



## NO<sub>x</sub> emissions changes from 2019 to 2021 in Eastern China as estimated through variational inversions and TROPOMI satellite data

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### Abstract

20 China is one of the largest emitters of nitrogen oxides NO<sub>x</sub> (= NO + NO<sub>2</sub>) globally. With ongoing efforts and global pressure to reduce its emissions, up-to-date estimates of NO<sub>x</sub> emissions remain essential. In this study we focus on the evolution of NO<sub>x</sub> emissions over Eastern China (101.75-132.25°E; 17.75-50.25°N) between 2019 and 2021, including the analysis of the impact of the COVID-19 outbreak and the Chinese Lunar New Year national break (LNY) on NO<sub>x</sub> emissions. We exploit the 25 high spatial resolution and coverage of the TROPOMI Sentinel-5 Precursor nitrogen dioxide (NO<sub>2</sub>) observations, and provide an estimate of this evolution down to the level of Chinese provinces. We assimilate TROPOMI NO<sub>2</sub> observations in NO<sub>x</sub> atmospheric inversions based on the variational inversion drivers of the Community Inversion Framework (CIF), coupled to a 0.5° resolution configuration of the CHIMERE regional chemistry transport model.

30 Our results show a decrease in NO<sub>x</sub> anthropogenic emissions in most of Eastern China during early 2020 compared to 2019. This decrease approaches -40% in February 2020 as compared to 2019 due to i) the lockdowns and strict regulations on mobility associated with the COVID-19 outbreak (lockdowns extended from 23 January to 8 April in Wuhan, and from late January or early February 2020 in other parts of China, extending till March 2020) and ii) the celebrations of the LNY that took 35 place during February 2020, before the lockdowns were imposed. In some Chinese provinces, such as Shanghai, Qinghai, Jiangsu, Hubei, Henan and Beijing, the reductions in NO<sub>x</sub> emissions were -38%, -29%, -31%, -36%, -24%, and -16% respectively during February 2020 relative to 2019. The annual total (anthropogenic + biogenic) emissions of NO<sub>x</sub> in Eastern China decreased by -0.2 TgNO<sub>2</sub>/year in 2020, compared to 2019. However, in 2021, our estimates show an annual increase of 40 +4% in NO<sub>x</sub> emissions as compared to 2019, amounting to 16.7 TgNO<sub>2</sub>/year in 2021.

This study advances emissions quantification by delivering NO<sub>x</sub> emissions down to the scale of Chinese provinces, a refinement that is not always addressed in regional inversions. Our approach, therefore, makes it possible to evaluate targeted policy interventions by resolving sub-national



emissions. This shows how crucial it is to use high-resolution data when working to improve air quality  
45 and address climate change.

## 1. Introduction

Besides being one of the most populated countries (1.4 Billion inhabitants in 2019 according to the Chinese census data (NBSC, 2023)), China's exports are the highest globally (\$2.75T vs. \$1.52T in the United States (U.S.) that comes next (OEC, 2022); China is the leading manufacturing country as it  
50 accounted for 29% out of the global manufacturing output during 2023 (China Power Team, 2024). Such industrial activities are directly linked to high levels of energy consumption and fossil fuel use (Abdelwahab et al., 2024). Since 2005, China has also been considered the main emitter of nitrogen oxides ( $\text{NO}_x = \text{NO} + \text{NO}_2$ ) in the world, followed by the U.S. and then the European Union (Wang et al., 2022).  $\text{NO}_x$  emissions are mainly driven by fossil fuel combustion, for instance due to transportation  
55 (Zheng et al., 2018a), power plants (Tang et al., 2020), and some types of industrial processes (e.g. steel, iron and cement productions among others) (Bo et al., 2021; J. Liu et al., 2021).

$\text{NO}_x$  are air pollutants that are crucial to tropospheric chemistry. For instance, the photo-oxidation of  $\text{NO}_x$  is a major source of nitrous acid (HONO), the latter accelerates the formation of tropospheric ozone ( $\text{O}_3$ ) (Song et al., 2023).  $\text{O}_3$ , when in the troposphere, is damaging to human health (Donzelli & Suarez-Varela, 2024) and to the ecosystem (Agathokleous et al., 2020).  $\text{NO}_x$  gases act as oxidizing agents and therefore intervene in the chemistry of volatile organic compounds (VOCs) (Atkinson, 2000), these gases have been linked to a variety of physical diseases (Soni et al., 2018). Due to their role in particulate matter (PM) formation and particle growth,  $\text{NO}_x$  gases were found to be linked to haze episodes in China (H. He et al., 2014). In addition to their impact on the environment,  $\text{NO}_x$  emissions are one of the drivers of the increase of premature death due to deteriorated air quality, with  
60 numbers of deaths that are expected to be higher in the future (Bressler, 2021). In a study carried out in the Netherlands, Zock et al. (2018) found a positive link between air pollution and diseases such as asthma, diabetes, headaches, among others; they included in their analysis pollution by PM ( $\text{PM}_{2.5} < 2.5 \mu\text{m}$ , and  $\text{PM}_{10} < 10 \mu\text{m}$ ) and  $\text{NO}_2$  gas. The exposure to high amounts of  $\text{NO}_x$  gases increases the  
65 risk of mental health disorders (e.g., anxiety, depression, and even suicide attempts), as Shaw and Van Heyst (2022) showed a positive correlation between the risk ratio -referring to the increased risk linked to an increase of  $\text{NO}_2$  concentration by  $10 \mu\text{g} \cdot \text{m}^{-3}$  and the exposure rate of  $\text{NO}_2$ . Ju et al. (2023) have  
70 also demonstrated the impact of  $\text{NO}_x$ -related air pollutants ( $\text{O}_3$  and  $\text{PM}_{2.5}$ ) on mental health in highly polluted cities over China.

75 In 2006, the Chinese government put in place the 11<sup>th</sup> 5-year plan (2006 – 2010) (NPC, 2006) to control air pollutants such as sulfur dioxide ( $\text{SO}_2$ ) and PM. In 2011,  $\text{NO}_x$  control was added to the 12<sup>th</sup> 5-year plan (2011 – 2015), to reduce the annual Chinese  $\text{NO}_x$  emissions by 10% compared to 2010. And later in 2018, an action plan for a duration of three years was introduced, this plan aimed at reducing the emissions of both  $\text{SO}_2$  and  $\text{NO}_x$  emissions gases by a minimum 15% as compared to 2015  
80 (S. Li et al., 2023). Chinese policies seem to succeed in reducing the  $\text{NO}_x$  emissions and therefore the  $\text{NO}_2$  concentrations after 2010 (Ding et al., 2017), and remarkably, the gross domestic product (GDP) started decoupling from the  $\text{NO}_x$  emissions in 2011 (S. Li et al., 2023). Nevertheless, the decrease in  $\text{NO}_2$  concentrations did not necessarily lead to an equivalent enhancement in air quality. Some studies argued that the alteration in the balance of the VOC/ $\text{NO}_x$  ratio did in fact lead to an increase in surface



85  $O_3$  concentrations (R. Li et al., 2024; Lu et al., 2023) causing haze episodes in Shanghai (Le et al., 2020). An accurate account of  $NO_x$  emissions in space and time is needed to i) assess the effectiveness of policies aimed at reducing  $NO_x$  emissions and ii) to understand the impact of the evolution of  $NO_x$  emissions and of  $NO_2$  concentrations on air quality.

90 The quantification of anthropogenic  $NO_x$  emissions following a bottom-up (BU) approach, based on the statistics of activity sectors and fuel consumption and relying on emission factors per activity type, suffers from relatively large uncertainties at the national and annual scales (Ding et al., 2017) and bear, in addition to that, large spatial and temporal differences (Ding et al., 2017; Jena et al., 2015). Saikawa et al. (2017) have compared five different inventories focusing on the estimates of Chinese  $NO_x$  emissions, including the Regional Emission inventory in ASia v2.1 (REAS), the Multi-resolution Emission Inventory for China (MEIC), the Emission Database for Global Atmospheric Research v4.2 (EDGAR), the inventory by Yu Zhao (ZHAO), and the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS). They have shown that large discrepancies between the BU estimates were mainly related to different statistical data and emission factors for the energy and transport sectors, as inventories used different provincial statistics (Saikawa et al., 2017). To develop accurate, high-resolution emission inventories, integrating independent information sources is essential. Atmospheric measurements can serve as a valuable complement to current BU approaches, thereby enhancing the spatial precision of emission inventories. These improvements are crucial for effectively assessing air quality policies.

105 Since the 2000s,  $NO_2$  tropospheric concentrations have been monitored around the world by space-borne instruments, such as the Global Ozone Monitoring Experiment (GOME) (Burrows et al., 1999) and GOME-2 (Munro et al., 2016; Munro et al., 2016), the SCanning Imaging Absorption spectrometer for Atmospheric CHartographY (SCIAMACHY) (Bovensmann et al., 1999; Burrows et al., 1995) and the Ozone Monitoring Instrument (OMI) (Levelt et al., 2018). In this context, attempts have been made to develop so-called top-down (TD) methods, complementary to BU inventories, to deduce  $NO_x$  emissions from  $NO_2$  satellite data. The OMI  $NO_2$  tropospheric vertical column densities (TVCDs) have been exploited previously to estimate  $NO_x$  emissions in China (Gu et al., 2014; Qin et al., 2023; Savas et al., 2023). For example, Savas et al. (2023) estimated the evolution of anthropogenic  $NO_x$  emissions in 2015 and 2019 compared to 2010 over Eastern China. They have found a decrease of  $NO_x$  emissions in the southern part of Eastern China over large urbanized and industrialized locations but an increase in the northern part in 2015 and 2019 compared to 2010.

110 The successor of OMI, the TROPOspheric Monitoring Instrument (TROPOMI) (Veefkind et al., 2012), on board the Copernicus Sentinel-5 Precursor (S-5P) satellite launched in 2017, has brought images of  $NO_2$  with higher spatial resolution (pixel size of about 5.6 km  $\times$  3.5 km since August 2019). With a swath as wide as approximately 2600 km on ground, TROPOMI also provides daily coverage. This higher spatial resolution increases the potential to quantify local emissions (from specific provinces, urban or industrial areas, sometimes from specific large industrial sites). It also supports improved precision in  $NO_x$  emission estimates at large scales by offering better insights into spatial heterogeneity in chemistry and emission processes (e.g., industrial vs. urban areas) at fine scales.

120 Both OMI and TROPOMI  $NO_2$  observations have also been used to quantify the impact on  $NO_2$  concentrations of the Lunar New Year national holiday (LNY), which is a 7 to 14-days of celebration with businesses and factories closed that takes place every year around late January – mid February



(Bauwens et al., 2020; Chu et al., 2021; Cooper et al., 2022; Pei et al., 2020; Tan et al., 2009). By the end of 2019, and in the middle of the 3-year action plan (2018 – 2020), the COVID-19 pandemic broke in Wuhan. The latter is one of the multiple megacities in China that hosted 11.21 million inhabitants in 130 2019 (HKTDC Research, 2022). As a response to the increasing rates of infections and deaths caused by the COVID-19 virus, the Chinese government implemented strict mobility regulations across various provinces starting early 2020 and extending until early April 2020 in some cases. The applied measures varied based on the number of infection cases reported (Chinazzi et al., 2020; R. Li et al., 2020). For instance, in Wuhan, a full 76-day lockdown was imposed, the longest in China, and it 135 extended from the 23<sup>rd</sup> of January 2020 up until the 8<sup>th</sup> of April 2020 (R. Li et al., 2020). In other Chinese cities, shorter lockdown periods accompanied by lighter measures were imposed, the periods varied between 3 and 60 days. Due to the reduction in mobility and the stay-home rule imposed by the Chinese government, the NO<sub>2</sub> TVCDs and the NO<sub>x</sub> emissions were expected to have significantly dropped during the period of the lockdown (Bauwens et al., 2020; Ding et al., 2020; Zhang et al., 2020; 140 Levelt et al., 2022; H. Li et al., 2024; S. Li et al., 2023).

If all the inverse modeling studies about the COVID-19 crisis agree that NO<sub>x</sub> emissions were indeed reduced during the lockdown period in 2020 (T.-L. He et al., 2022; Q. Zhang et al., 2020; R. Zhang et al., 2020; Wei et al., 2023; Ding et al., 2020; H. Li et al., 2024), they differ on the extent of these reductions. The simplest approaches extrapolate the relationship between the tropospheric 145 concentrations and the emissions from a single perturbation of the emissions in the CTM simulations, and from the assumption that this relationship is purely local (occurring within the scale of the CTM grid cell) (H. Li et al., 2024; Zheng et al., 2021). These simplifications and assumptions provide a limited capability to account for the chemistry. A more detailed account of the complex NO<sub>x</sub> chemistry with more elaborate approaches using chemistry-transport models (CTMs) with ensemble Kalman filter 150 inverse modelling techniques or variational approaches should support more accurate derivations of NO<sub>x</sub> emissions from NO<sub>2</sub> satellite data.

In this study, we leverage the high spatial resolution and extensive coverage of NO<sub>2</sub> tropospheric columns provided by TROPOMI instrument, onboard the Sentinel 5-Precursor satellite, to estimate 155 NO<sub>x</sub> emissions in Eastern China, where the Chinese NO<sub>x</sub> emissions are the highest, at the relatively high horizontal resolution of 0.5°×0.5° for the period 2019 – 2021, that includes changes in emissions due to Chinese emissions reduction policies, lockdown measures, and LNY holidays. The relatively fine spatial resolution makes it possible to estimate the decrease/increase of NO<sub>x</sub> emissions down to the provincial level. We therefore study the impact of COVID-19 lockdown measures, as well as the effect of the LNY on the emissions of NO<sub>x</sub> in a total of 26 provinces over Eastern China. To estimate 160 these emissions, we conduct variational inversions at 0.5° and 1-day resolution with the Community Inversion Framework (CIF, Berchet et al., 2021), using the CHIMERE CTM (Mailler et al., 2017; Menut et al., 2013), including a chemistry module taking into account the complex NO<sub>x</sub> chemistry in gas-phase, its non-linearities, and its adjoint (Fortems-Cheiney et al., 2021). This framework and the regional inversion configuration are built upon the work done by Fortems-Cheiney et al. (2021, 2024), 165 Savas et al. (2023) and Plauchu et al. (2024). **Section 2** presents the data sources and pre-processing, as well as the regional inverse modelling configuration. **Section 3** presents the results and discusses them, and the last section summarizes the conclusions.



## 2. Data and Methods

### 2.1. Configuration of the CHIMERE CTM for the simulation of NO<sub>2</sub> concentrations in Eastern China

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Study domain boundaries	101.75-132.25°E; 17.75-50.25°N
Horizontal resolution	0.5°×0.5°, 61 × 65 grid cells (longitude × latitude), i.e., 3965 grid cells per level
Vertical resolution	17 layers extending from the surface up to 200 hPa (around 12 km above the sea level)
Meteorological fields	European Centre for Medium-Range Weather Forecasts (ECMWF) operational meteorological forecast (Owens and Hewson, 2018)
Initial and boundary conditions (excluding NO <sub>x</sub> )	LMDZ-INCA (Szopa et al., 2009)
Anthropogenic emissions	A combination of Carbon-Monitor (Z. Liu et al., 2020), Community Emissions Data System (CEDS, Hoesly et al., 2019, 2018), MEIC data (Zheng et al., 2018b), and EDGAR-HTAP-v2.2 for VOCs (Janssens-Maenhout et al., 2015)
Biogenic soil emissions (NO+VOCs)	Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006)
Chemical Scheme	MELCHIOR-2 (Carter, 1990; Derognat et al., 2003)

**Table 1.** The configuration of CHIMERE for the simulation of NO<sub>2</sub> concentrations in Eastern China. See **Figure 1** for a map of the domain. MELCHIOR-2: Modèle Lagrangien de Chimie de l'Ozone à l'échelle Régionale, gas-phase chemical reaction scheme.

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CHIMERE CTM is designed for the study of regional atmospheric pollution events (e.g., Ciarelli et al., 2019; Menut et al., 2020), and is part of the operational ensemble of the Copernicus Atmosphere Monitoring Service (CAMS) regional services. The long series of publications on air quality modelling with this CTM, including comparisons to observations, support the reliability and the accuracy of the CHIMERE simulations over regions such as Eastern China. In particular, C. Gao et al. (2024) showed that CHIMERE provides reliable simulations of air quality and NO<sub>2</sub> concentrations over this region, effectively capturing annual and seasonal spatiotemporal characteristics when evaluated against surface and satellite observations.

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The configuration set for the CHIMERE simulations in this study is summarized in **Table 1**. CHIMERE is driven here by meteorological fields at a fine resolution of 0.25°, from the European Centre for Medium-Range Weather Forecasts (ECMWF) operational meteorological forecast (Owens and Hewson, 2018). The MELCHIOR-2 scheme includes 120 oxidation reactions of 40 gaseous species.

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**Figure 1.** The study domain covers the Chinese provinces of mainland China, hereafter called “Eastern China”. The model grid cells are shown in dashed grey.

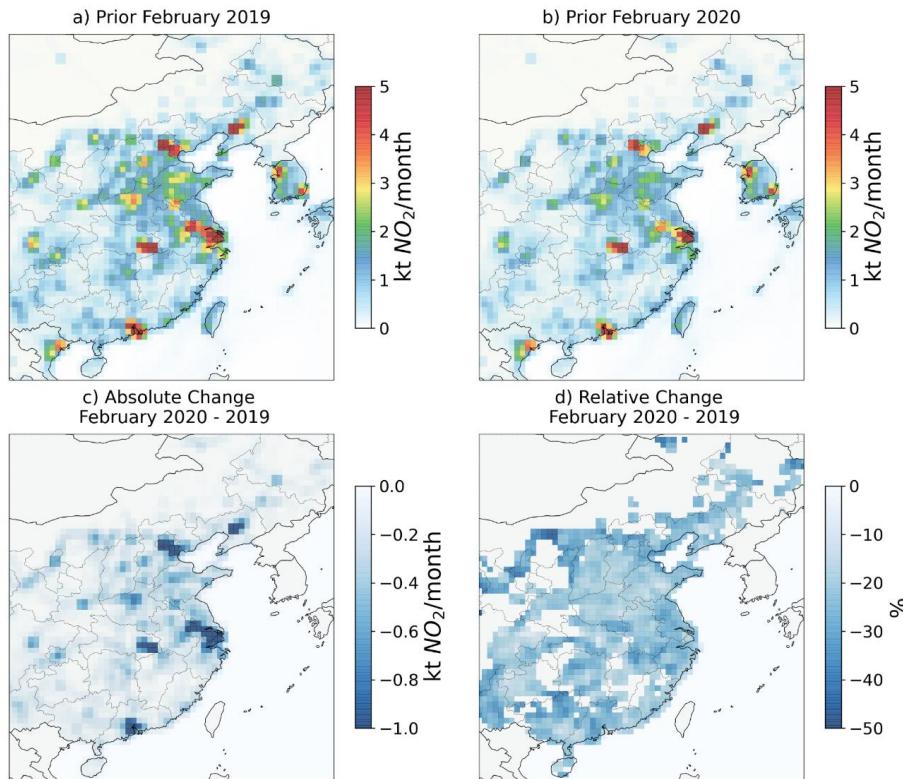
190 Considering the short lifetime of  $\text{NO}_2$ , we do not consider its influx from outside the domain: its  
boundary conditions are set to zero, as in Savas et al. (2023), Plauchu et al. (2024) and in Fortems-  
Cheiney et al. (2024). Nevertheless, the lateral and top boundaries for other species, such as  $\text{O}_3$ , nitric  
acid ( $\text{HNO}_3$ ), peroxyacetyl nitrate (PAN) and formaldehyde (HCHO), participating in the  $\text{NO}_x$   
chemistry are considered, as in Savas et al. (2023), in Plauchu et al. (2024) and in Fortems-Cheiney et  
al. (2024). In this study, Eastern China refers to the Chinese mainland included in the study domain  
(Figure 1).

## 2.2. Prior estimates of $\text{NO}_x$ emissions in Eastern China

200 The main goal of an inversion is to correct a-priori emission maps, that we refer to as “prior”  
emissions/estimates. In this study, the anthropogenic prior estimates are a combination of three  
inventories: the Community Emissions Data System (CEDS) (R. Hoesly et al., 2018, 2019), the Carbon  
Monitor (Z. Liu et al., 2020), and the Multi-resolution Emission Inventory model for Climate and air  
pollution research (MEIC) (Zheng et al., 2018b). The resulting prior estimates used for this study are  
therefore referred to as CEDS-CarbonMonitor-MEIC (CCMM). In the combination of these three  
inventories, CEDS provides gridded monthly emissions for the year 2019 at a  $0.5^\circ \times 0.5^\circ$  horizontal



205 resolution; these monthly estimates are downscaled to daily emissions based on daily variations from  
Carbon Monitor for the period 2019 - 2021; finally, the process of temporal disaggregation from daily  
to hourly emissions relies on typical diurnal emission profiles by sector derived from MEIC (Zheng et  
al., 2018b). CEDS employs a comprehensive approach that integrates various emissions inventories,  
factors, and activity data to derive emission estimates at national and sectoral scales (Hoesly et al.,  
210 2018, 2019). CEDS monthly emissions are produced from aggregated estimates, leveraging spatial  
proxy data from the EDGAR gridded emissions dataset and distributed for nine final gridded sectors  
(agriculture, energy, industrial, transportation, residential and commercial, solvents production and  
application, waste, international shipping, and aviation). The agricultural sector includes soil emissions  
due to agricultural practices, derived from the EDGAR inventory (Crippa et al., 2018). Among these  
215 sectors, we exclude aviation for this study. To obtain daily emissions, Carbon Monitor daily variations  
for each month, sector, and region are used. These variations are derived from CO<sub>2</sub> daily estimates. The  
impact of the 2020 lockdown is therefore considered in CCMM via the relative temporal variations  
extracted from the Carbon Monitor (illustrated in **Figure 2** for February, shown for the other months of  
the year in Figures S1, S2, and S3). NO<sub>x</sub> emissions are expressed in either teragram or kiloton in  
220 equivalent NO<sub>2</sub> (TgNO<sub>2</sub> or ktNO<sub>2</sub>) throughout the study.



225 **Figure 2.** Prior estimates of NO<sub>x</sub> total (anthropogenic + biogenic) emissions for February 2019 (a) and 2020 (b), and the  
absolute (c) and relative (d) change between these estimates. Values below 0.3 kt NO<sub>2</sub> in the prior estimate for February  
2019 are set to zero when calculating the relative difference (d) to reduce the noise in the figure. The total relative difference  
across the entire domain (shown in panel d) is -22%.



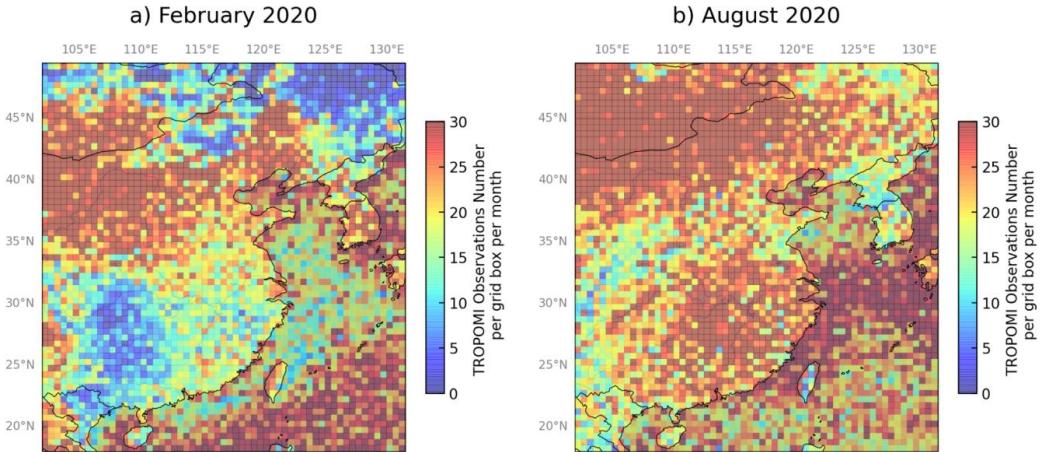
In addition to  $\text{NO}_x$  anthropogenic emissions,  $\text{NO}$  biogenic emissions from soil microbial activity are considered. These  $\text{NO}$  biogenic soil emissions are based on simulations from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006), with a  $\sim 1 \text{ km} \times 1 \text{ km}$  spatial resolution. These biogenic simulations do not provide soil emissions due to agricultural practices (these  
230 are included in the anthropogenic CCMM inventory). The lightning-generated  $\text{NO}_x$  are not taken into account in the prior estimation. In China, the lightning-produced  $\text{NO}_x$  ( $\text{LNO}_x$ ) emissions are estimated at 516 to 1054  $\text{ktNO}_2.\text{yr}^{-1}$  (Q. Li et al., 2023), and they contribute up to 7.5% to the total tropospheric  $\text{NO}_x$  emissions (Fengxia et al., 2016). Fire emissions are neglected, as their contribution to the  $\text{NO}_x$  total emissions in China is relatively small 230 (50–450)  $\text{ktNO}_2.\text{yr}^{-1}$  (Yin et al., 2019), which amount  
235 to approximately 1.15% of the total national  $\text{NO}_x$  emissions. All the emission data are spatially aggregated at the horizontal resolution of  $0.5^\circ \times 0.5^\circ$  of the CHIMERE grid.

### 2.3. $\text{NO}_2$ TVCDs from TROPOMI

In this study we use  $\text{NO}_2$  TVCDs from the TROPOMI instrument (Veefkind et al., 2012). TROPOMI  
240 is a nadir-viewing hyperspectral spectrometer that covers a large spectral range (270 – 2385 nm) from the ultraviolet (UV) to the shortwave infrared (SWIR) (Lambert et al., 2024). S-5P is following a sun-synchronous polar orbit, and has a daily overpass at 13:30 solar local time. TROPOMI has a swath of  $\sim 2600 \text{ km}$ , with a spatial resolution of  $3.5 \times 5.5 \text{ km}^2$  (since 6 August 2019, and  $3.5 \times 7 \text{ km}^2$  prior to that time), and it covers latitudes between  $7^\circ$  and  $7^\circ$ .

We use reprocessed (RPRO) version 02.04.00 from TROPOMI  $\text{NO}_2$  products, from January 2019 to  
245 December 2021. The main update in this version, compared to previous versions, is that it uses a spectral surface reflectivity climatology derived from TROPOMI (Tilstra et al., 2024) replacing the older OMI and GOME-2 (the Global Ozone Monitoring Experiment-2) datasets used in versions 1.0.2 to 2.3.1. This replacement, which also accounts for the directionality of the Lambertian Equivalent Reflectivity (LER), leads to significant changes in the cloud retrievals (van Geffen et al., 2022).

250 TROPOMI RPRO-v02.04.00 data and ground-based MAX-DOAS data, from 31 stations globally, continue to show discrepancies that vary with the level of pollution near the station (Lambert et al., 2024). However, when MAX-DOAS profile data are vertically smoothed using the S-5P averaging kernels (AKs), the bias is positive (+10%) over non-polluted areas and negative (-32%) over highly polluted areas. Furthermore, comparisons of  $\text{NO}_2$  total columns with the Pandora direct-sun spectrometer (Herman et al., 2009), which provides robust column measurements due to its trivial light path, show a median bias of -8% overall, with a positive bias of 4% at clean, high-altitude sites and a negative bias of -15% in polluted areas (Lambert et al., 2025). These biases are reduced when AKs are used, as in this study. We select data with a quality assurance (QA) value of 0.75, following the criteria of Lambert et al. (2024, 2025).



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**Figure 3.** Number of TROPOMI observations (with a quality flag  $qa > 0.75$ ) of tropospheric  $\text{NO}_2$  (cloud-free) per  $0.5^\circ$  resolution grid-cell, during a winter month (a) and a summer month(b) in 2020.

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In winter, some areas have a relatively lower observation number, as compared to the rest of the domain and to summer months (Figure 3). This is mainly due to the larger cloud cover, but other factors have an impact, such as the solar zenith angle, aerosol optical thickness and surface albedo (M. Gao et al., 2023). Overall, TROPOMI provides reliable and consistent coverage of the studied domain, during both summer and winter, which makes it possible to estimate emissions down to the provincial level.

#### 2.4. Consistent comparison between simulated and observed $\text{NO}_2$ TVCDs

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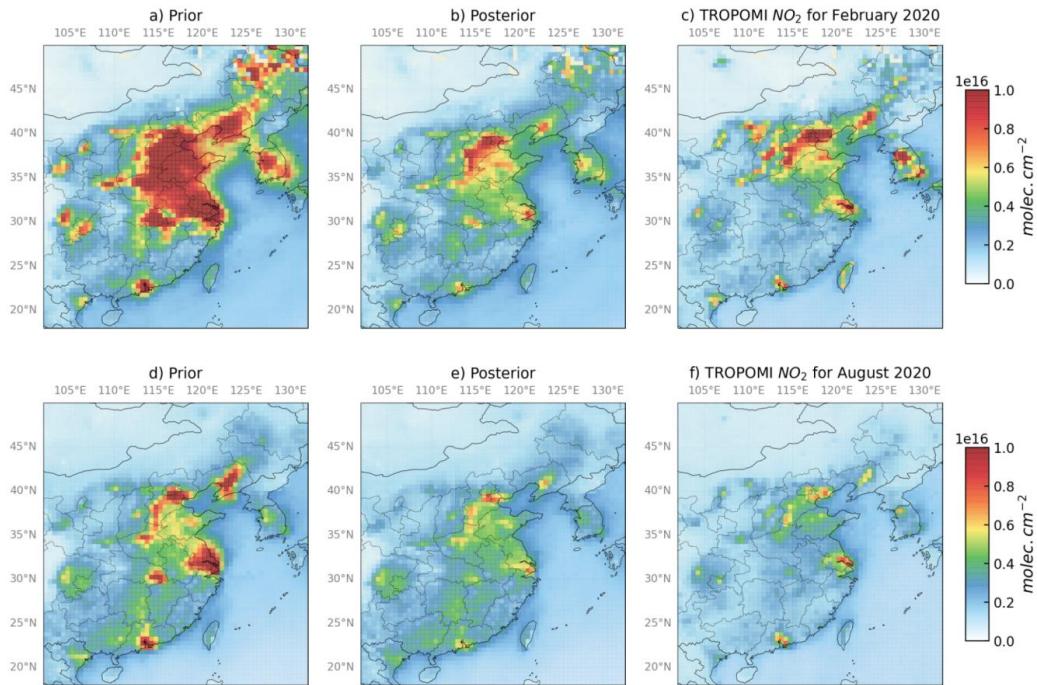
In order to compare  $\text{NO}_2$  TVCDs simulated by CHIMERE with those provided by TROPOMI observations, we need to tackle several differences between the satellite instrument and the CTM 3D simulated  $\text{NO}_2$  concentration fields. For example, these differences include: 1) spatial and temporal resolutions, and 2) variations of the instrument retrieval sensitivity to the concentrations along the vertical columns characterized by the retrieval AKs. A “super-observation” approach (Eskes et al., 2003; Miyazaki et al., 2012) is employed to aggregate the observations and derive errors (uncertainties from the retrieval process, including the propagation of the instrumental noise) and vertical sensitivity at the model resolution (Fortems-Cheiney et al., 2021; Plauchu et al., 2024; Savas et al., 2023). The temporal resolution is addressed, by matching the overpass of the S-5P satellite to the time stamps in the simulated TVCDs from CHIMERE. At a given time, in each model grid-cell covered by TROPOMI observations, the super-observation is defined as the observation (TVCD, errors and AKs) corresponding to the TVCD value closest to the mean of all the observations within this  $0.5^\circ \times 0.5^\circ$  grid cell, and within the corresponding CHIMERE physical time step of approximately 5 to 10 minutes, following the method described by Plauchu et al. (2024). The super-observation errors were assigned as in Plauchu et al. (2024), so as to account for spatially correlated systematic uncertainties in TROPOMI  $\text{NO}_2$  retrievals.

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The simulated NO<sub>2</sub> TVCDs (Figure 4a, d for two months) broadly capture the spatial and temporal variabilities of those provided by TROPOMI (Figure 4c, f).



**Figure 4.** Monthly means of the NO<sub>2</sub> TVCDs corresponding to the TROPOMI super-observations at 0.5° resolution during February (upper row) and August (bottom row) 2020. The CHIMERE prior TVCDs are shown in the first column (a, d), the posterior estimates using the correction of the prior are shown in the second column (b, e), and the TVCDs from TROPOMI are shown in the last column (c, f).

## 2.5. Variational inversion of NO<sub>x</sub> anthropogenic and biogenic soil emissions

290 The NO<sub>x</sub> emission estimates from the inversion (called “posterior” estimate of the emissions) consist in a correction of the prior estimates of NO<sub>x</sub> emissions (described in Section 2.2), which reduce the differences between the simulated NO<sub>2</sub> and the TROPOMI NO<sub>2</sub> super-observations. We use a Bayesian variational inversion framework, similar to the one used in Fortems-Cheiney et al. (2021, 2024), Savas et al. (2023) and Plauchu et al. (2024). To cover the period from 2019 to 2021, we conduct 295 a series of monthly inversions (covering 1-month windows) that are later combined; each inversion is independent of the others. In each iteration, the posterior emissions, are derived from the minimization of the cost function  $J(x)$ , stated below as **Equation 1**:

$$J(x) = \frac{1}{2} (x - x^b)^T B^{-1} (x - x^b) + (H(x) - y)^T R^{-1} (H(x) - y) \quad \text{Equation 1}$$



300 The control vector  $\mathbf{x}$ , contains variables that are corrected by the inversion, which, here, all underlay the estimates of the surface emissions of NO and NO<sub>2</sub> in input of CHIMERE, while  $\mathbf{x}^b$  is the prior estimates of the control vector.  $\mathbf{y}$  combines the TROPOMI super-observations.  $\mathbf{H}$ , the observation operator, links the variables in the control vector (log-space) to the observation space defined by the TROPOMI super-observations. It includes: the exponential operator and scaling of the prior emission maps that convert the maps of the logarithm of the coefficient for the emission) into emission maps at 305 the spatial and temporal resolutions of CHIMERE; the atmospheric chemistry and transport model CHIMERE itself; and the extraction of the TVCDs from CHIMERE when TROPOMI super-observations are available (spatially and temporally).

310  $\mathbf{B}$  is the prior error covariance matrix: it fully characterizes the uncertainty in  $\mathbf{x}^b$ , called prior uncertainty under the assumption that it follows an unbiased and normal distribution  $N(\mathbf{0}, \mathbf{B})$ ;  $\mathbf{R}$  is the observation error covariance matrix: it corresponds to the sum of uncertainties from the observation operator and from the super-observations; assuming that these errors have an unbiased and normal distribution,  $N(\mathbf{0}, \mathbf{R})$ , they are fully characterized by  $\mathbf{R}$ .

315 Here,  $\mathbf{x}$  is built so that the uncertainties in the prior estimates of the emissions are implicitly modelled assuming that they have a log-normal distribution, in order to reflect the uncertainty in the order of magnitude of these emissions and forcing the analyzed emissions to be positive (Plauchu et al., 2024).  $\mathbf{x}$  is thus composed of logarithms of scaling coefficients to be applied to the prior estimate of the emissions.

320  $\mathbf{x}$  is split according to the anthropogenic and biogenic components of the NO and NO<sub>2</sub> emissions in each of the model grid cells at 0.5° resolution for each day. The information provided by the TROPOMI observations, together with the spatial and temporal resolutions of CHIMERE, do not support an accurate separation of the total NO<sub>x</sub> emissions between NO and NO<sub>2</sub> emissions. However, the control vector  $\mathbf{x}$  maintains this separation for technical simplicity, as this approach does not adversely impact the estimation of the total NO<sub>x</sub> emissions. There is no further sectorization of the control of anthropogenic emissions. More specifically,  $\mathbf{x}$  contains:

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- the logarithms of the scaling coefficients for NO anthropogenic emissions, at a 1-day temporal resolution, and a 0.5°×0.5° (longitude, latitude) horizontal resolution; and over the 17 vertical layers of CHIMERE i.e. for each of the corresponding 61×65×17 grid cells,
- the logarithms of the scaling coefficients for NO<sub>2</sub> anthropogenic emissions, at the same vertical, temporal, horizontal resolutions as for NO,
- the logarithms of the scaling coefficients for NO biogenic soil emissions at a 1-day temporal resolution, and a 0.5°×0.5° (longitude, latitude) horizontal resolution at the surface (1 vertical layer), i.e. for each of the corresponding 61×65×1 grid cells.

330 Accordingly,  $\mathbf{B}$ , is a 3-block diagonal matrix: the first block corresponds to the logarithms of the NO anthropogenic emissions, in which each diagonal element is set at (0.35)<sup>2</sup>, this variance value in the log-space corresponds to a factor ranging between 70 to 140% in the emission space, at the 1-day and model's grid scale. Similarly, the second block corresponds to the logarithms of the NO<sub>2</sub> anthropogenic emissions, in which each diagonal element is set at (0.35)<sup>2</sup>, this variance value in the log-space corresponds to a factor ranging between 70 to 140% in the emission space, at the 1-day and model's grid scale.



340 The third block of  $\mathbf{B}$  is set for NO biogenic soil emissions, in which each diagonal element is set at  $(0.5)^2$ , corresponding to a factor ranging between 60 to 164% in the emission space at the 1-day and model's grid scale.

345 We account for spatial correlations in the uncertainties both for anthropogenic and biogenic parts. Spatial correlations are described by exponentially decaying functions with an e-folding length of 50 km over land and over the sea as in Fortems-Cheiney et al. (2024). However, we do not account for temporal correlations between different days.

350 The uncertainties on the observations  $y$  and on the observation operator  $\mathbf{H}$  are characterized by the so-called observation error covariance matrix  $\mathbf{R}$ , set up here as a diagonal matrix based on the assumption that these errors are not correlated in space or time when aggregated at the model  $0.5^\circ$  and 1 h resolution. The variance in the observation errors corresponding to individual observations in the diagonal of  $\mathbf{R}$  is the quadratic sum of the error we have assigned to the TROPOMI super-observations and of an estimate of the errors from the observation operator. We assume that the observation operator error is dominated by the chemistry-transport modelling errors and -in case when the model grid-cell is covered by few TROPOMI observations- by the errors associated with the discrepancies between the 355 spatial representativeness of the super-observations and of the model corresponding column. It is set at 20 % of the retrieval value for each super-observation, as in Fortems-Cheiney et al. (2021, 2024).

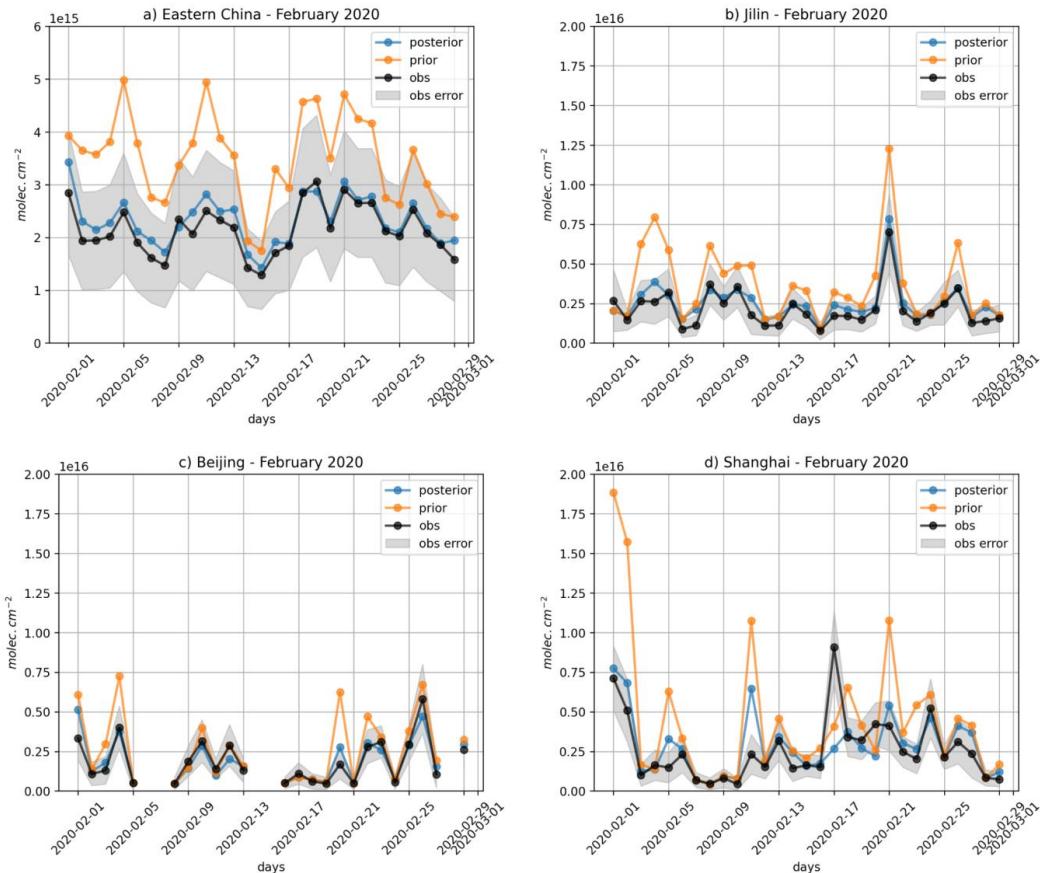
360 The inversion system searches for the minimum of the cost function  $\mathbf{J}$  using the iterative M1QN3 limited-memory quasi-Newton minimization algorithm (Gilbert & Lemaréchal, 1989). At each iteration, the computation of the gradient of  $\mathbf{J}$  relies on the adjoint of CHIMERE, which is the adjoint of the observation operator. We impose a reduction of the norm of the gradient of  $\mathbf{J}$  by 95% as a constraint for the interruption of the minimization process.

### 3. Results

#### 3.1. Improving the fit between CHIMERE simulations and TROPOMI super-observations

365 Before analyzing the results in terms of  $\text{NO}_x$  emissions, we check the behavior of the inversion by comparing the performance of the prior and posterior simulations in reproducing the spatial and temporal variations of the observations. We analyses the months of February and August 2020 to illustrate the typical behavior of the inversions in winter and summer, but the analysis is performed on all the months for the years 2019, 2020 and 2021 (Figures S4, S5, and S6).

370 TROPOMI super-observations and the corresponding CHIMERE  $\text{NO}_2$  TVCDs present similar spatial patterns, with hotspots over Shanghai, Beijing, Wuhan, and Hong Kong, as well as in the north-eastern province, Liaoning (Figure 4). However, the prior simulation overestimates the  $\text{NO}_2$  TVCDs in Eastern China compared to the observations, with a mean bias of about +30% and +12%, for February and August 2020 respectively.



**Figure 5.** Daily mean posterior (blue) and prior (orange) estimations of NO<sub>2</sub> TVCDs as compared to that of the TROPOMI super-observations (obs, black) during February 2020, with the 1-sigma error associated to the super-observations as a grey shaded area (obs error). The top row features averages for Eastern China (a) and the province of Jilin (b), and the bottom row shows averages for the provinces of Beijing (c) and Shanghai (d). The NO<sub>2</sub> TVCDs are expressed in molecules.cm<sup>-2</sup>. Note that the y axis limits are different for the provinces as compared to Eastern China. The provinces are shown in Figure 1 on the map of the domain.

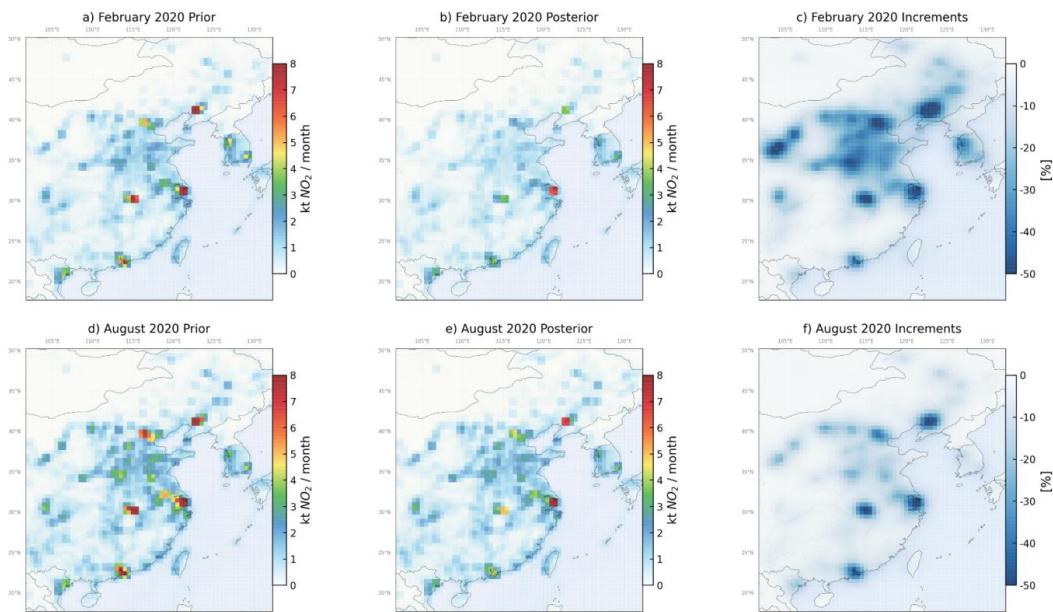
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The inversion brings the simulated NO<sub>2</sub> TVCDs closer to the TROPOMI super-observations (Figure 4; see also the time series of daily mean TVCDs in Figure 5). The improvement of the fit between the simulated and the observed NO<sub>2</sub> differs among polluted and rural regions, for instance, it is the best over polluted regions such as Shanghai, Beijing, Wuhan, Hong Kong, and Liaoning (Figure 4).

380 Nevertheless, the posterior simulation still presents positive biases compared to the observations (Figure 4 and Figure 5). This is partly due to 1) the limitations in the spatial and temporal coverage of TROPOMI, particularly in winter (Figure 3), 2) the errors associated with TROPOMI super-observations and with the CTM (assigned in the *R* matrix, Section 2.5), and 3) the potential lack of sensitivity of NO<sub>2</sub> TVCDs to NO<sub>x</sub> emissions depending on the chemical regime. As the CHIMERE



385 prior simulation overestimates the NO<sub>2</sub> TVCDs, the inversion brings the CHIMERE NO<sub>2</sub> columns closer to the TROPOMI data by reducing NO<sub>x</sub> emissions (Figure 6, Section 3.2).



**Figure 6.** NO<sub>x</sub> emissions during the months of February (top, a to c) and August (bottom, d to f) 2020. The prior (a, d) and the posterior (b, e) estimates are expressed in kilotons equivalent NO<sub>2</sub> (ktNO<sub>2</sub>/month), and the relative increments (c, f) to the prior emissions from the inversion, in %.

### 3.2. NO<sub>x</sub> annual budgets in Eastern China from 2019 to 2021

This section focuses on comparing the total (biogenic + anthropogenic) posterior NO<sub>x</sub> emission estimates and the prior ones. NO<sub>x</sub> posterior total emissions over Eastern China were reduced by -18% ( $\pm 0.4\%$ ) compared to the prior inventory, for all years considered here (Figure 7). This result may be partly due to the artificial propagation of errors from the TROPOMI observations (Lambert et al., 2024; Section 2.3) and from the CTM. Nevertheless, the potential biases of the TROPOMI observations are considered in the characterization of our **R** matrix (Section 2.5). In addition, CHIMERE has been shown to be reliable for simulating air quality over Eastern China (C. Gao et al., 2024) and no large-scale modeling bias has been identified in the CHIMERE simulation of NO<sub>2</sub> concentrations. Our results therefore suggest that NO<sub>x</sub> emissions are overestimated by inventories.

The increase of NO<sub>x</sub> emissions between 2019 and 2021 -which is probably linked to the evolution of the total consumption of fossil fuels in China (including coal, oil, and natural gas)- is the same in the posterior (+4.4%) as in the prior estimates (+4%). This increase over the 3-year period includes a decrease of NO<sub>x</sub> emissions in 2020 due to COVID-19 lockdowns (Section 3.4) and a rebound in 2021. Our posterior estimates indeed show a rebound of NO<sub>x</sub> emissions in 2021 compared to 2019 (Figure 7)

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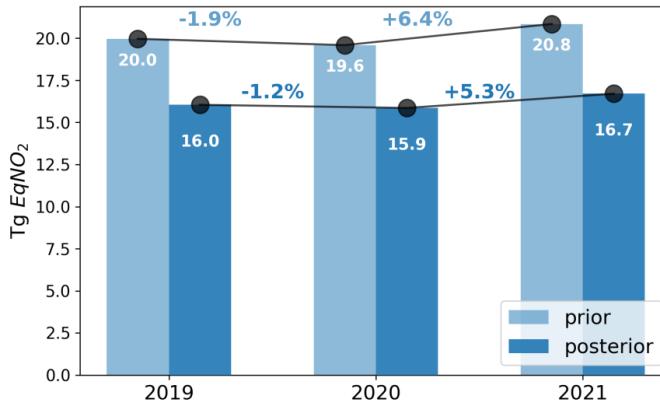
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405 while the annual mean TROPOMI NO<sub>2</sub> TVCDs decrease over the same period (Figure A 1). However, this rebound is not in agreement with H. Li et al. (2024): they estimated an increase of NO<sub>x</sub> emissions over China for the first quarter in 2021 compared to 2019 but a decrease of about -3.5% at the annual scale. Appendix A details the sensitivity tests we conducted to assess the effect of meteorological fluctuations from 2019 to 2021 on the observed variations in NO<sub>2</sub> TVCDs between these years. This analysis highlights the large impact of meteorology on the variations of the TVCDs between the two years. Both our inversion framework and that of H. Li et al. (2024) use a CTM to deconvolve this impact from that of the change in emissions when analyzing the variations of the TVCDs. However, CTMs bear relative uncertainties which limit this capability and which are propagated into the emission estimates. The uncertainties in diagnosing emission changes over this period are thus likely high, which may explain the divergent conclusions between the results of H. Li et al. (2024) and our inversion results for 2021.

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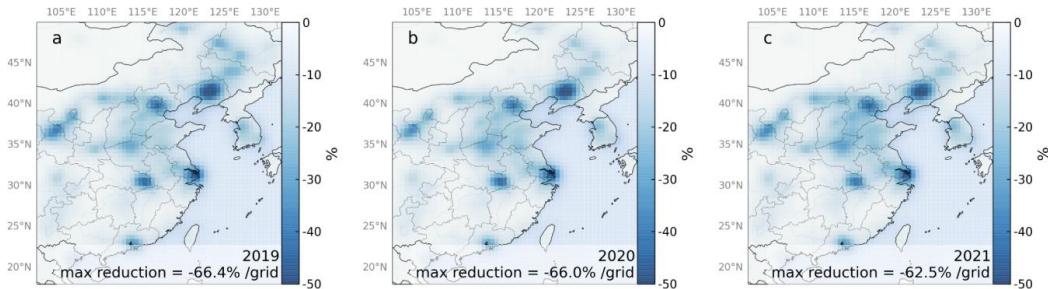


**Figure 7.** Annual total NO<sub>x</sub> emissions (biogenic + anthropogenic) for the period 2019 – 2021, expressed in Tg EqNO<sub>2</sub>/year for Eastern China in the domain of study. The prior (light blue) and posterior (dark blue) total estimates are shown in white on each bar. The year-on-year changes are shown in percentage above the black lines.

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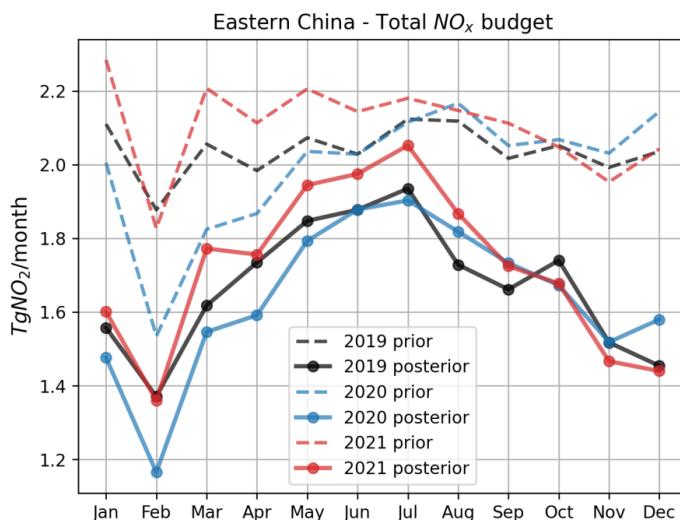
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At the scale of the province, the highest increments over the 3-year period are obtained in five main provinces: Liaoning (-49%), Beijing (-38%), Shanghai (-50%), Hubei (-28%), and Guangdong (-18%). There is no correlation between the reduction of prior estimates and the amount of NO<sub>x</sub> emitted per province (Figure S9). For instance, although Shanghai emits 471 ktNO<sub>2</sub>/year, while Liaoning emits 1398 ktNO<sub>2</sub>/year, we find that Shanghai and Liaoning show similar increments of about -50%; in Jiangsu the prior NO<sub>x</sub> emissions amount to 1304 ktNO<sub>2</sub>/year (similar to Liaoning), while the increment is -25% (compared to an increment of -49% in Liaoning). At the grid-cell resolution, however, the reduction of the prior estimates is highly correlated ( $r>0.97$ ) to the prior estimates. Similar results are found in the study of Plauchu et al. (2024) over France, with strong corrections mainly over dense urban areas. The corrections applied to the prior estimates in Eastern China, on a yearly scale are similar interannually, and they are -66% in 2019 and 2020, and -62% in 2021, per grid box and per year (Figure 8).



430 **Figure 8.** Maps of the yearly increments relative to the prior estimates (%) of annual NO<sub>x</sub> emissions at a 0.5° resolution, for the years 2019 to 2021 (a to c).

### 3.3. Seasonal cycle of NO<sub>x</sub> total emissions in Eastern China



**Figure 9.** Times series of the monthly prior (in dashed lines) and posterior (in solid lines) total NO<sub>x</sub> budget over Eastern China for the years 2019 (black), 2020 (blue) and 2021 (red), expressed in Tg equivalent NO<sub>2</sub> per month.

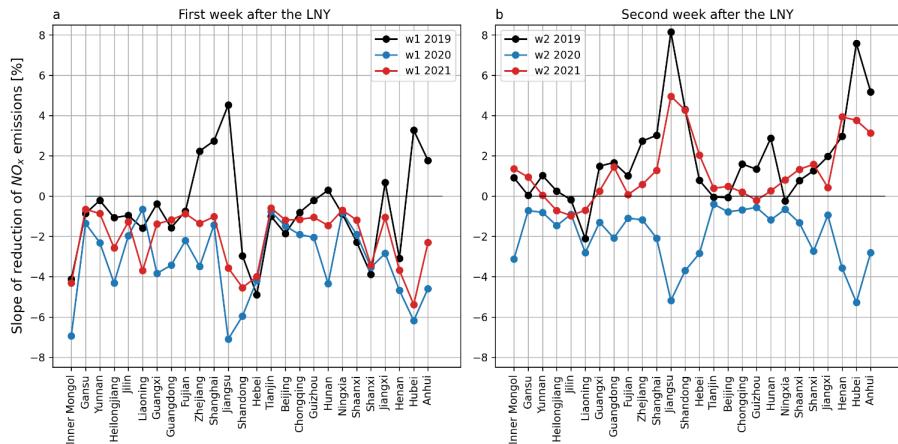
435 In contrast with the prior estimate of the emissions (Section 2.2), the posterior estimates show an annual maximum systematically occurring in July (Figure 9). The peak in summer agrees with the Daily Emissions Constraint by Satellite Observations (DECSO) emissions estimated from inversions assimilating OMI satellite observations (Ding et al., 2015). It suggests that the biogenic emissions may be underestimated in our prior estimates of the NO<sub>x</sub> emissions over Eastern China, hence the absence of  
440 the peak in the prior estimates during summer; the peak in August 2020 appearing mostly because of the reduction in emissions in winter. This suggestion would be in line with the study of Visser et al. (2019), showing a strong underestimation of the NO<sub>x</sub> biogenic emissions from MEGAN in summer over Europe.



Such peaks in summer could also be partly due to anthropogenic emissions, through electricity consumption and the resulting power load, highly correlated with rising temperatures in summer, as shown by Zhang et al. (2009).

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In agreement with the prior estimates, the posterior emissions show a minimum in February each year which can be linked to the LNY.



**Figure 10.** Slope of change in the posterior estimates of  $\text{NO}_x$  emissions during the first (a) and second (b) weeks following the LNY dates, in 2019, 2020, and 2021 in the Chinese provinces. These slopes of change are calculated for each week separately by computing the slope of a line that connects the first and the last days of the week in question. Week1 of LNY starts on the first day of the New Year, (assigned day1), and week2 starts on day 8. Note that the LNY dates for each year are different; and they fall on 5 February 2019, 25 January 2020, and 12 February 2021.

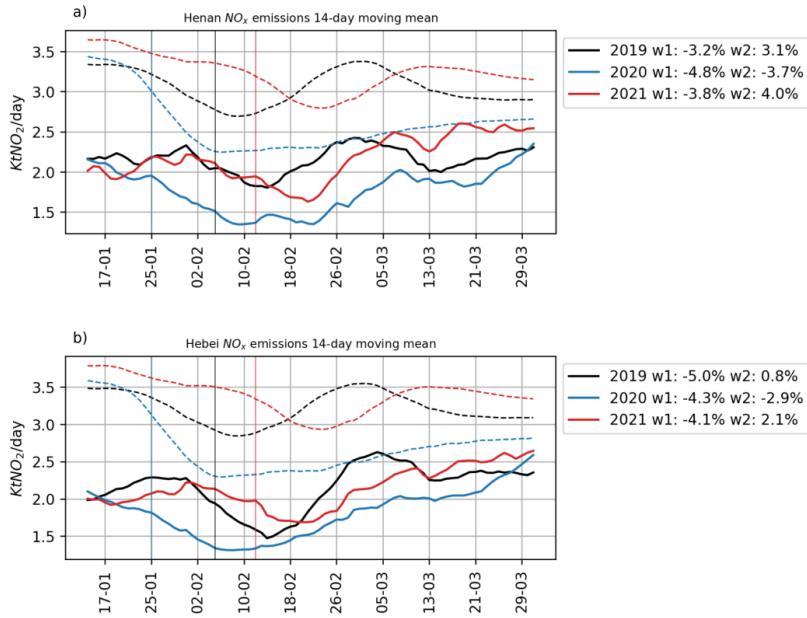
450 Our posterior estimates show a consistent relative reduction in  $\text{NO}_x$  emissions during the first week after LNY in 2019, 2020 and 2021 for 19 out of 26 provinces, by  $-7\%$  to  $-0.5\%$  (Figure 10). In all provinces, the emissions rebound in the second week during 2019 and 2021, but not in 2020, with reductions between  $-5.5\%$  and  $-0.1\%$  (Figure 10). We also looked at the 14-day moving means of  $\text{NO}_x$  emissions in all provinces, but we show only two in Figure 11, Henan and Hebei, as both are some of the most densely populated provinces in China (7+ million inhabitants). During the first week following the LNY date, we observe reductions of  $\text{NO}_x$  emissions, that are around  $-3.1\%$ ,  $-4.7\%$ , and  $-3.9\%$  for 2019, 2020 and 2021 respectively in Henan (Figure 11a). These reductions are explained by the reduced industrial activities during the LNY holidays. After this decline in emissions, the activity goes back to normal, which is reflected by the rebound in  $\text{NO}_x$  emissions during the second week following the LNY date in 2019 and 2021 (Figure 11). However, in 2020, the reduction is extended to the second week reaching  $-3.6\%$  in Henan and  $-2.8\%$  in Hebei, which emphasises the influence of pandemic-related disruptions on  $\text{NO}_x$  emissions (see Section 3.4). Miyazaki et al. (2020) showed that nearly 80% of the emission reductions, during the period 23 Jan – 29 Feb 2020, are due to the lockdown measures in each province. The outdoor restrictions took effect 7 days after the LNY in Henan, falling on 3

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February 2020, and 11 days after LNY in Hebei (7 February 2020).



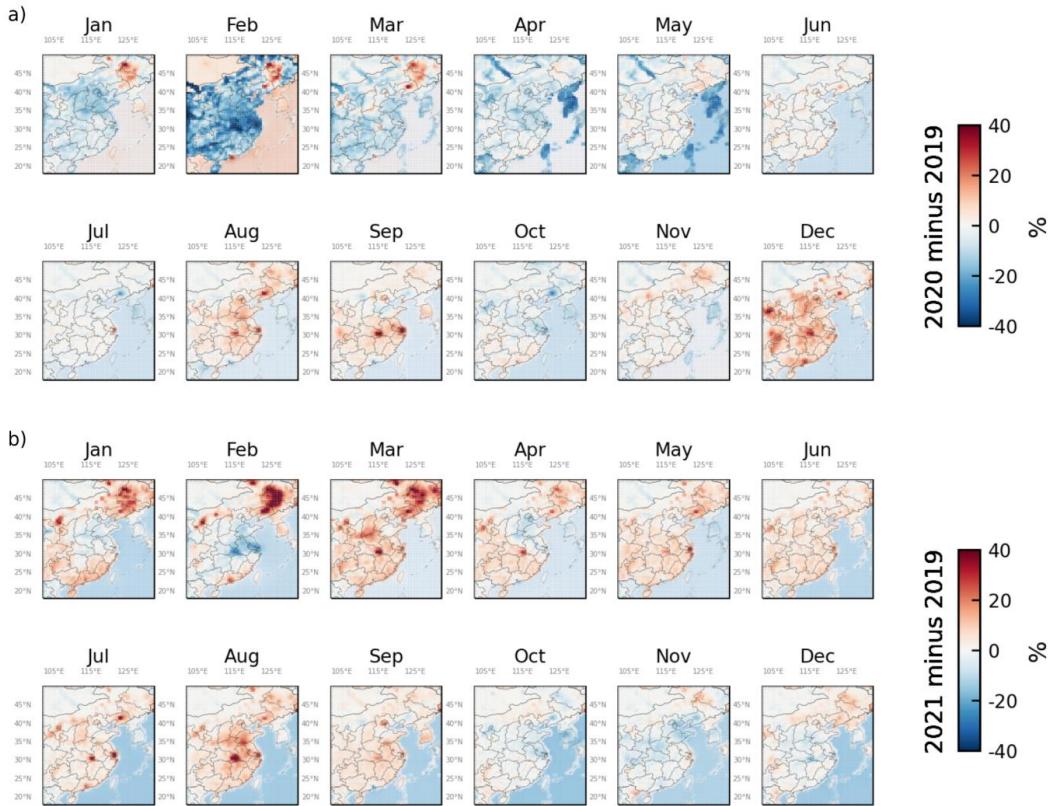
**Figure 11.** NO<sub>x</sub> emissions during the early period of the year, during 2019 (black), 2020 (blue) and 2021 (red). The dotted lines are the prior estimates, and the solid lines are the posterior (corrected) estimates presented as 14-day moving means, for the province of Henan (a) and Hebei (b). In the legend, we show the slope of change during the first (w1) and the second (w2) weeks following the new year date. The Lunar New Year dates are shown as dashed lines in black (for 2019), blue (2020) and red (2021). The slopes of change are calculated on a section of the line (for the posterior estimates). For instance, to get the slopes of week 1, we take the first and last days of the week, we then calculate the slope on a line that connects these two days; it is finally presented as a percentage of change from day 1 to day 7 during the week. The same calculation is applied for week 2, when we take the eighth day after the New Year date, and we count 7 days (day 8 to 14).

### 3.4. Impact of the COVID-19 lockdowns in Eastern China

Following a usual diagnostic in the literature to assess the change in air pollutant concentrations and emissions due to COVID-19 policies, we characterize the impact of the COVID-19 lockdown in Eastern China in terms of changes in anthropogenic emission estimates from 2019 to 2020 (Fig 12a). While Eastern China total emissions decrease by about -1.8% in 2020 compared to 2019 in CCMM, the posterior emissions show a smaller decrease of about -1.15% (Figure 7), less than the one estimated by H. Li et al. (2024) at -2.7% in 2020 compared to 2019.

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**Figure 12.** Relative difference (in %) in the posterior estimates of the  $0.5^{\circ}$  resolution  $\text{NO}_x$  monthly emission maps during 2020 (a) and 2021 (b) as compared to 2019, over the domain of study. Each plot is the result of the following equation:  $\frac{\text{emissions}_{\text{year}} - \text{emissions}_{2019}}{\text{emissions}_{2019}} \times 100$ , with  $\text{emissions}_{\text{year}}$  being the posterior monthly emissions of the year in question.

February marks the month that witnessed the highest decrease in  $\text{NO}_x$  emissions:  $-40\%$  in 2020 compared to 2019. This result agrees with other studies. Zheng et al. (2021) for instance found a decrease of about  $-36\%$  of the  $\text{NO}_x$  emissions, leading to a good agreement between  $\text{NO}_2$  surface observations and simulations. Miyazaki et al. (2020) found that Chinese  $\text{NO}_x$  emissions were reduced by  $-36\%$  from early January to mid-February in 2020 compared to 2019. Finally, Kong et al. (2023) and Stavrakou et al. (2021) also found a decrease of about  $-40\%$  of the  $\text{NO}_x$  emissions in February 2020 compared to 2019.

Our configuration allows us to access the spatial distribution of emissions variations. At the provincial resolution, the changes in emissions compared to 2019 are between  $-3.3\%$  and  $+3.3\%$  in 2020, and between  $-1.8\%$  and  $+11\%$  in 2021 (Figure 14). In 2021, all provinces, except Tianjin, show an increase in emissions, most of them between  $+0.05\%$  and  $+5.5\%$ ; the highest increases are mostly in industrial regions, e.g. Liaoning ( $+11\%$ ), Heilongjiang ( $+10\%$ ), and Jilin ( $+9.7\%$ ).

In February 2020, the reduction of emissions occurred in most of Eastern China (Figure 12a). The highest decrease of the  $\text{NO}_x$  emissions, by more than  $-30\%$  ( $\pm 7\%$ ), are found in the provinces of

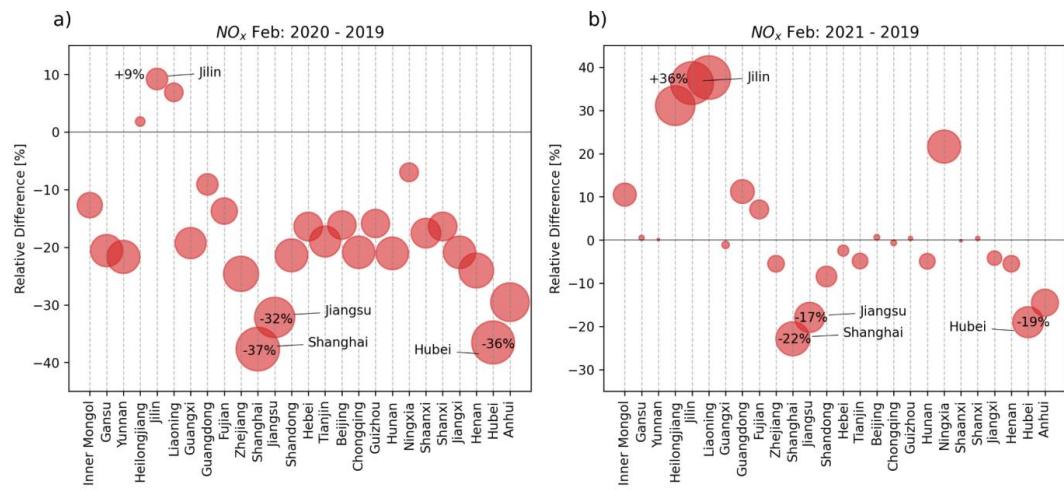
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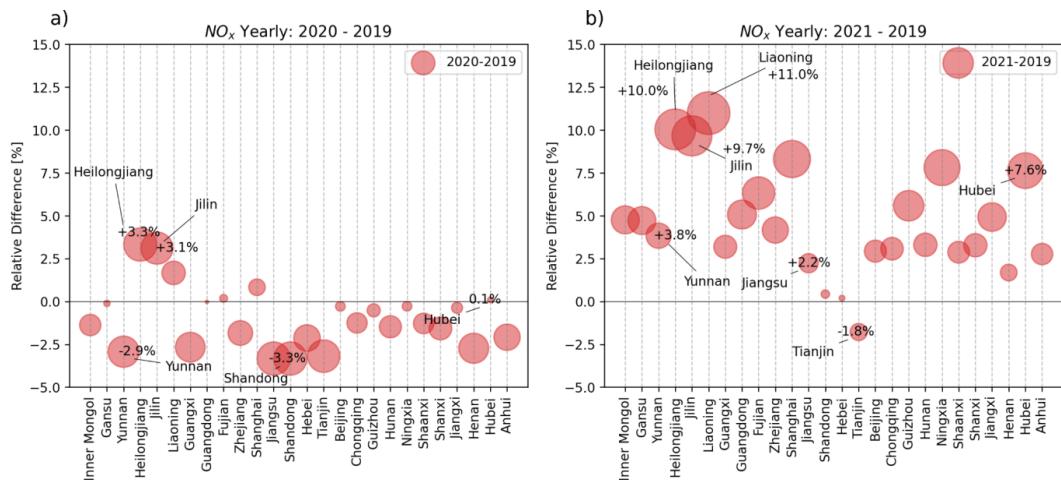
490 Shanghai, Hubei and Jiangsu (**Figure 13**). On the contrary, emissions increase in the three provinces of Heilongjiang, Jilin and Liaoning located in Northeast China (**Figure 13**). This result agrees with the study of R. Li et al. (2024) also finding an increase in NO<sub>x</sub> emissions in these three provinces, due to an increase in industrial production.

495 During March-April-May of 2020, emissions decreased by -20% along the China-Mongolia-Russia Economic Corridor (**Figure 12a**), one of the main economic belts connecting China through Inner Mongolia, then Mongolia and Russia (World Bank, 2019). This reduction in NO<sub>x</sub> emissions may be a result of the decrease in the Chinese exports in April and May 2020 by -15% (GACC, 2020), due to the lock-down measures applied by the Chinese government and to the decrease in Russian exports to China, from ~\$60 billion in 2019 to ~\$50 billion in 2020 (GZERO, 2022; OEC, 2022).



**Figure 13.** Relative difference in the posterior NO<sub>x</sub> emissions during February 2020 (a) and 2021 (b) as compared to 2019, in %, for the provinces illustrated in Figure 1. Note that the scale of the y axes is not the same.

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**Figure 14.** Same as **Figure 13**, but at the annual scale.

#### 4. Conclusions

Monitoring NO<sub>x</sub> emissions makes it possible to assess existing air quality policies. In addition to BU approaches (inventories and process-based models), the assimilation of atmospheric data into inverse 505 modelling systems can provide a means for monitoring emissions at a finer time resolution. This study evaluates the potential of satellite data from TROPOMI to provide estimates of NO<sub>x</sub> emissions across Eastern China at the monthly and provincial resolutions, using regional-scale atmospheric inversions. For this, TROPOMI NO<sub>2</sub> TVCDs are assimilated in the variational inversion model of the Community Inversion Framework (CIF), coupled to the CHIMERE regional chemistry transport model.

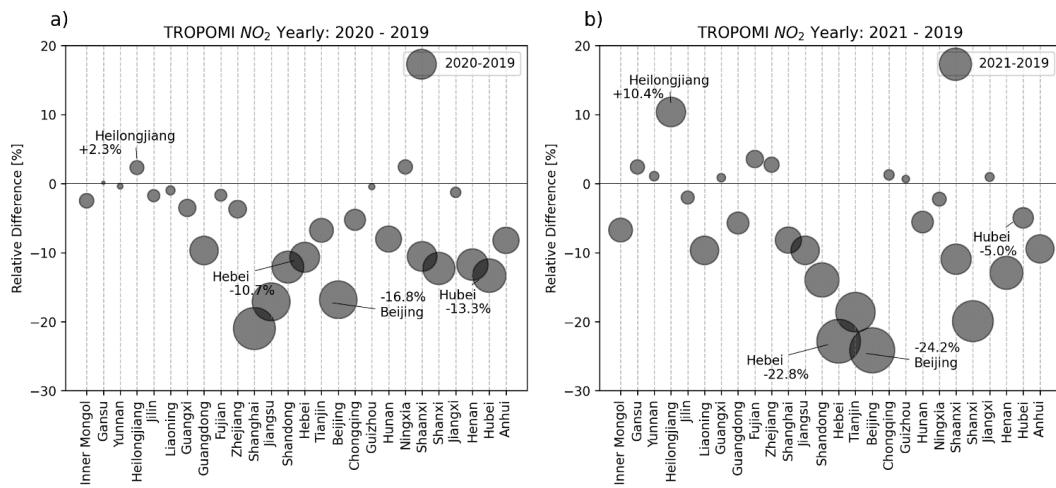
510 We focused on the changes in Chinese NO<sub>x</sub> emissions from 2019 to 2021, with an analysis of the impacts of the Chinese Lunar New Year (LNY) in the three years and of the COVID-19 outbreak in 2020. Each year in 2019, 2020 and 2021, the LNY holidays led to a reduction of emissions during the first week following the New Year day and to an increase of emissions during the second week, except in 2020. Driven by the impact of the LNY combined with the COVID-19 outbreak and the associated 515 lock-down measures, our estimates show a reduction in NO<sub>x</sub> emissions of  $-40\%$  in February 2020 relative to 2019, in Eastern China, in agreement with previous studies. In addition to this, the relatively fine spatial resolution of our framework shows a reduction in emissions along the China-Mongolia-Russia Economic Corridor, of  $-20\%$  during the spring of 2020, correlated with the measures applied regarding the import-export activities in China. During spring 2020, NO<sub>x</sub> emissions decreased in all 520 studied provinces, except three industrial provinces in the North Eastern part of China. At the yearly scale, however, while emissions in 2020 decreased by  $-1\%$  compared to 2019, we found a rebound of  $+4\%$  in 2021 compared to 2019.

This study highlights the changes in emissions during the LNY and the COVID-19 periods, and adds a refinement down to the level of provinces. Our regional configuration and inversion model therefore



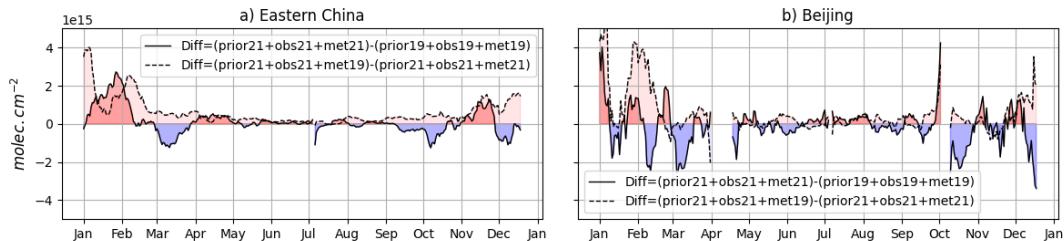
525 allow for the estimation of emissions at such a scale, which is crucial for the evaluation of policies at sub-national scales.

#### Appendix A: Impact of the meteorology on the simulation of NO<sub>2</sub> TVCDs in CHIMERE



**Figure A 1.** Relative differences in the NO<sub>2</sub> TROPOMI TVCDs during February 2020 (a) and 2021 (b) as compared to 2019, in %, for the provinces illustrated in **Figure 1**.

530 Our posterior estimates show a rebound of NO<sub>x</sub> emissions in 2021 compared to 2019 (**Figure 7**), with an increase over almost all the provinces (except one) at the annual scale (**Figure 14b**). However, the annual mean TROPOMI NO<sub>2</sub> TVCDs decrease between 2019 and 2021 over most provinces, and, overall, by about  $-8.3\%$  over Eastern China (**Figure A 1**). These variations in the NO<sub>2</sub> TVCDs are driven by changes in NO<sub>x</sub> emissions between 2021 and 2019 but also by fluctuations of the meteorological conditions from 2019 to 2021. In order to understand the relative impact of the meteorology on these variations, we performed a sensitivity test by simulating the NO<sub>2</sub> TVCDs in 2021, using the CCMM inventory for the year 2021 but meteorological fields from the year 2019. This led to increased simulated TVCDs in 2021 by +17% in Eastern China compared to the reference simulation for this year. This change in the simulation of NO<sub>2</sub> TVCDs in 2021 due to using the meteorological forcing from 2019 is shown to be positive throughout the year, but enhanced in January, February, April, and later in November, for Eastern China (**Figure A 2, a**), and Beijing (**Figure A 2, b**), as an illustration of a polluted province.



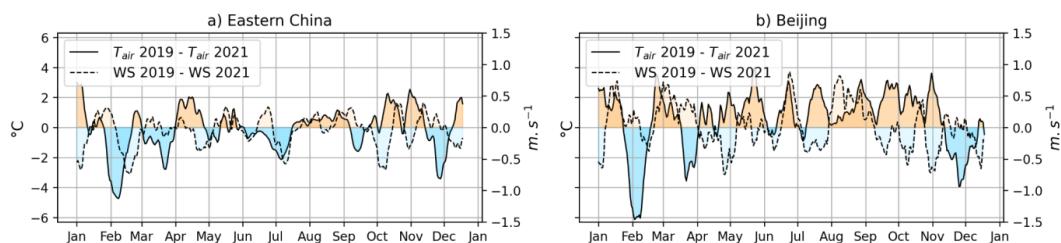
545 **Figure A 2.** Time series of the differences between daily 14-day running averages of the NO<sub>2</sub> TVCDs over Eastern China (a) and Beijing (b) from different simulations. The solid lines show the difference between the averages from the reference prior simulations of the TVCDs estimates for 2019 and 2021 (one simulating the TROPOMI observations in 2021 with the meteorological forcing for 2021: prior21+obs21+met21, and the other simulating the data in 2019 with the meteorological forcing for 2019: prior19+obs19+met19). The dotted lines represent the sensitivity to meteorology: they show the difference between the averages from the two simulations of the TROPOMI observations in 2021: the reference prior simulation: prior21+obs21+met21, and the sensitivity test with the meteorological forcing for 2019: prior21+obs21+met19.

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555 While various meteorological factors play a role in the simulation of NO<sub>2</sub> TVCDs, we focus our diagnostics on two parameters: air temperature at an altitude of 2m, and the wind speed at 10m. In Eastern China and at the provincial scale, the two main peaks of sensitivity of the simulated NO<sub>2</sub> TVCDs to the meteorology in January and February, coincide with the positive difference of temperature in January between 2019 and 2021 (**Figure A 2**), and the negative difference of wind speed in February between 2019 and 2021 at both scales (**Figure A 3a, b**). This confirms that the impact of changes in the meteorology on the atmospheric chemistry and transport can explain the decrease of NO<sub>2</sub> concentrations between 2019 and 2021 despite the increase of the NO<sub>x</sub> emissions diagnosed from our inversions.

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565 The effect of meteorological variability is thus substantial, and the relative uncertainties in the simulation of such an effect that is inherent in the CTM are propagated into the emission estimates, and thus into the estimate of the variations of these emissions from 2019 to 2021, within the atmospheric inversion frameworks. This likely explains the differences between our results in terms of interannual variability from 2019 to 2021 and those of H. Li et al. (2024), who also used a CTM in their inversion framework, but who reported a decrease in emissions in 2021 relative to 2019.



580 **Figure A 3.** Time series of the absolute differences of air temperature (at 2m) and wind speed averages, for Eastern China (a) and Beijing (b). All curves represent 14-day running means.



### Author contributions

570 RA, AFC, and GB conceptualized the study and carried out the results analysis. RA carried out the inversions, and the output data analysis, and input data pre-processing. IP, AB, EP developed the CIF inversion system. RP and AFC contributed to the preprocessing for fluxes and satellite observations. BZ provided the CEDS-CarbonMonitor-MEIC (CCMM) inventory used as prior emissions in this study. All the co-authors read the manuscript.

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### Data availability

TROPOMI-RPRO-v02.04.00 data are freely available through the website [https://tropomi.gesdisc.eosdis.nasa.gov/data/S5P\\_TROPOMI\\_Level2/S5P\\_L2\\_NO2\\_HiR.2/](https://tropomi.gesdisc.eosdis.nasa.gov/data/S5P_TROPOMI_Level2/S5P_L2_NO2_HiR.2/).

580 The NO<sub>x</sub> emissions of this study are available to download through the world-emission platform <https://app.world-emission.com/detail/regional/nox/nox>. The CEDS-CarbonMonitor-MEIC (CCMM) are available upon a reasonable request from Bo Zheng at [bozheng@sz.tsinghua.edu.cn](mailto:bozheng@sz.tsinghua.edu.cn).

### Code availability

The CHIMERE code is available here: <http://www.lmd.polytechnique.fr/chimere/> (Menut et al., 2013;

585 Mailler et al., 2017). The CIF inversion system (Berchet et al., 2021) is available at <https://doi.org/10.5281/zenodo.6304912> (Berchet et al., 2022).

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## Competing Interests

The authors declare that they have no conflict of interest.

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