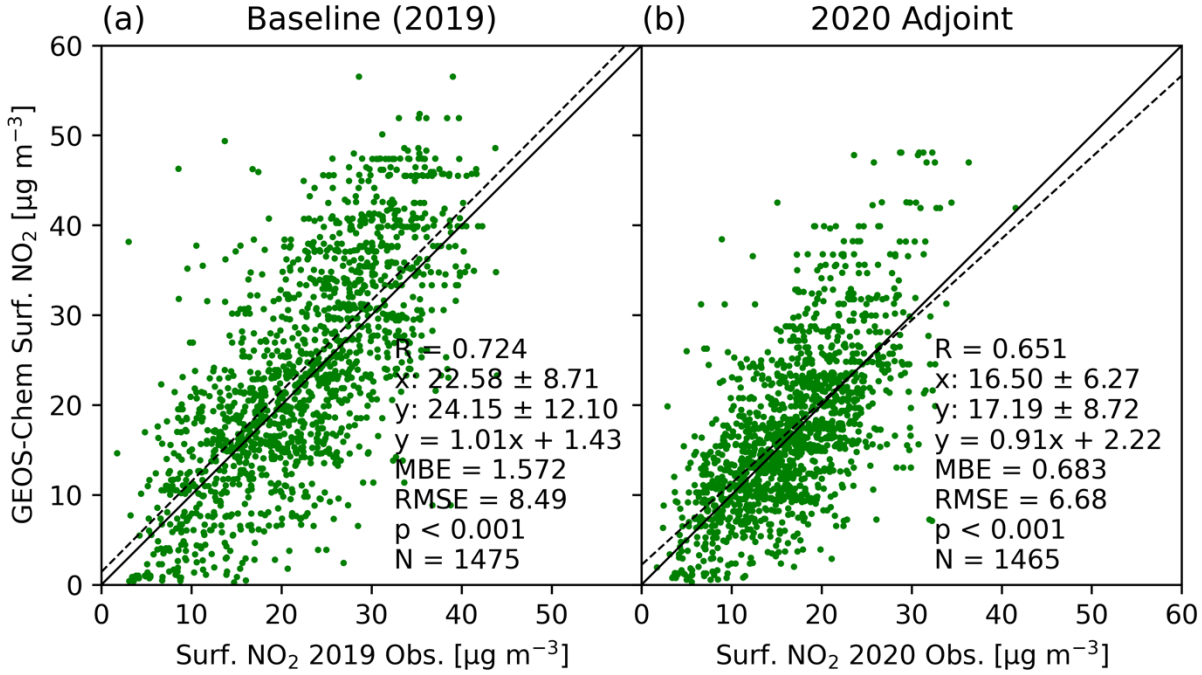


Figure 6. Scatter plots for comparing the surface NO₂ concentrations from GEOS-Chem simulations and ground measurements. (a) GEOS-Chem simulation Baseline (2019) versus ground measurements in 2019 Feb.-Mar. (b) GEOS-Chem simulation 2020 Adjoint versus ground measurements in 2020 Feb.-Mar. Note: the number of ground sites differ in these two years.

Table 2. Evaluation statistics for modeled surface NO₂ compared with the in-situ measurements*.

Experiments	R	MBE ($\mu\text{g m}^{-3}$)	RMSE ($\mu\text{g m}^{-3}$)	Slope	Intercept ($\mu\text{g m}^{-3}$)
Baseline (2019)	0.724	1.572	8.49	1.01	1.43
2020 Default	0.661	6.021	10.43	1.19	2.95
2020 NOx	0.608	1.726	7.74	0.92	3.03
2020 Adjoint	0.651	0.683	6.68	0.91	2.22

* 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 NO₂ 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 NO_x emission, 2020 Default features a relative bias of ~37%. After updating the NO_x emissions, 2020 NO_x 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 NO_x. We also validated the VOC emissions by comparing the simulated HCHO VCD with TROPOMI measurements (Appendix D).

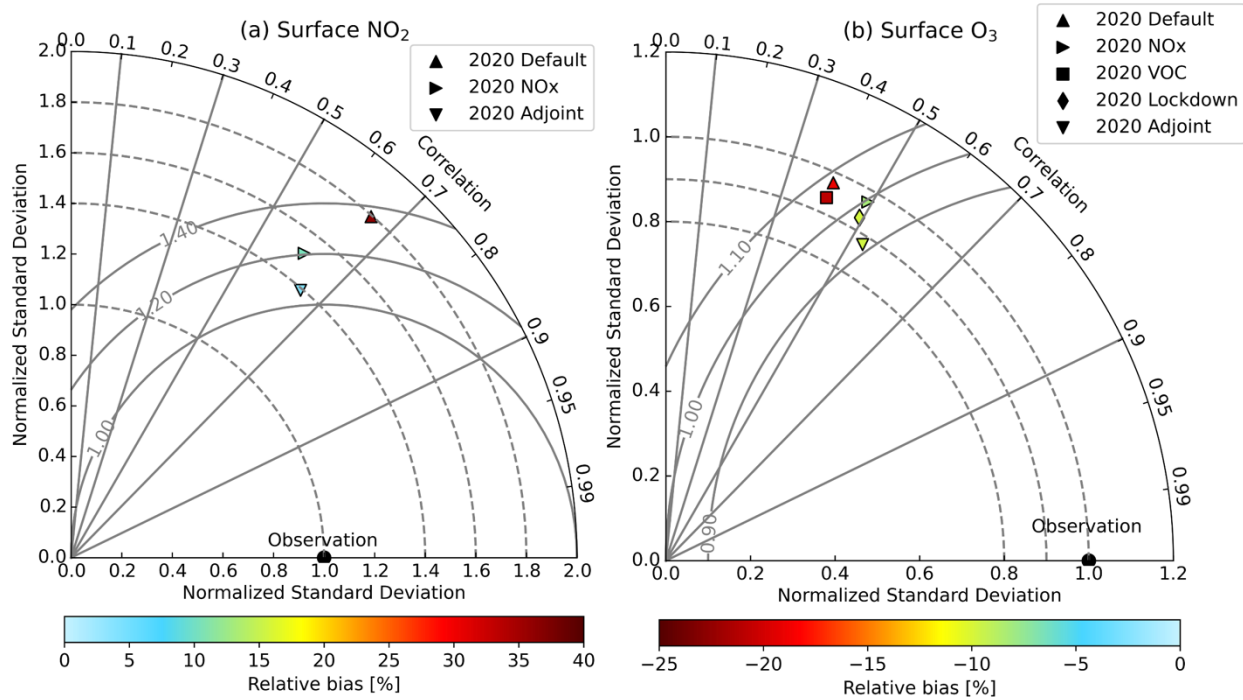


Figure 7. Taylor diagram for evaluating the GEOS-Chem simulations of (a) surface NO₂ and (b) surface O₃ during lockdown period (2020 Feb.-Mar.) using ground observations for different simulation experiments listed in Table 1. The evaluation of surface O₃ only includes the areas where the NO_x emissions optimized by 4D-Var reduced by more than 10%.

3.3 Evaluation of surface O₃ simulations

We evaluated the GEOS-Chem simulations of MDA8 surface O₃ from different simulation experiments listed in Table 1 using ground measurements. Fig. 7(b) is the Taylor diagram for comparing the surface O₃ concentrations during 2020 Feb.-Mar. from ground measurements and GEOS-Chem simulations. We focused on areas with significant NO_x emissions reduction to better assess the role of updated NO_x emissions in improving surface O₃ simulations. The ground sites where the NO_x 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 NO_x emissions, the relative bias reduced to around -10% while the correlation coefficients remain at ~0.50 for model simulations 2020 NO_x, 2020 Lockdown and 2020 Adjoint. This indicates the NO_x emission updates significantly improve the surface O₃ simulations. Comparing the simulations 2020 Default and 2020 VOC, or 2020 NO_x and 2020 Lockdown, the results show that scaling VOC emissions does not improve the surface O₃ 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 NO_x emissions. Therefore, we used the 2020 Adjoint to evaluate the impacts of NO_x emission on surface O₃ in the following analysis.

Fig. 8 compares the modeled surface O₃ 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 O₃ pollution occurs in North China and southwest of China. The average MDA8 O₃ in two months can reach up to ~110 $\mu\text{g m}^{-3}$ 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 $\mu\text{g m}^{-3}$. GEOS-Chem model underestimates the surface O₃ over North China for both years compared with ground observations, which could be a result of the underestimation of biogenic VOC emissions (Appendix D). The underestimation of the simulated O₃ over North China will not significantly affect our study purpose since this study focuses on revealing the impacts of emissions and meteorology change

on the surface O_3 change by each region. The bias is predominantly systematic and will be substantially cancelled when we compute the relative difference of the surface O_3 . The model 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)). In Sichuan Basin, the trend of the surface O_3 change from the model is opposite to the measurements, which is probably caused by the inaccurate simulation of the meteorological effects (See Section 4.1) due to the complex terrain features in this region. The localized improvement of the model simulation is further needed for the regional study. 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 O_3 is indeed small and the model is able to capture the part of O_3 decrease in the southwest part of the North China domain (Fig. 8(c)). We note that some previous studies showed large increase of O_3 in North China, but such increase is in comparison with the O_3 in the month right before the lockdown (not the same time in 2019; (Shi & Brasseur, 2020; Y. M. Liu et al., 2021)).

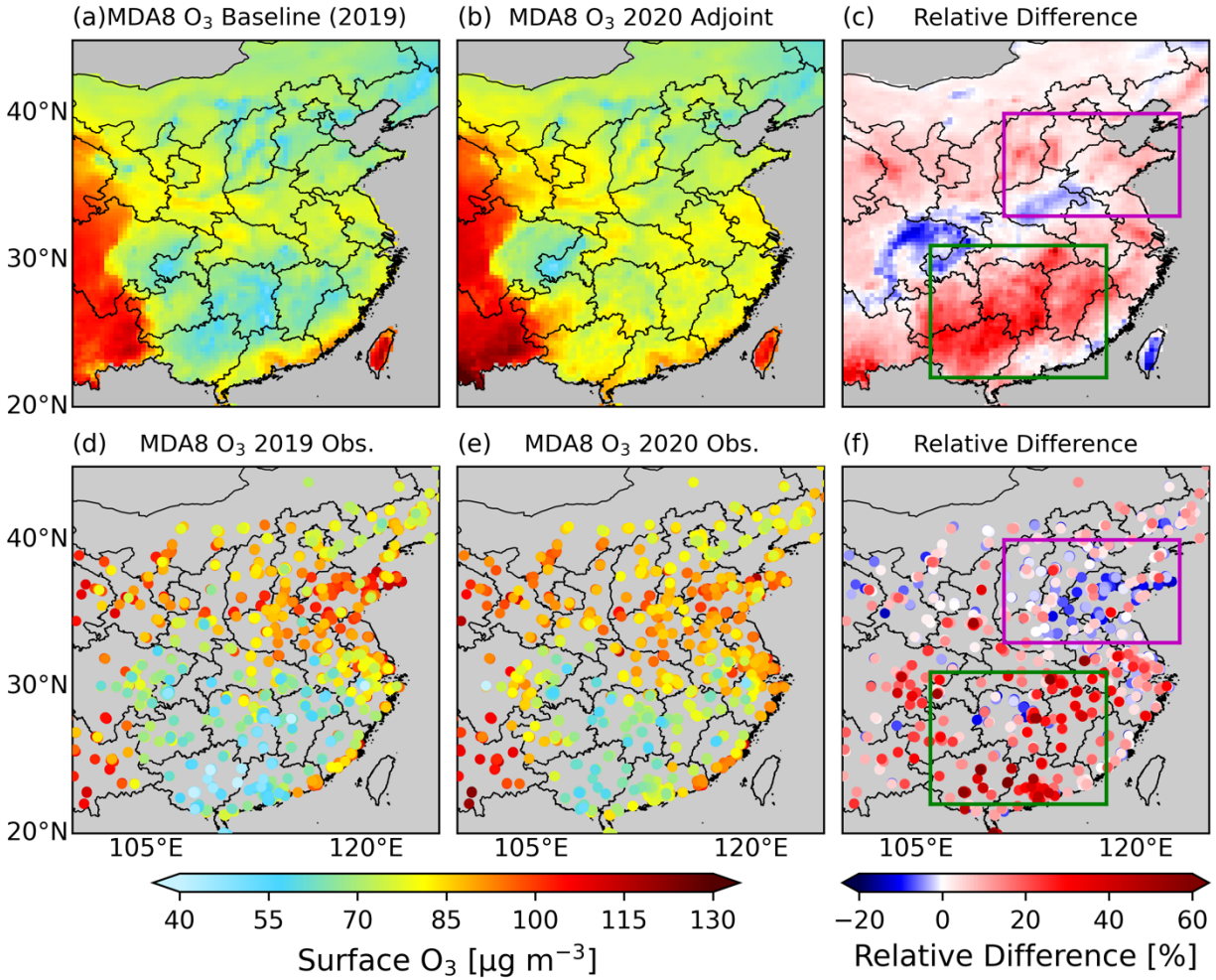


Figure 8. Comparison of MDA8 surface O₃ in 2019 and 2020 Feb.-Mar. and the relative difference between two years from GEOS-Chem model (a-c) versus ground observations (d-f). GEOS-Chem mean MDA8 O₃ at 9 m above the surface under standard temperature and pressure (STP; 273.15 K, 101.325 kPa) from (a) Baseline (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). NO_x 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 NO_x 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. Wang et al. (2022) also showed ~80% of the O₃ MDA8 increase during the COVID lockdown period is caused by the meteorological factors, and ~20% is caused by the emission decline in East China. Liu et al. (2020b) reported that the surface O₃ increase in the major cities 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), Wang et al. (2022) and Liu et al. (2020b) only focused on the lockdown period of one to five weeks in reference to the time period right before the lockdown instead of the same period in previous years, which did not provide a comprehensive analysis over the whole lockdown period, and Zhao et al. (2020) and Liu et al. (2020b) did not separate the effects of seasonal trend from the meteorological anomalies. Moreover, Liu et al. (2020b) and Wang et al. (2022) only analyzed four representative cities or the East China region 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. Previous studies found that TROPOMI NO₂ product has a negative bias of -7% to -

20% (Verhoelst et al., 2021; Judd et al., 2020; Li et al., 2021). The sensitivity simulations indicate this low bias does not significantly affect the model evaluation and our main conclusions (figures not shown).

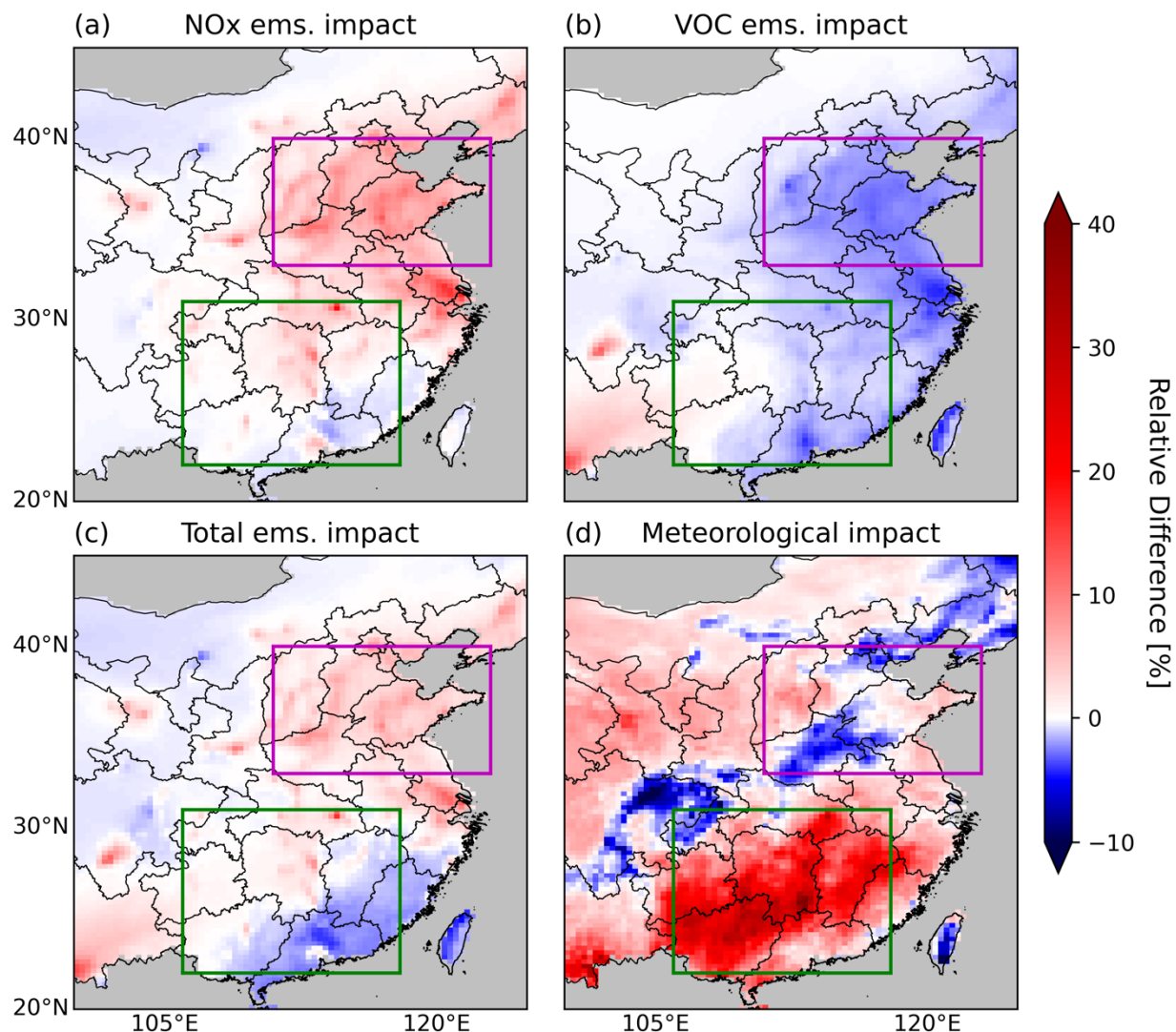


Figure 9. Relative difference in simulated surface O_3 caused by (a) NOx emission reduction, (b) VOC emission reduction, (c) overall emission reduction and (d) meteorological variations due to COVID-19 lockdown. The pink and green boxes in each panel define the North China and South China domain.

4.2 Critical meteorological variables causing aggravated surface O₃ pollution in South China

To identify the critical meteorological variables that lead to the aggravated surface O₃ pollution in South China, we investigated the correlations between the surface O₃ concentrations and 2-m air temperature, downward visible direct flux at surface, cloud fraction, relative humidity, and wind speed. 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, downward visible direct flux at surface and cloud fraction 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-c)) and North China (111°E - 123°E, 33°N - 40°N, pink box in Fig. 10(a-c)). Fig. 10 displays the difference of 2-month mean 2-meter air temperature (Fig. 10(a)), downward visible direct flux at surface (Fig. 10(b)) and cloud fraction (Fig. 10(c)) in Feb.-Mar. between 2020 and 2019 and the scatter plot between the daily difference of measured surface O₃ concentration and 2-meter air temperature (Fig. 10(d)), downward visible direct flux at surface (Fig. 10(e)) and cloud fraction (Fig. 10(f)) over both South China (green dots in Fig. 10(d-f)) and North China (pink dots in Fig. 10(d-f)). 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 temperature increase contributed to the significant aggravated surface O₃ pollution in South China. The enhanced solar radiation at surface could also promote the production of O₃ via photochemical reactions. The correlation coefficient between the daily difference of surface O₃ concentration and downward visible direct flux at surface is as high as 0.740 in South China (Fig. 10(e)). The reason for the increase of temperature and solar radiation at surface is the lower cloud fraction. Via analyzing the GEOS-FP data, we found the cloud fraction decreases by ~5% (Fig. 10(c)), and the downward visible direct flux at surface increased by 5 W m⁻² (Fig. 10(b)) 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 change of cloud fraction is negatively

correlated with the change of surface O₃ in South China with the correlation coefficient of -0.619 (Fig. 10(f)).

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(d) 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. This negative intercept is caused by the net effects of factors other than temperature, including chemistry, emissions, and other meteorological factors. It is a challenge to quantify the contributions of each individual factors, because these factors are thermodynamically or dynamically related. 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(d) 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 decreases by 2.2 µg m⁻³ in North China, although the average temperature increased in both South and North China. On the contrary to South China, the change of solar radiation at surface and cloud fraction are poorly correlated with the change of surface O₃ concentrations in North China (Fig. 10(e, f)), mainly due to a more important role of emission change in regulating the surface O₃ in North China.

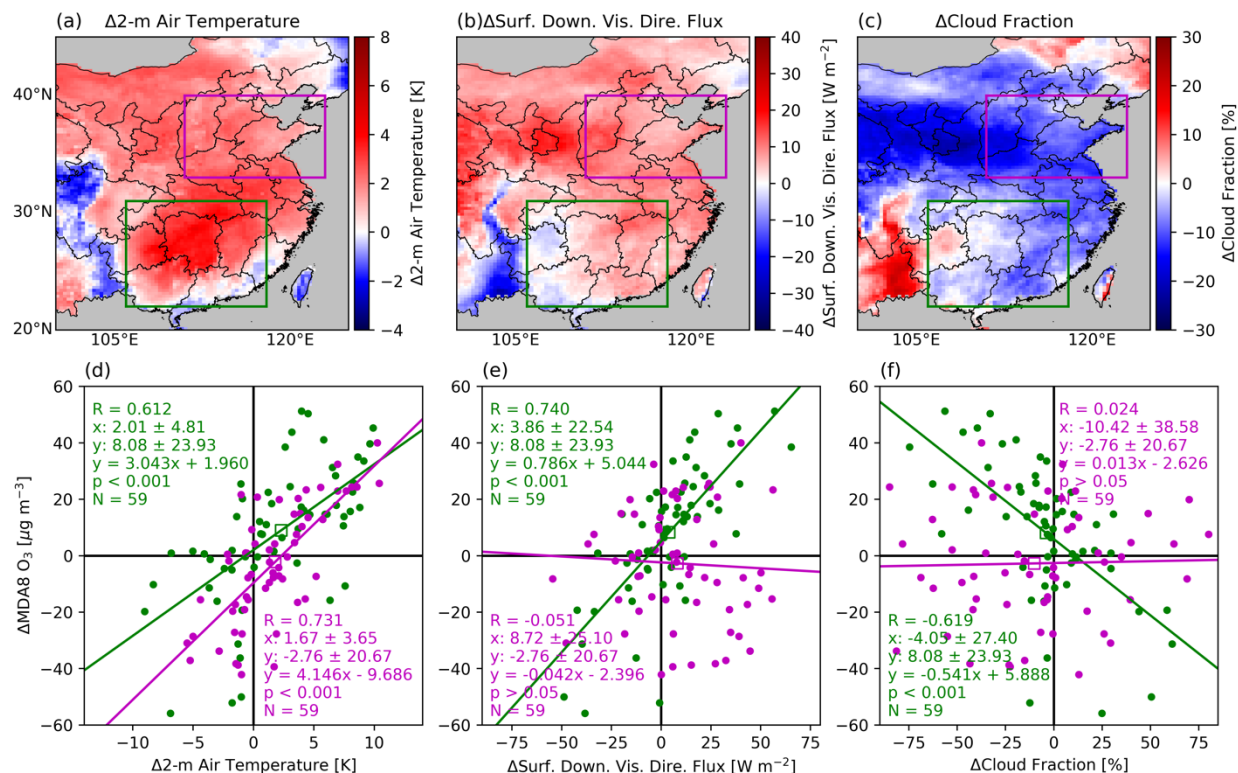


Figure 10. The changes of (a) 2-meter air temperature, (b) downward visible direct flux at surface and (c) cloud fraction from 2019 Feb.-Mar. to 2020 Feb.-Mar and the scatter plots between the daily difference of surface O₃ measurements and (d) 2-meter air temperature, (e) downward visible direct flux at surface and (f) cloud fraction in Feb.-Mar. between 2020 and 2019 over South China (green dots) and North China (pink dots). The green and pink open squares in (d-f) mark the predicted average change of surface MDA8 O₃ in South China and North China, respectively, based on the linear regression against the change of meteorological variables.

The impacts of relative humidity and wind speed on the surface O₃ change are also investigated (Fig. 11). The relative humidity increased by ~5.1% in North China and decreased by ~3.0% in South China on average. The strong correlation ($R = -0.675$) between the change of relative humidity and surface O₃ in South China indicates the decrease of relative humidity also contributes to the increase of surface O₃ pollution in South China, but the correlation between them in North China is very low. The wind speed also changed oppositely in South China and North China, but we cannot identify any significant impact of wind speed on the surface O₃ pollution since the correlation coefficients are low in both South China and North China. In summary, the significant increase of surface O₃ pollution during the lockdown period in South China could be

primarily attributed to the higher temperature, enhanced solar radiation at surface and declined relative humidity.

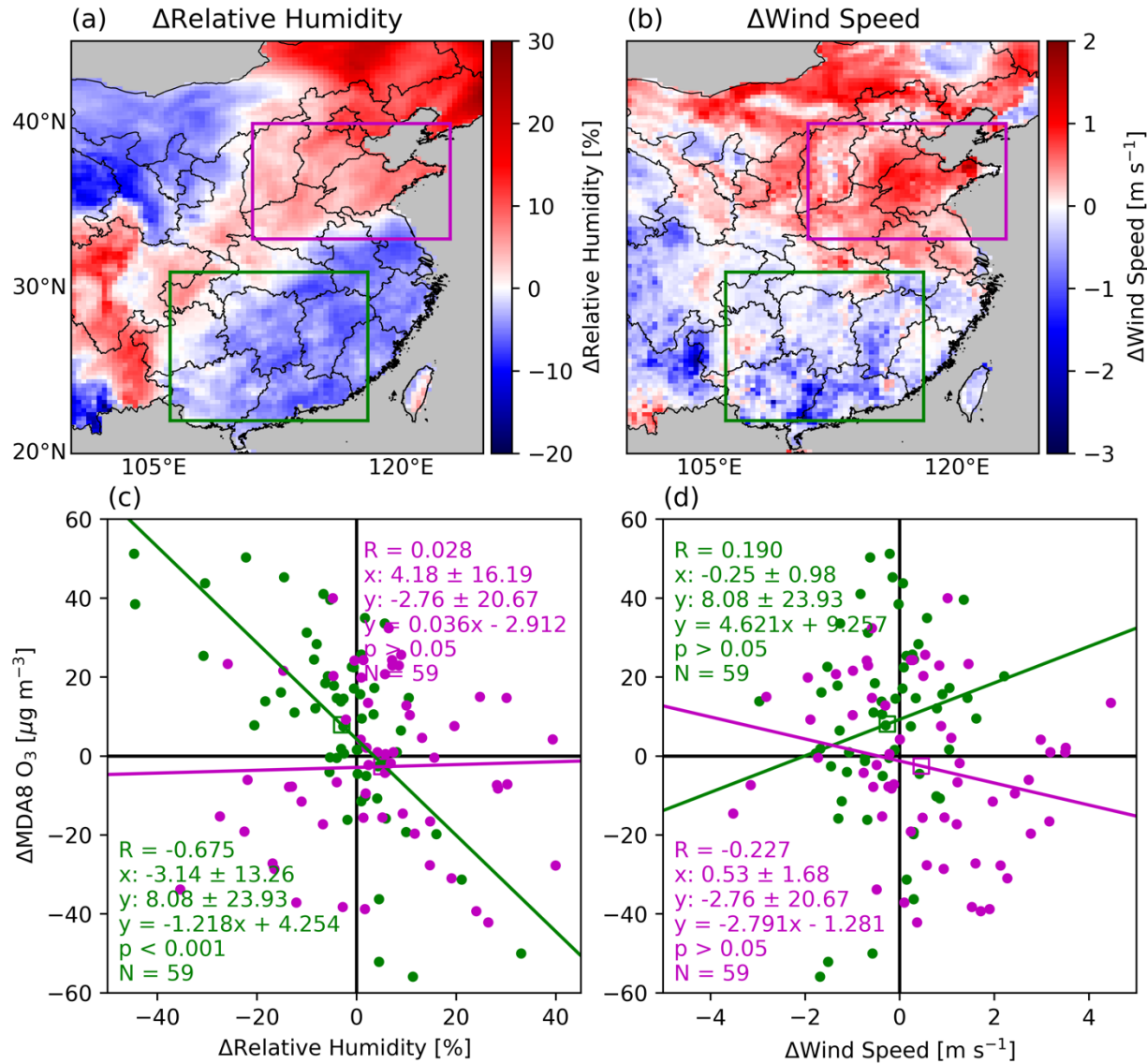


Figure 11. Same as Fig. 10 but for (a, c) relative humidity and (b, d) wind speed.

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 O₃. In this study, we analyzed the reasons for the enhanced surface O₃ pollution from two perspectives: anthropogenic emissions reduction and inter-annual meteorological variations. We constrain the NO_x emissions based on the TROPOMI

NO₂ product using both the mass balance and 4D-Var methods. The VOC emissions 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 O₃ increases through a series of sensitivity simulations using the GEOS-Chem model.

The updated NO_x emissions from the 4D-Var and mass balance approaches share a similar spatial pattern. However, the NO_x 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 NO_x emissions from the 4D-Var inversion leads to better model performance than that from the mass balance approach. The updated anthropogenic VOC emissions with mass balance method are evaluated by comparing the GEOS-Chem HCHO and TROPOMI HCHO. However, the updated VOC emissions may still suffer from large uncertainties because of the low retrieval accuracy of TROPOMI HCHO (Vigouroux et al., 2020), large biogenic sources of VOC emissions and limited representativeness of HCHO for the whole VOC species.

The anthropogenic NO_x emission decreased by ~30% over East China during 2020 Feb.-Mar. compared to the same period in 2019. Over North China, NO_x 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 NO_x 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, enhanced solar radiation at surface and declined relative humidity during the lockdown period. The lower cloud fraction increases the downward solar radiation at the surface during the lockdown period, leading to higher surface air temperature. We cannot identify any significant impact of wind speed on the surface O₃ pollution.

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

The default anthropogenic NO_x emission over East Asia in GEOS-Chem is MIX 2010 (Li et al., 2017). To generate the anthropogenic NO_x 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 NO_x emission has declined significantly as a result of the clean air actions of Chinese government (Zheng et al., 2018).

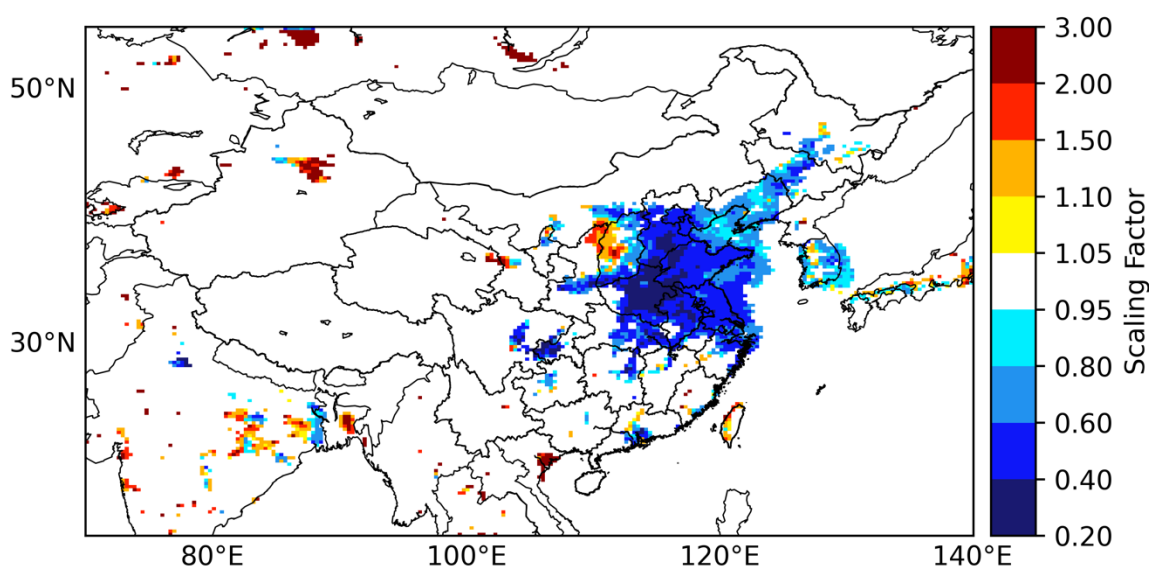


Figure A1. The scaling factor of anthropogenic NO_x emission from year 2010 to 2019.

Appendix B: Applying the TROPOMI NO₂ averaging kernel in the observation operator

To optimize the NO_x 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 NO_x 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 NO₂ concentration shown as Equation (B1).

$$F = \frac{\partial J}{\partial \mathbf{c}} = \mathbf{S}_{\text{obs}}^{-1} [H(\mathbf{c}) - \mathbf{s}] \frac{\partial H(\mathbf{c})}{\partial \mathbf{c}} \quad (\text{B1})$$

For each single TROPOMI NO₂ observation, the adjoint forcing component f and cost function component j are computed as Equation (B2) and Equation (B3).

$$f = \frac{M_{\text{gc}} v_{\text{gc}} - M_{\text{obs}} v_{\text{obs}}}{e_{\text{obs}} M_{\text{obs}}} M_{\text{gc}} \quad (\text{B2})$$

$$j = \frac{0.5 f (M_{\text{gc}} v_{\text{gc}} - M_{\text{obs}} v_{\text{obs}})}{M_{\text{gc}}} \quad (\text{B3})$$

Here M_{gc} is GEOS-Chem air mass factor applying the GEOS-Chem NO₂ vertical profiles and TROPOMI NO₂ averaging kernel. M_{obs} is TROPOMI air mass factor. v_{gc} and v_{obs} are the tropospheric NO₂ VCD from GEOS-Chem model and TROPOMI observation, respectively. The product of air mass factor and NO₂ VCD is NO₂ slant column density. e_{obs} is the standard error of TROPOMI tropospheric NO₂ VCD.

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

$$M_{\text{gc}} = \frac{\sum_{i \in \text{trop.}} c_i^{\text{gc}} \Delta p_i^{\text{gc}} w_i^{\text{gc}}}{\sum_{i \in \text{trop.}} c_i^{\text{gc}} \Delta p_i^{\text{gc}}} \quad (\text{B4})$$

Here c_i^{gc} is GEOS-Chem NO₂ mixing ratio at vertical layer i , Δp_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 NO₂ 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-B6) (Eskes and Boersma, 2003; Palmer et al., 2001).

$$w_l^{\text{TM5}} = \frac{M_{\text{obs}} A_l^{\text{TM5}}}{M_{\text{geo}}} \quad (\text{B5})$$

$$M_{\text{geo}} = \sec \theta_0 + \sec \theta \quad (\text{B6})$$

where M_{geo} is geometric air mass factor, θ_0 and θ are solar zenith angle and viewing zenith angle, respectively.

Appendix C: Validation of the TROPOMI NO₂ 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.

$$\frac{\partial J(\ln \sigma)}{\partial \ln \sigma} \approx \frac{J(\ln(\sigma + 0.05)) - J(\ln(\sigma - 0.05))}{\ln(\sigma + 0.05) - \ln(\sigma - 0.05)} \quad (C1)$$

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 $0.25^\circ \times 0.3125^\circ$. 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 NO_x emission at a much finer resolution of $0.25^\circ \times 0.3125^\circ$ than in their study ($2^\circ \times 2.5^\circ$).

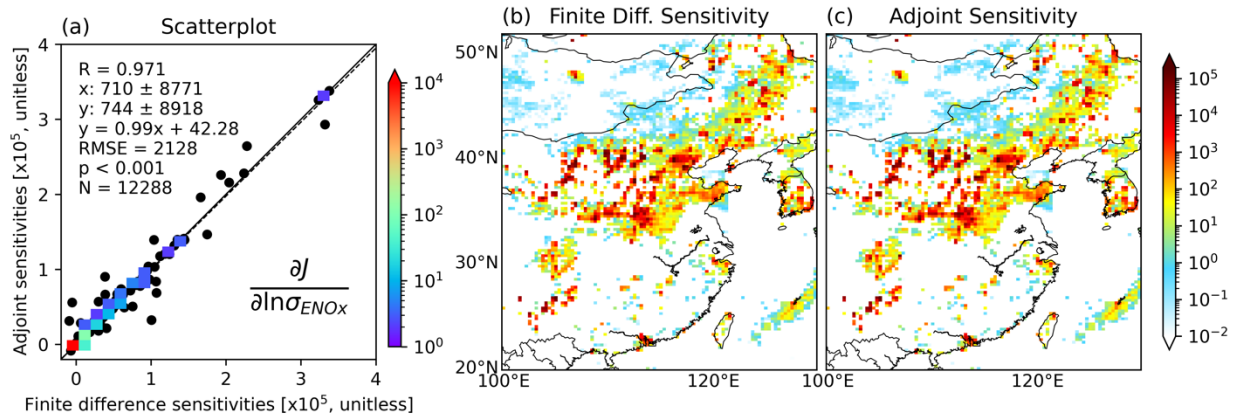


Figure C1. Comparison of adjoint sensitivities and finite difference sensitivities. (a) Scatter plot of the adjoint sensitivity of the cost function with respect to the logarithm of NO_x emission scaling factor versus the finite difference sensitivities. The color scheme for panel (a) encodes the number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

Appendix D: Validation of HCHO simulations

It is not practical to validate the VOC emissions species by species in this study. We compare the GEOS-Chem simulation of HCHO with TROPOMI data for a rough validation of the VOC emissions. Overall, the GEOS-Chem simulations Baseline (2019) and 2020 Adjoint agree well with TROPOMI HCHO in Feb.-Mar. of 2019 and 2020, respectively (Fig. D1). The correlation coefficients for the two comparisons are 0.878 and 0.874, and the MBE are -0.036 DU and -0.042 DU, respectively. This good agreement also supports our assumption of ignoring the change of VOC emissions from 2010 to 2019, since the anthropogenic VOC emission used for Baseline (2019) is equivalent to MIX 2010. However, Fig. D1 shows GEOS-Chem tends to overestimate the HCHO VCD over urban regions and underestimate it over rural regions, which indicates the partition of total emissions may overestimate the anthropogenic emissions and underestimate the biogenic emissions, at least for HCHO. A more comprehensive evaluation for various species and emission sectors would be helpful for further improvement of the VOC emissions used in the model.

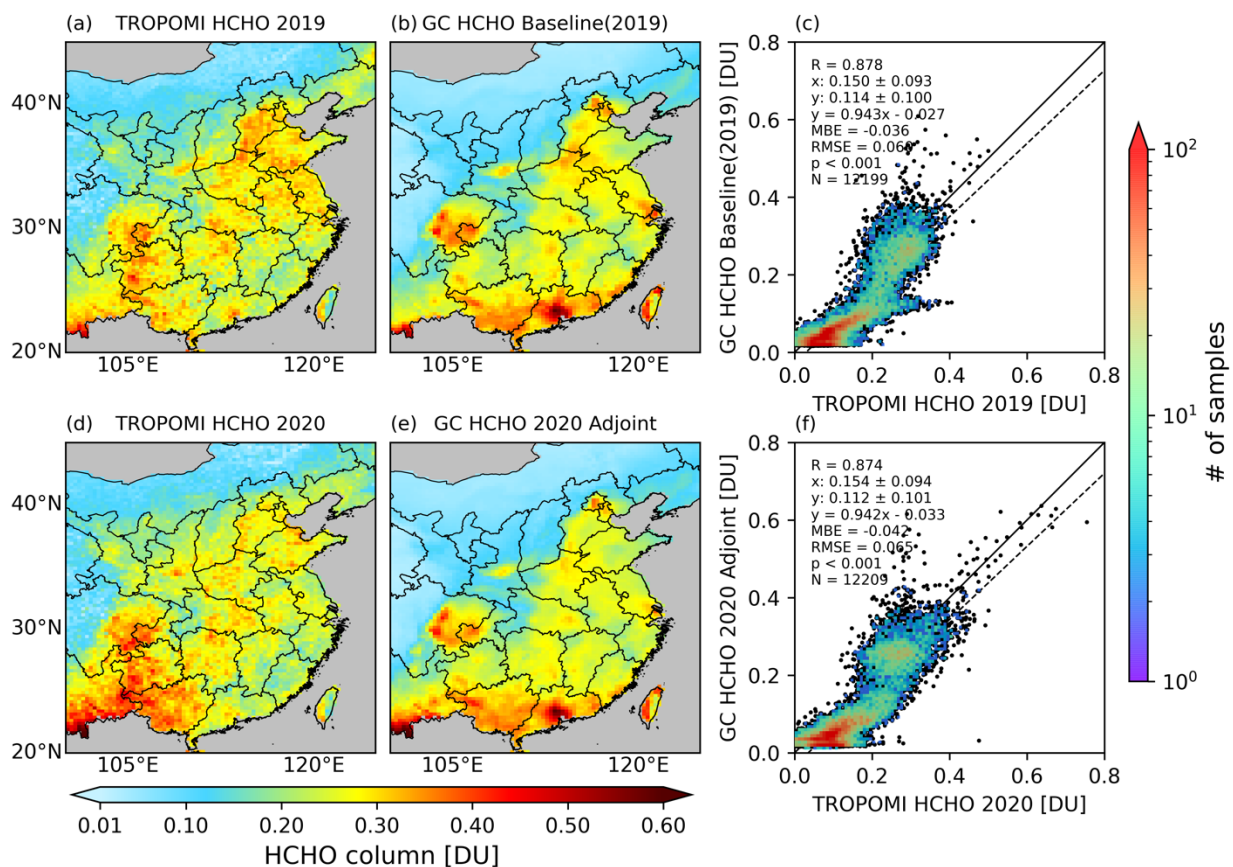


Figure D1. Comparison of tropospheric HCHO VCD from TROPOMI and GEOS-Chem. (a): TROPOMI HCHO in 2019 Feb.-Mar. (b): GEOS-Chem HCHO from Baseline (2019). (c): Scatter plot between (a) and (b). (d): TROPOMI HCHO in 2020 Feb. -Mar. (e): GEOS-Chem HCHO from 2020 Adjoint. (f): Scatter plot between (d) and (e). The emissions and meteorology configurations for GEOS-Chem simulations Baseline (2019) and 2020 Adjoint are listed in Table 1.

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>). The ground O₃ and NO₂ measurements are available at the China National Environmental Monitoring Center (<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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References

- Atkinson, R.: Atmospheric chemistry of VOCs and NO_x, *Atmospheric Environment*, 34, 2063-2101, 10.1016/s1352-2310(99)00460-4, 2000.
- Bauwens, M., Compernelle, S., Stavrakou, T., Muller, J. F., van Gent, J., Eskes, H., Levelt, P. F., van der A, R., Veefkind, J. P., Vlietinck, J., Yu, H., and Zehner, C.: Impact of Coronavirus Outbreak on NO₂ Pollution Assessed Using TROPOMI and OMI Observations, *Geophys Res Lett*, 47, 10.1029/2020GL087978, 2020.
- Bey, I., Jacob, D. J., Yantosca, R. M., Logan, J. A., Field, B. D., Fiore, A. M., Li, Q., Liu, H. Y., Mickley, L. J., and Schultz, M. G.: Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *Journal of Geophysical Research: Atmospheres*, 106, 23073-23095, 10.1029/2001jd000807, 2001.
- Bi, Z., Ye, Z., He, C., and Li, Y.: Analysis of the meteorological factors affecting the short-term increase in O₃ concentrations in nine global cities during COVID-19, *Atmospheric Pollution Research*, 13, 101523, <https://doi.org/10.1016/j.apr.2022.101523>, 2022.
- Chen, D., Wang, Y., McElroy, M. B., He, K., Yantosca, R. M., and Le Sager, P.: Regional CO pollution and export in China simulated by the high-resolution nested-grid GEOS-Chem model, *Atmospheric Chemistry and Physics*, 9, 3825-3839, 10.5194/acp-9-3825-2009, 2009.
- Cooper, M., Martin, R. V., Padmanabhan, A., and Henze, D. K.: Comparing mass balance and adjoint methods for inverse modeling of nitrogen dioxide columns for global nitrogen oxide emissions, *Journal of Geophysical Research: Atmospheres*, 122, 4718-4734, 10.1002/2016jd025985, 2017.
- De Smedt, I., Theys, N., Yu, H., Danckaert, T., Lerot, C., Compernelle, S., Van Roozendaal, M., Richter, A., Hilboll, A., Peters, E., Pedernana, M., Loyola, D., Beirle, S., Wagner, T., Eskes, H., van Geffen, J., Boersma, K. F., and Veefkind, P.: Algorithm theoretical baseline for formaldehyde retrievals from S5P TROPOMI and from the QA4ECV project, *Atmospheric Measurement Techniques*, 11, 2395-2426, 10.5194/amt-11-2395-2018, 2018.
- Eskes, H. J. and Boersma, K. F.: Averaging kernels for DOAS total-column satellite retrievals, *Atmospheric Chemistry and Physics*, 3, 1285-1291, 10.5194/acp-3-1285-2003, 2003.
- Ghahremanloo, M., Lops, Y., Choi, Y., and Mousavinezhad, S.: Impact of the COVID-19 outbreak on air pollution levels in East Asia, *Science of the Total Environment*, 754, 10.1016/j.scitotenv.2020.142226, 2021.

772 Gong, C., Lei, Y., Ma, Y., Yue, X., and Liao, H.: Ozone–vegetation feedback through dry
 773 deposition and isoprene emissions in a global chemistry–carbon–climate model, *Atmos. Chem.*
 774 *Phys.*, 20, 3841–3857, 10.5194/acp-20-3841-2020, 2020.

775 Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and
 776 Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1):
 777 an extended and updated framework for modeling biogenic emissions, *Geoscientific Model*
 778 *Development*, 5, 1471–1492, 10.5194/gmd-5-1471-2012, 2012.

779 Guo, J., Zhang, X. S., Gao, Y., Wang, Z. W., Zhang, M. G., Xue, W. B., Herrmann, H., Brasseur,
 780 G. P., Wang, T., and Wang, Z.: Evolution of Ozone Pollution in China: What Track Will It Follow?,
 781 *Environmental Science & Technology*, 57, 109–117, 10.1021/acs.est.2c08205, 2023.

782 Henze, D. K., Hakami, A., and Seinfeld, J. H.: Development of the adjoint of GEOS-Chem,
 783 *Atmospheric Chemistry and Physics*, 7, 2413–2433, 10.5194/acp-7-2413-2007, 2007.

784 Henze, D. K., Seinfeld, J. H., and Shindell, D. T.: Inverse modeling and mapping US air quality
 785 influences of inorganic PM_{2.5} precursor emissions using the adjoint of GEOS-Chem, *Atmospheric*
 786 *Chemistry and Physics*, 9, 5877–5903, 10.5194/acp-9-5877-2009, 2009.

787 Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert,
 788 J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J.-
 789 i., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., and Zhang, Q.: Historical (1750–2014)
 790 anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data
 791 System (CEDS), *Geoscientific Model Development*, 11, 369–408, 10.5194/gmd-11-369-2018,
 792 2018.

793 Huang, X., Ding, A., Gao, J., Zheng, B., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W.,
 794 Chi, X., Xu, Z., Chen, L., Li, Y., Che, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu,
 795 W., Fu, Q., Liu, B., Chai, F., Davis, S. J., Zhang, Q., and He, K.: Enhanced secondary pollution
 796 offset reduction of primary emissions during COVID-19 lockdown in China, *National Science*
 797 *Review*, 8, 10.1093/nsr/nwaa137, 2020.

798 Hudman, R. C., Moore, N. E., Mebust, A. K., Martin, R. V., Russell, A. R., Valin, L. C., and
 799 Cohen, R. C.: Steps towards a mechanistic model of global soil nitric oxide emissions:
 800 implementation and space based-constraints, *Atmospheric Chemistry and Physics*, 12, 7779–7795,
 801 10.5194/acp-12-7779-2012, 2012.

802 Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G.,
 803 Keating, T., Zhang, Q., Kurokawa, J., Wankmuller, R., van der Gon, H. D., Kuenen, J. J. P.,
 804 Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and
 805 global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution,
 806 *Atmospheric Chemistry and Physics*, 15, 11411-11432, 10.5194/acp-15-11411-2015, 2015.
 807 Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y. L., Calle, E.,
 808 and Thun, M.: Long-Term Ozone Exposure and Mortality, *New England Journal of Medicine*, 360,
 809 1085-1095, 10.1056/NEJMoa0803894, 2009.
 810 Judd, L. M., Al-Saadi, J. A., Szykman, J. J., Valin, L. C., Janz, S. J., Kowalewski, M. G., Eskes,
 811 H. J., Veefkind, J. P., Cede, A., Mueller, M., Gebetsberger, M., Swap, R., Pierce, R. B., Nowlan,
 812 C. R., Abad, G. G., Nehrir, A., and Williams, D.: Evaluating Sentinel-5P TROPOMI tropospheric
 813 NO₂ column densities with airborne and Pandora spectrometers near New York City and Long
 814 Island Sound, *Atmospheric Measurement Techniques*, 13, 6113-6140, 10.5194/amt-13-6113-2020,
 815 2020.
 816 Le, T. H., Wang, Y., Liu, L., Yang, J. N., Yung, Y. L., Li, G. H., and Seinfeld, J. H.: Unexpected
 817 air pollution with marked emission reductions during the COVID-19 outbreak in China, *Science*,
 818 369, 702-706, 10.1126/science.abb7431, 2020.
 819 Leue, C., Wenig, M., Wagner, T., Klimm, O., Platt, U., and Jähne, B.: Quantitative analysis of
 820 NO_x emissions from Global Ozone Monitoring Experiment satellite image sequences, *Journal of*
 821 *Geophysical Research: Atmospheres*, 106, 5493-5505, 10.1029/2000jd900572, 2001.
 822 Levelt, P. F., Zweers, D. C. S., Aben, I., Bauwens, M., Borsdorff, T., De Smedt, I., Eskes, H. J.,
 823 Lerot, C., Loyola, D. G., Romahn, F., Stavrakou, T., Theys, N., Van Roozendaal, M., Veefkind, J.
 824 P., and Verhoelst, T.: Air quality impacts of COVID-19 lockdown measures detected from space
 825 using high spatial resolution observations of multiple trace gases from Sentinel-5P/TROPOMI,
 826 *Atmospheric Chemistry and Physics*, 22, 10319-10351, 10.5194/acp-22-10319-2022, 2022.
 827 Li, M., McDonald, B. C., McKeen, S. A., Eskes, H., Levelt, P., Francoeur, C., Harkins, C., He, J.,
 828 Barth, M., Henze, D. K., Bela, M. M., Trainer, M., de Gouw, J. A., and Frost, G. J.: Assessment
 829 of Updated Fuel-Based Emissions Inventories Over the Contiguous United States Using
 830 TROPOMI NO₂ Retrievals, *Journal of Geophysical Research-Atmospheres*, 126,
 831 10.1029/2021jd035484, 2021.

832 Li, M., Zhang, Q., Kurokawa, J.-i., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G.,
 833 Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng,
 834 B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration
 835 framework of the MICS-Asia and HTAP, *Atmospheric Chemistry and Physics*, 17, 935-963,
 836 10.5194/acp-17-935-2017, 2017.

837 Liu, F., Page, A., Strode, S. A., Yoshida, Y., Choi, S., Zheng, B., Lamsal, L. N., Li, C., Krotkov,
 838 N. A., Eskes, H., van der A, R., Veefkind, P., Levelt, P. F., Hauser, O. P., and Joiner, J.: Abrupt
 839 decline in tropospheric nitrogen dioxide over China after the outbreak of COVID-19, *Science*
 840 *Advances*, 6, 10.1126/sciadv.abc2992, 2020a.

841 Liu, S. C., Trainer, M., Fehsenfeld, F. C., Parrish, D. D., Williams, E. J., Fahey, D. W., Hubler,
 842 G., and Murphy, P. C.: Ozone production in the rural troposphere and the implications for regional
 843 and global ozone distributions, *Journal of Geophysical Research-Atmospheres*, 92, 4191-4207,
 844 10.1029/JD092iD04p04191, 1987.

845 Liu, T., Wang, X., Hu, J., Wang, Q., An, J., Gong, K., Sun, J., Li, L., Qin, M., Li, J., Tian, J.,
 846 Huang, Y., Liao, H., Zhou, M., Hu, Q., Yan, R., Wang, H., and Huang, C.: Driving Forces of
 847 Changes in Air Quality during the COVID-19 Lockdown Period in the Yangtze River Delta
 848 Region, China, *Environmental Science & Technology Letters*, 7, 779-786,
 849 10.1021/acs.estlett.0c00511, 2020b.

850 Liu, Y. M., Wang, T., Stavrakou, T., Elguindi, N., Doumbia, T., Granier, C., Bouarar, I., Gaubert,
 851 B., and Brasseur, G. P.: Diverse response of surface ozone to COVID-19 lockdown in China,
 852 *Science of the Total Environment*, 789, 10.1016/j.scitotenv.2021.147739, 2021.

853 Lu, X., Zhang, L., Chen, Y. F., Zhou, M., Zheng, B., Li, K., Liu, Y. M., Lin, J. T., Fu, T. M., and
 854 Zhang, Q.: Exploring 2016-2017 surface ozone pollution over China: source contributions and
 855 meteorological influences, *Atmospheric Chemistry and Physics*, 19, 8339-8361, 10.5194/acp-19-
 856 8339-2019, 2019.

857 Mao, J., Paulot, F., Jacob, D. J., Cohen, R. C., Crounse, J. D., Wennberg, P. O., Keller, C. A.,
 858 Hudman, R. C., Barkley, M. P., and Horowitz, L. W.: Ozone and organic nitrates over the eastern
 859 United States: Sensitivity to isoprene chemistry, *Journal of Geophysical Research: Atmospheres*,
 860 118, 11,256-211,268, 10.1002/jgrd.50817, 2013.

861 Mao, J., Jacob, D. J., Evans, M. J., Olson, J. R., Ren, X., Brune, W. H., Clair, J. M. S., Crounse, J.
 862 D., Spencer, K. M., Beaver, M. R., Wennberg, P. O., Cubison, M. J., Jimenez, J. L., Fried, A.,

863 Weibring, P., Walega, J. G., Hall, S. R., Weinheimer, A. J., Cohen, R. C., Chen, G., Crawford, J.
 864 H., McNaughton, C., Clarke, A. D., Jaeglé, L., Fisher, J. A., Yantosca, R. M., Le Sager, P., and
 865 Carouge, C.: Chemistry of hydrogen oxide radicals (HOx) in the Arctic troposphere in spring,
 866 *Atmospheric Chemistry and Physics*, 10, 5823-5838, 10.5194/acp-10-5823-2010, 2010.
 867 Martin, R. V., Jacob, D. J., Chance, K., Kurosu, T. P., Palmer, P. I., and Evans, M. J.: Global
 868 inventory of nitrogen oxide emissions constrained by space-based observations of NO₂ columns,
 869 *Journal of Geophysical Research-Atmospheres*, 108, 10.1029/2003jd003453, 2003.
 870 Miyazaki, K., Bowman, K., Sekiya, T., Jiang, Z., Chen, X., Eskes, H., Ru, M., Zhang, Y., and
 871 Shindell, D.: Air Quality Response in China Linked to the 2019 Novel Coronavirus (COVID-19)
 872 Lockdown, *Geophys Res Lett*, 47, 10.1029/2020gl089252, 2020.
 873 Murray, L. T., Jacob, D. J., Logan, J. A., Hudman, R. C., and Koshak, W. J.: Optimized regional
 874 and interannual variability of lightning in a global chemical transport model constrained by
 875 LIS/OTD satellite data, *Journal of Geophysical Research: Atmospheres*, 117,
 876 10.1029/2012jd017934, 2012.
 877 Ott, L. E., Pickering, K. E., Stenchikov, G. L., Allen, D. J., DeCaria, A. J., Ridley, B., Lin, R.-F.,
 878 Lang, S., and Tao, W.-K.: Production of lightning NO_x and its vertical distribution calculated from
 879 three-dimensional cloud-scale chemical transport model simulations, *Journal of Geophysical*
 880 *Research*, 115, 10.1029/2009jd011880, 2010.
 881 Palmer, P. I., Jacob, D. J., Chance, K., Martin, R. V., Spurr, R. J. D., Kurosu, T. P., Bey, I.,
 882 Yantosca, R., Fiore, A., and Li, Q. B.: Air mass factor formulation for spectroscopic measurements
 883 from satellites: Application to formaldehyde retrievals from the Global Ozone Monitoring
 884 Experiment, *Journal of Geophysical Research-Atmospheres*, 106, 14539-14550,
 885 10.1029/2000jd900772, 2001.
 886 Pusede, S. E., Steiner, A. L., and Cohen, R. C.: Temperature and Recent Trends in the Chemistry
 887 of Continental Surface Ozone, *Chemical Reviews*, 115, 3898-3918, 10.1021/cr5006815, 2015.
 888 Qu, Z., Henze, D. K., Capps, S. L., Wang, Y., Xu, X., Wang, J., and Keller, M.: Monthly top-down
 889 NO_x emissions for China (2005–2012): A hybrid inversion method and trend analysis, *Journal of*
 890 *Geophysical Research: Atmospheres*, 122, 4600-4625, 10.1002/2016JD025852, 2017.
 891 Qu, Z., Henze, D. K., Li, C., Theys, N., Wang, Y., Wang, J., Wang, W., Han, J., Shim, C.,
 892 Dickerson, R. R., and Ren, X.: SO₂ Emission Estimates Using OMI SO₂ Retrievals for 2005-2017,
 893 *J Geophys Res Atmos*, 124, 8336-8359, 10.1029/2019JD030243, 2019.

894 Sha, T., Ma, X. Y., Zhang, H. X., Janecek, N., Wang, Y. Y., Wang, Y., Garcia, L. C., Jenerette,
 895 G. D., and Wang, J.: Impacts of Soil NO_x Emission on O₃ Air Quality in Rural California,
 896 Environmental Science & Technology, 55, 7113-7122, 10.1021/acs.est.0c06834, 2021.

897 Shi, X. and Brasseur, G. P.: The Response in Air Quality to the Reduction of Chinese Economic
 898 Activities During the COVID-19 Outbreak, Geophys Res Lett, 47, 10.1029/2020gl088070, 2020.

899 Sillman, S., Logan, J. A., and Wofsy, S. C.: The sensitivity of ozone to nitrogen-oxides and
 900 hydrocarbons in regional ozone episodes, Journal of Geophysical Research-Atmospheres, 95,
 901 1837-1851, 10.1029/JD095iD02p01837, 1990.

902 Steinbacher, M., Zellweger, C., Schwarzenbach, B., Bugmann, S., Buchmann, B., Ordóñez, C.,
 903 Prevot, A. S. H., and Hueglin, C.: Nitrogen oxide measurements at rural sites in Switzerland: Bias
 904 of conventional measurement techniques, Journal of Geophysical Research: Atmospheres, 112,
 905 <https://doi.org/10.1029/2006JD007971>, 2007.

906 Streets, D. G., Canty, T., Carmichael, G. R., de Foy, B., Dickerson, R. R., Duncan, B. N., Edwards,
 907 D. P., Haynes, J. A., Henze, D. K., Houyoux, M. R., Jacob, D. J., Krotkov, N. A., Lamsal, L. N.,
 908 Liu, Y., Lu, Z., Martin, R. V., Pfister, G. G., Pinder, R. W., Salawitch, R. J., and Wecht, K. J.:
 909 Emissions estimation from satellite retrievals: A review of current capability, Atmospheric
 910 Environment, 77, 1011-1042, 10.1016/j.atmosenv.2013.05.051, 2013.

911 Sun, K., Zhu, L., Cady-Pereira, K., Chan Miller, C., Chance, K., Clarisse, L., Coheur, P.-F.,
 912 González Abad, G., Huang, G., Liu, X., Van Damme, M., Yang, K., and Zondlo, M.: A physics-
 913 based approach to oversample multi-satellite, multispecies observations to a common grid,
 914 Atmospheric Measurement Techniques, 11, 6679-6701, 10.5194/amt-11-6679-2018, 2018.

915 Tong, L., Liu, Y., Meng, Y., Dai, X. R., Huang, L. J., Luo, W. X., Yang, M. R., Pan, Y., Zheng,
 916 J., and Xiao, H.: Surface ozone changes during the COVID-19 outbreak in China: An insight into
 917 the pollution characteristics and formation regimes of ozone in the cold season, Journal of
 918 Atmospheric Chemistry, 80, 103-120, 10.1007/s10874-022-09443-2, 2023.

919 Travis, K. R., Jacob, D. J., Keller, C. A., Kuang, S., Lin, J., Newchurch, M. J., and Thompson, A.
 920 M.: Resolving ozone vertical gradients in air quality models, Atmos. Chem. Phys. Discuss., 2017,
 921 1-18, 10.5194/acp-2017-596, 2017.

922 Travis, K. R., Jacob, D. J., Fisher, J. A., Kim, P. S., Marais, E. A., Zhu, L., Yu, K., Miller, C. C.,
 923 Yantosca, R. M., Sulprizio, M. P., Thompson, A. M., Wennberg, P. O., Crounse, J. D., St Clair, J.
 924 M., Cohen, R. C., Laughner, J. L., Dibb, J. E., Hall, S. R., Ullmann, K., Wolfe, G. M., Pollack, I.

925 B., Peischl, J., Neuman, J. A., and Zhou, X.: Why do Models Overestimate Surface Ozone in the
 926 Southeastern United States?, *Atmos Chem Phys*, 16, 13561-13577, 10.5194/acp-16-13561-2016,
 927 2016.

928 van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y., Rogers, B. M.,
 929 Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J., Yokelson, R. J., and Kasibhatla, P. S.:
 930 Global fire emissions estimates during 1997–2016, *Earth System Science Data*, 9, 697-720,
 931 10.5194/essd-9-697-2017, 2017.

932 TROPOMI ATBD of the total and tropospheric NO₂ data products:
 933 <https://sentinel.esa.int/documents/247904/2476257/sentinel-5p-tropomi-atbd-no2-data-products>,
 934 last access: Sep. 29.

935 van Geffen, J., Boersma, K. F., Eskes, H., Sneep, M., ter Linden, M., Zara, M., and Veefkind, J.
 936 P.: S5P TROPOMI NO₂ slant column retrieval: method, stability, uncertainties and comparisons
 937 with OMI, *Atmospheric Measurement Techniques*, 13, 1315-1335, 10.5194/amt-13-1315-2020,
 938 2020.

939 Veefkind, J. P., Aben, I., McMullan, K., Förster, H., de Vries, J., Otter, G., Claas, J., Eskes, H. J.,
 940 de Haan, J. F., Kleipool, Q., van Weele, M., Hasekamp, O., Hoogeveen, R., Landgraf, J., Snel, R.,
 941 Tol, P., Ingmann, P., Voors, R., Kruizinga, B., Vink, R., Visser, H., and Levelt, P. F.: TROPOMI
 942 on the ESA Sentinel-5 Precursor: A GMES mission for global observations of the atmospheric
 943 composition for climate, air quality and ozone layer applications, *Remote Sensing of Environment*,
 944 120, 70-83, <https://doi.org/10.1016/j.rse.2011.09.027>, 2012.

945 Venter, Z. S., Aunan, K., Chowdhury, S., and Lelieveld, J.: COVID-19 lockdowns cause global
 946 air pollution declines, *Proceedings of the National Academy of Sciences*, 117, 18984-18990,
 947 doi:10.1073/pnas.2006853117, 2020.

948 Verhoelst, T., Compernelle, S., Pinardi, G., Lambert, J. C., Eskes, H. J., Eichmann, K. U., Fjæraa,
 949 A. M., Granville, J., Niemeijer, S., Cede, A., Tiefengraber, M., Hendrick, F., Pazmiño, A., Bais,
 950 A., Bazureau, A., Boersma, K. F., Bogner, K., Dehn, A., Donner, S., Elokhov, A., Gebetsberger,
 951 M., Goutail, F., de la Mora, M. G., Gruzdev, A., Gratsea, M., Hansen, G. H., Irie, H., Jepsen, N.,
 952 Kanaya, Y., Karagkiozidis, D., Kivi, R., Kreher, K., Levelt, P. F., Liu, C., Müller, M., Comas, M.
 953 N., Piters, A. J. M., Pommereau, J. P., Portafaix, T., Prados-Roman, C., Puertedura, O., Querel,
 954 R., Remmers, J., Richter, A., Rimmer, J., Cárdenas, C. R., de Miguel, L. S., Sinyakov, V. P.,
 955 Stremme, W., Strong, K., Van Roozendaal, M., Veefkind, J. P., Wagner, T., Wittrock, F., González,

956 M. Y., and Zehner, C.: Ground-based validation of the Copernicus Sentinel-5P TROPOMI NO₂
 957 measurements with the NDACC ZSL-DOAS, MAX-DOAS and Pandonia global networks,
 958 Atmospheric Measurement Techniques, 14, 481-510, 10.5194/amt-14-481-2021, 2021.

959 Vigouroux, C., Langerock, B., Aquino, C. A. B., Blumenstock, T., Cheng, Z. B., De Mazière, M.,
 960 De Smedt, I., Grutter, M., Hannigan, J. W., Jones, N., Kivi, R., Loyola, D., Lutsch, E., Mahieu, E.,
 961 Makarova, M., Metzger, J. M., Morino, I., Murata, I., Nagahama, T., Notholt, J., Ortega, I., Palm,
 962 M., Pinardi, G., Röhling, A., Smale, D., Stremme, W., Strong, K., Sussmann, R., Té, Y., van
 963 Roozendaal, M., Wang, P. C., and Winkler, H.: TROPOMI-Sentinel-5 Precursor formaldehyde
 964 validation using an extensive network of ground-based Fourier-transform infrared stations,
 965 Atmospheric Measurement Techniques, 13, 3751-3767, 10.5194/amt-13-3751-2020, 2020.

966 Vinken, G. C. M., Boersma, K. F., van Donkelaar, A., and Zhang, L.: Constraints on ship NO_x
 967 emissions in Europe using GEOS-Chem and OMI satellite NO₂ observations, Atmos. Chem. Phys.,
 968 14, 1353-1369, 10.5194/acp-14-1353-2014, 2014.

969 Wang, H. L., Huang, C., Tao, W., Gao, Y. Q., Wang, S. W., Jing, S. A., Wang, W. J., Yan, R. S.,
 970 Wang, Q., An, J. Y., Tian, J. J., Hu, Q. Y., Lou, S. R., Pöschl, U., Cheng, Y. F., and Su, H.:
 971 Seasonality and reduced nitric oxide titration dominated ozone increase during COVID-19
 972 lockdown in eastern China, Npj Climate and Atmospheric Science, 5, 10.1038/s41612-022-00249-
 973 3, 2022.

974 Wang, Y., Wang, J., Xu, X., Henze, D. K., Qu, Z., and Yang, K.: Inverse modeling of SO₂ and
 975 NO_x emissions over China using multisensor satellite data – Part 1: Formulation and sensitivity
 976 analysis, Atmospheric Chemistry and Physics, 20, 6631-6650, 10.5194/acp-20-6631-2020, 2020a.

977 Wang, Y., Wang, J., Zhou, M., Henze, D. K., Ge, C., and Wang, W.: Inverse modeling of SO₂ and
 978 NO_x emissions over China using multisensor satellite data – Part 2: Downscaling techniques for
 979 air quality analysis and forecasts, Atmospheric Chemistry and Physics, 20, 6651-6670,
 980 10.5194/acp-20-6651-2020, 2020b.

981 Wang, Y. X., McElroy, M. B., Jacob, D. J., and Yantosca, R. M.: A nested grid formulation for
 982 chemical transport over Asia: Applications to CO, Journal of Geophysical Research: Atmospheres,
 983 109, n/a-n/a, 10.1029/2004jd005237, 2004.

984 Williams, J. and Koppmann, R.: Volatile Organic Compounds in the Atmosphere: An Overview,
 985 in: Volatile Organic Compounds in the Atmosphere, 1-32,
 986 <https://doi.org/10.1002/9780470988657.ch1>, 2007.

987 Zhang, L., Jacob, D. J., Knipping, E. M., Kumar, N., Munger, J. W., Carouge, C. C., van Donkelaar,
 988 A., Wang, Y. X., and Chen, D.: Nitrogen deposition to the United States: distribution, sources, and
 989 processes, *Atmospheric Chemistry and Physics*, 12, 4539-4554, 10.5194/acp-12-4539-2012, 2012.
 990 Zhang, Q., Pan, Y., He, Y., Walters, W. W., Ni, Q., Liu, X., Xu, G., Shao, J., and Jiang, C.:
 991 Substantial nitrogen oxides emission reduction from China due to COVID-19 and its impact on
 992 surface ozone and aerosol pollution, *Science of The Total Environment*, 753,
 993 10.1016/j.scitotenv.2020.142238, 2021.
 994 Zhang, R. X., Zhang, Y. Z., Lin, H. P., Feng, X., Fu, T. M., and Wang, Y. H.: NO_x Emission
 995 Reduction and Recovery during COVID-19 in East China, *Atmosphere*, 11,
 996 10.3390/atmos11040433, 2020.
 997 Zhang, Y.-L. and Cao, F.: Fine particulate matter (PM_{2.5}) in China at a city level, *Scientific*
 998 *Reports*, 5, 14884, 10.1038/srep14884, 2015.
 999 Zhao, Y., Zhang, K., Xu, X., Shen, H., Zhu, X., Zhang, Y., Hu, Y., and Shen, G.: Substantial
 1000 Changes in Nitrogen Dioxide and Ozone after Excluding Meteorological Impacts during the
 1001 COVID-19 Outbreak in Mainland China, *Environmental Science & Technology Letters*, 7, 402-
 1002 408, 10.1021/acs.estlett.0c00304, 2020.
 1003 Zheng, B., Tong, D., Li, M., Liu, F., Hong, C., Geng, G., Li, H., Li, X., Peng, L., Qi, J., Yan, L.,
 1004 Zhang, Y., Zhao, H., Zheng, Y., He, K., and Zhang, Q.: Trends in China's anthropogenic emissions
 1005 since 2010 as the consequence of clean air actions, *Atmospheric Chemistry and Physics*, 18,
 1006 14095-14111, 10.5194/acp-18-14095-2018, 2018.
 1007