Quantifying Contributions of Local Emissions and Regional Transport to NO\textsubscript{X} in Beijing Using TROPOMI Constrained WRF-Chem Simulation

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Abstract: Air quality is strongly influenced by both local emissions and regional transport. Atmospheric chemical transport models can distinguish between emissions and regional transport sources in air pollutant concentrations. However, quantifying model inventories is challenging due to emission changes caused by the recent strict control measures taken by the Chinese government. In this study, we use NO\textsubscript{2} column observations from the Tropospheric Monitoring Instrument to retrieve top-down nitrogen oxide (NO\textsubscript{X}) emissions and quantify the contributions of local emissions and regional transport to NO\textsubscript{X} in Beijing (BJ), from 1 November 2018 to 28 February 2019 (W\textsubscript{2018}) and 1 November 2019 to 29 February 2020 (W\textsubscript{2019}). In W\textsubscript{2018} and W\textsubscript{2019}, the BJ bottom-up NO\textsubscript{X} emissions from the multi-resolution emission inventory for China in 2017 were overestimated by 11.8\% and 40.5\%, respectively, and the input of NO\textsubscript{X} from other cities to BJ was overestimated by 10.9\% and 51.6\%, respectively. The simulation using our adjusted inventory exhibited a much higher spatial agreement (slope = 1.0, \(R^2 = 0.79\)) and reduced a mean relative error by 45\% compared to those of bottom-up NO\textsubscript{X} emissions. The top-down inventory indicated that (1) city boundary transport contributes approximately 40\% of the NO\textsubscript{X} concentration in BJ; (2) in W\textsubscript{2019}, NO\textsubscript{X} emissions and transport in BJ decreased by 20.4\% and 17.2\%, respectively, compared to those of W\textsubscript{2018}; (3) in W\textsubscript{2019}, NO\textsubscript{X} influx substantially decreased (−69 g/s) in BJ compared to that of W\textsubscript{2018} despite negative meteorological conditions that should have increased NO\textsubscript{X} influx by +503 g/s. Overall, the contribution of intercity input to NO\textsubscript{X} in BJ has declined with decreasing emissions in the surrounding cities due to regional cooperative control measures, and the role of local emissions in BJ NO\textsubscript{X} levels was more prominent. Our findings indicate that local emissions may play vital roles in regional center city air quality.

Keywords: tropospheric monitoring instrument; weather research and forecasting with coupled chemistry; top-down nitrogen oxide emissions; transport; meteorology
1. Introduction

In the past decade, haze in China has been frequently reported at unprecedented PM$_{2.5}$ concentrations during the autumn and winter, particularly in the North China Plain (NCP). Nitrogen oxides (NO$_x$, NO + NO$_2$), which are primarily discharged by anthropogenic activities, such as fossil fuel combustion [1], are a group of reactive trace gases. NO$_x$ is not only toxic to human health, but also play a key role in the formation of secondary aerosol and tropospheric ozone [2]. Therefore, NO$_x$ is crucial atmospheric air pollutants.

A series of strict emissions reduction measures was implemented in China beginning in September 2013 [3]. In the past seven years, substantial manpower and material resources have been invested to improve air quality, and major measures have been taken for the atmosphere and ecosystem. To evaluate the effectiveness of air pollution control measures, an accurate and high spatiotemporal resolution of NO$_2$ distribution must be obtained [4–6]. Most previous studies have primarily used satellite observations, regional air quality model simulations, or ground-based observations to obtain the trace gas distribution [7–11]. Compared with these methods, the NO$_2$ concentration produced by models (Global 3-D model of atmospheric chemistry driven by meteorological input from the Goddard Earth Observing System (GEOS-Chem) [12], Community Multiscale Air Quality (CMAQ) model, Comprehensive Air Quality Model with Extensions (CAMx)) exhibits a higher spatiotemporal resolution in both the horizontal and vertical directions. Thus, timely NO$_x$ emission data are necessary.

A top-down inversion using satellite retrieval products of tropospheric vertical column densities (VCDs) of NO$_2$ is widely used to estimate NO$_x$ emissions. This method accounts for the nonlinear effects of horizontal transport, chemical loss, and deposition. Previous studies have estimated NO$_x$ emissions from various regions worldwide, including North America, Asia, the Middle East, and Europe [13–17]. This inversion has also been used to produce and validate NO$_x$ emission estimations from sources such as soil, lightning, power plants, aircraft, marine vessels, and urban centers [8,18–21].

The NO$_x$ concentration distribution is not only related to regional NO$_x$ emissions but also linked with the regional transport of NO$_x$. Several quantitative and qualitative approaches have been applied to evaluate the regional transport of emissions and its effect on local air quality [22–25]. The backward trajectory based on meteorology analysis has been used to qualitatively identify and describe the major transport direction and pathway of a target city [23,26–28]. Additionally, certain studies have used meteorological models or have combined meteorology and air quality observations to determine the relative importance of different source regions as a semi-qualitative assessment. Such studies include footprint analysis [29,30], the potential source contribution function analysis [31–33], and flux calculation [34]. Recently, regional transport studies have mostly been based on the chemical transport model [35] to sufficiently consider both the physical and chemical processes. For example, [36] found three primary transport pathways inside the Beijing–Tianjin–Hebei (BTH) region, i.e., the southwest, southeast, and anticlockwise pathway, with the Community Multiscale Air Quality and Integrated Source Apportionment Model (CMAQ-ISAM) [37] quantified the contribution of pollutant transport via PM$_{2.5}$ concentrations in 13 cities in the BTH region using the CMAQ-ISAM model. These findings have noted the necessity of a regional joint-control strategy. However, these studies are based on inventories relying on indirect information that is often outdated or incomplete, which is insufficient to support the development of a comprehensive joint-control strategy.

Herein, we combined the Tropospheric Monitoring Instrument (TROPOMI) observations and Weather Research and Forecasting with coupled chemistry (WRF-Chem) simulations to obtain accurate NO$_x$ emissions for the NCP region (covering 33° N–43° N, 109° E–123° E). We then identify the sources and sinks of NO$_x$ in NCP based on the regional transport flux. Finally, we quantify the contributions of NO$_x$ local emissions and regional transport to NO$_x$ concentrations, distinguishing the roles of meteorology and emissions to the city-
boundary transport flux, thus helping the government implement emissions reduction policies at the city level. The remainder of this paper is organized as follows. Section 2 provides detailed information about the model configuration, TROPOMI NO₂ retrieval, top-down NOₓ emissions inversion, and transport flux calculation. The top-down NOₓ inventory evaluation, relative contribution of NOₓ emissions and transport flux to NOₓ concentration, and role of emissions control and meteorology analysis are presented in Section 3. Section 4 presents the discussion. Finally, conclusions are present in Section 5.

2. Materials and Methods

2.1. Model Description and Configuration

The meteorological parameters and NOₓ concentration were provided by WRF-Chem version 4.0. This modeling system runs in two parts: the dynamic and chemical modules. The WRF model, which is a mesoscale numerical weather prediction system designed for meteorological research and numerical weather forecasting, is used as the dynamic module. A detailed description of the WRF model is available at the WRF website (http://www.wrf-model.org/index.php, last access: 15 March 2020). In addition to dynamical calculations, the chemical module is fully coupled with the WRF model online [38]. A detailed illustration of the chemical composition is provided by Grell et al. [39]. In this work, the simulation domain covered East China and its surrounding area, with a center point of 38.0° N, 115.6° E. The model’s horizontal resolution was selected to be 20 × 20 km², with 89 × 79 grids (there are 89 grids in the east-west direction and 79 grids in the north-south direction). From the ground level to the top pressure of 10 hPa, there were 44 vertical sigma layers for all grids. The initial meteorological fields and boundary conditions were from the 6-h final operational global analysis (FNL) data. The data were provided by the National Centers for Environmental Prediction (NCEP), and it exhibited a 1° × 1° spatial resolution. Furthermore, the NCEP Administrative Data Processing (ADP) Global Surface Observational Weather Data (ds461.0) and Upper Air Observational Weather Data (ds351.0) with 6-hourly temporal resolution were used to accurately reproduce the meteorology. The physical and chemical parameterization schemes adopted in this study are detailed in Table 1. Further configuration options of the model can be found in our previous study [40]. Table 2 demonstrates that the simulated wind fields are reproducible in comparison with those of the National Office for Oceanic and Atmospheric Administration (NOAA) observations (https://www.ncdc.noaa.gov/, last access: 25 June 2020).

The Carbon-Bond Mechanism version Z photochemical mechanism combined with the Model for Simulating Aerosol Interactions and Chemistry was used to simulate the chemical process in the atmosphere. The anthropogenic emissions were taken from the multi-resolution emission inventory for China in 2017 (MEIC-2017; http://www.meicmodel.org/, last access: 17 March 2020) [41,42]. The biogenic emissions were calculated online using the Model of Emissions of Gases and Aerosols from Nature embedded in the WRF-Chem model. The near-real-time fire emissions from the fire inventory of NCAR based on Moderate Resolution Imaging Spectroradiometer rapid response fire counts (https://www.acom.ucar.edu/acresp/forecast/fire-emissions.shtml, last access: 18 March 2020).

| Schemes                      | Description                                      |
|------------------------------|--------------------------------------------------|
| Microphysics                | Purdue Lin Scheme [43]                           |
| Longwave radiation          | Rapid radiative transfer model (RRTMG) scheme [44]|
| Shortwave radiation         | RRTMG scheme                                     |
| Cumulus parameterization     | Grell–Freitas Ensemble Scheme [45]               |
| Land surface                 | Unified Noah Land Surface Model [46]             |
| Planetary boundary layer     | Yonsei University scheme [47]                   |
| Chemical mechanism          | Carbon-Bond Mechanism version Z                  |
| Photolysis scheme            | Fast-J photolysis                                |
Table 2. Comparison of wind field from WRF simulation and NOAA observations.

| Meteorological Parameter | Statistic    | Unit    | Mean | Standard Deviation |
|--------------------------|--------------|---------|------|--------------------|
| Wind Speed               | MeanOBS      | (m/s)   | 4.32 |                     |
|                          | MeanPRD      | (m/s)   | 3.97 |                     |
|                          | Bias         | (m/s)   | 0.37 | ≤±0.5              |
|                          | GrossError   | (m/s)   | 1.13 | <2                 |
|                          | Root mean square error (RMSE) | (m/s) | 1.86 | ≤±0.5              |
| Wind Direction           | MeanOBS      | (°)     | 337  |                     |
|                          | MeanPRD      | (°)     | 288  |                     |
|                          | Bias         | (°)     | 5.25 | ≤10                |
|                          | GrossError   | (°)     | 48.32| ≤±30               |
|                          | RMSE         | (°)     | 79.81|                     |

2.2. TROPOMI Satellite Observation

TROPOMI is a passive trace gas spectrometer aboard the Sentinel-5 Precursor satellite, which was launched on 13 October 2017. TROPOMI measures the reflected sunlight of the atmosphere with spectral bands in 270–500 nm (ultraviolet–visible) 675–775 nm (near-infrared), and 2305–2385 nm (short-wave infrared) at a moderate resolution (0.25 nm to 0.6 nm), enabling daily global coverage with a spatial resolution of 7 km × 3.5 km at an exact nadir point [48,49]. In this work, the TROPOMI NO2 product typically followed the satellite trace gas retrieval algorithms by USTC (University of Science and Technology of China) [50]. During NO2 slant column density retrieval, the wavelength range of 405–465 nm was selected for the NO2 spectral fit performed with the QDOAS software package [51]. The QDOAS configurations follow the suggestions in the QA4ECV NO2 project [52]. For NO2 air mass factor (AMF) calculations, the Vector Linearized Discrete Ordinate Radiative-Transfer version 2.7 model [53] was used to calculate the stratospheric and tropospheric NO2 AMF pixel by pixel. During the RTM calculations, a priori NO2 profile with a high-resolution of 20 km × 20 km was taken from the monthly WRF-Chem simulations. Other information, such as cloud top pressure, cloud fraction, and surface albedo, was obtained from the operational TROPOMI cloud dataset [54]. To separate the stratospheric contribution from the total NO2 VCDs, a modified reference sector method, i.e., the STREAM algorithm [55], was applied. For the final tropospheric NO2 VCDs, we utilized a novel P-spline method to re-grid the NO2 VCDs to the Level-3 product at a resolution of 0.2° × 0.2° [56]. TROPOMI data were considered cloud-contaminated and filtered out by the re-gridding algorithm when the cloud radiance fraction was greater than 50% [57,58].

2.3. TROPOMI-Derived Top-Down NOx Emissions

TROPOMI-detected NO2 columns are sensitive to NOx emissions at the surface and are influenced by the NOx lifetime within the plumes. Previous studies [18,59,60] have confirmed that there is a non-linear relationship between changes in surface NOx emissions and changes in tropospheric NO2 columns. This is because an increase in the NOx concentration may promote or inhibit oxidation losses [11]. In the monthly assimilated inversion, we referenced the framework by [61]. The following four steps were followed. (1) Two simulations were performed, one with a priori emissions MEIC-2017 (Emeic) and another with anthropogenic NOx emissions, which increased by 20%. (2) The dimensionless scaling factor β, which reflects the sensitivity of NO2 VCDs to local NOx emissions via NOx-OH chemistry (indicated by Formula (1)), was calculated. (3) Top-down NOx emissions (Etop-down) were estimated with the modeled sensitivity β to scale the a priori emissions (indicated by Equation (2)). (4) The monthly regional NOx distribution was modeled with WRF-chem to combine the top-down emissions.

\[ \beta = \frac{AE/Emeic}{AC/C_{meic}} \] (1)
where $\Delta E$ is the change in anthropogenic NO$_X$ emissions in $E_{meic}$, which increased by 20%; $C_{meic}$ is the simulated NO$_2$ columns with $E_{meic}$; $\Delta C$ is the change in the monthly average tropospheric-simulated NO$_2$ VCDs after perturbing $E_{meic}$ anthropogenic NO$_X$ emissions by +20%. $C_{tropomi}$ is the monthly average tropospheric VCDs based on WRF-Chem NO$_2$ vertical profile with $E_{meic}$. Equation (2) in this work is a simplified calculation from [61]. In our study, the difference of top-down NO$_X$ emissions from Equation (2) is ~0.2% lower than that calculated by [61].

### 2.4. Horizontal Transportation Flux

To further investigate the source and sink centers of air pollution in the NCP region, the spatial distribution of the transport flux was calculated as follows:

**Step 1:** Calculate the average wind.

The weight coefficient was derived from the vertical distribution of NO$_2$ under the height of the tropopause layer in each grid. A high NO$_2$ concentration corresponds to a large coefficient. The average wind is equal to the sum of the wind of each layer multiplied by the weight coefficient of the corresponding height.

**Step 2:** Calculate the horizontal transport flux of grid A ($i, j$) [62].

\[
\text{East–west direction } \text{flux}_a(i,j) = C_{(i+1,j)} \times U_{(i+1,j)} - C_{(i-1,j)} \times U_{(i,j)}
\]

\[
\text{North–south direction } \text{flux}_b(i,j) = C_{(i,j+1)} \times V_{(i,j+1)} - C_{(i,j-1)} \times V_{(i,j)}
\]

**Horizontal net flux**

\[
\text{flux}_{\text{net}}(i,j) = \text{flux}_a(i,j) + \text{flux}_b(i,j)
\]

Equations (3) and (4) provide the basic equations to calculate the transport flux of each grid in the region. Where $C$ is the tropospheric NO$_2$ column from the WRF-Chem NO$_2$ simulation. $U$ and $V$ represent the winds in the east–west and north–south directions, respectively. Easterly and northerly are both positive. Equation (5) quantifies the net flux of air pollutants in each grid. A positive net-flux represents the grid outputting NO$_2$ to its surrounding four grids. By contrast, negative values indicate that the four surrounding grids input NO$_2$ into the grid.

### 2.5. Ancillary Data

In-situ measurements for NO$_2$ analysis were provided by the operational stations of the China Environmental Observation Network operated by the China National Environmental Monitoring Centre (CNEMC; http://www.cnemc.cn/en/, last access: 28 June 2020). The 0–23-h concentrations of these measurements spanning from November 2018 to February 2019 and from November 2019 to February 2020 were used in this study. FNL data were obtained from NCEP FNL Operational Model Global Tropospheric Analyses (https://rda.ucar.edu/datasets/ds083.2/index.html, last access: 15 March 2020). NCAR archive ds351.0 data were downloaded from NCEP ADP Global Upper Air Observational Weather Data Centre (https://rda.ucar.edu/datasets/ds351.0/index.html, last access: 15 March 2020).

### 3. Results and Discussion

#### 3.1. Top-down Emissions Evaluation

Wintertime air pollution is extremely severe in northern Chinese cities. In February 2017, the Ministry of Environmental Protection in China released the Air Pollution Prevention and Control Work Plan for BTH and its neighboring regions, which referred to the cities that transmit air pollution as “2 + 26” cities (represented by triangles in Figure 1) for the first time. The “2 + 26” cities represent 28 NO$_X$ polluted cities in the Beijing-Tianjin-Hebei and surrounding region, including 2 cities with significant NO$_X$ pollution (Beijing (BJ) and Tianjin (TJ)) and 26 cities (the rest of the cities) with high NO$_X$ concentrations. Since the implementation of air emissions control in the “2 + 26” cities, the concentration of air pollu-
tion has been significantly reduced, and the air quality has been greatly improved. Monthly top-down NOx emissions were derived using the method described in Section 2. Figure 1 displays the spatial distribution of monthly averaged MEIC-2017 and top-down surface NOx emissions. "2 + 26" cities dominated in high NOx emissions. Figure 2 illustrates the regional emission differences. As exhibited in Figure 2a,b, the top-down NOx emissions in the south of BJ, southeast of Hebei province, northwest of Shandong province, and northern of Henan province were significantly reduced by 25–58% when compared to those of MEIC-2017. By contrast, NOx emissions in Taiyuan (TY) and Tangshan (TS) slightly increased. This phenomenon can also be observed in Figure S2a–f, except for January 2020. In February 2020, NO2 VCDs in the south of TY simulated from top-down W_2019 are slightly higher than that simulated from MEIC-2017 (Figure S2h). However, NO2 VCDs in the south of TY simulated from top-down W_2019 is slightly lower than that simulated from top-down W_2018 (Figure S2i) during February 2020. This means that NOx emissions in the south of TY during February 2020 are higher than that in the same period of 2017, but lower than that in the same period of 2019. Figure 2c displays the difference between the top-down inventories in W_2018 and W_2019. NOx emissions in all "2 + 26" cities significantly decreased. The monthly variation analysis (Figure S2j–l) shows that NOx emissions in the south of BJ and southeast of TJ and Langfang (LF) slightly increased, while NOx emissions in the other cities decreased in November 2019 compared to those in November 2018, particularly in central Shanxi. In December 2019, most cities in the southeast of Taihang Mountain exhibited significant NOx emission reductions compared to those in December 2018, while NOx emissions in central Shanxi province and Tangshan increased. In January 2020, NOx emissions across the entire study area exhibited a pronounced decreasing trend compared to those in January 2019; and the NO2 concentration in the BTH demonstrated a reduction of approximately 50% compared with that in January 2019. This result may be attributed to the lockdown policy during the outbreak of the COVID-19 pandemic. Additionally, it may have also resulted from the influence of the Spring Festival holiday.

Figure 3 compares the monthly averaged NO2 tropospheric VCDs simulated from MEIC-2017, top-down NOx emissions, and their differences. Figure 3a illustrates the monthly average distribution of tropospheric NO2 VCDs in W_2018, which was simulated using the MEIC-2017 inventory, and Figure 3b is the same as Figure 3a, except that it uses the top-down W_2018 NOx inventory, which was retrieved from the TROPOMI NO2 concentration during W_2018. Both Figures 3a and 3b exhibit peak NO2 in BJ, TJ, TS, and certain cities in southeast of the Taihang mountains. Figure 3c displays the monthly averaged distribution of tropospheric NO2 in W_2019, which uses the same inventory as that in Figure 3a. The role of meteorological conditions can be understood based on the difference between Figures 3a and 3c. The overall meteorology changes between W_2018 and W_2019 slightly influenced the NO2 VCDs and their spatial distribution. Figure 3d is the same as Figure 3c, except that it uses the top-down W_2018 inventory, the VCD distribution of which is similar to that of Figure 3b. Figure 3e is the same as Figure 3c, except that it uses the top-down W_2019 inventory, which was retrieved from the TROPOMI NO2 observations during W_2019. Figure 3f indicates the difference between Figures 3a and 3b, reflecting the changes in the tropospheric NO2 VCDs during W_2018 when using the calibration inventory compared to that when using MEIC-2017. In most cities, the NO2 concentration dropped significantly, while strong (18–32%) NO2 concentration increases occurred in the center of Shanxi province and TS after NOx emission adjustments during W_2018. In February 2019, the simulated NO2 VCDs using top-down W_2018 inventory was significantly lower than that using MEIC-2017 in the study area (Figure S3a–d,i–l). This result was due to reduced NOx emissions over the Spring Festival holiday. Figure 3g indicates the difference between Figures 3c and 3d, reflecting the changes in the tropospheric NO2 VCDs during W_2019 when using the top-down W_2018 inventory compared to that when using MEIC-2017. The decline in NO2 VCDs in Figure 3g is more pronounced than that in Figure 3f, which was caused by the unfavorable meteorological condition
during W_2019 compared to that in W_2018. Figure 3h indicates the differences between Figures 3c and 3e, reflecting the changes in the tropospheric NO2 VCDs during W_2019 when using the calibration inventory compared to that when using MEIC-2017. The NO2 VCDs simulated from the top-down-W_2019 are much lower than those from MEIC-2017, especially in January and February 2020. The NO2 concentration in the BTH region particularly decreased. The decline in NO2 VCDs in Figure 3h is more pronounced than that in Figure 3f, which was caused by the significantly decreased NOx emissions during W_2019. This substantial drop in the NO2 concentration was due to a combination of strict emission controls and the COVID-19 lockdown.

Figure 1. Four-month averaged nitrogen oxide (NOx) emissions for (a) the multi-resolution emission inventory for China in 2017 (MEIC-2017), (b) top-down 1 November 2018 to 28 February 2019 (W_2018), and (c) top-down 1 November 2019 to 29 February 2020 (W_2019). (a) is four-month average for January, February, November, and December. ‘Four-month averaged’ in this paper refers to average of four months as indicated above, while the supplementary material is average of each month.

Figure 2. Differences in four-month averaged NOx emissions between MEIC-2017 and top-down NOx emissions; (a) indicates top-down W_2018 minus MEIC-2017; (b) indicates top-down W_2019 minus MEIC-2017. (c) Change in surface NOx emissions between top-down W_2018 and top-down W_2019 (top-down W_2019 minus top-down W_2018).
Figure 3. Four-month averaged tropospheric NO$_2$ VCDs simulated from MEIC-2017, top-down NO$_X$ emissions, and their differences. (a) Simulated NO$_2$ VCDs in W 2018 with MEIC-2017 inventory. (b) Same as (a) except with top-down W 2018 inventory. (c) Simulated NO$_2$ VCDs in W 2019 with MEIC-2017 inventory. (d) Same as (c) except with top-down W 2018 inventory. (e) Same as (c) except with top-down W 2019 inventory. (f) Difference between (a) and (b) ((b) minus (a)). (g) Difference between (c) and (d) ((d) minus (c)). (h) Difference between (c) and (e) ((e) minus (c)).

Figure 4 illustrates the simulation with optimized NO$_X$ emissions and removing the model’s systematic bias with respect to the hourly surface in-situ NO$_2$ concentration, leading to an improved spatial agreement (slope = 1.0, $R^2 = 0.79$) and reducing the mean relative error by 45%. Our findings demonstrate that air quality model simulations combined with satellite observations can be used to adjust surface NO$_X$ emissions before more rigorous bottom-up emissions inventories are released.
3.2. Regional Transport Flux

Studies have been extensively performed to investigate the impact of regional transport on BJ airborne species [63–65]. Figure 5a–h display the NO$_2$ regional transport fluxes and their differences calculated using formulas (3–5), which correspond to Figure 3a–h, respectively. During the two periods, TS, TJ, and cities in southeast of Taihang mountain (Baoding (BD), Shijiazhuang, Xingtai, Handan, Anyang, and Hebi) were significant NO$_2$ sources when using MEIC-2017 (Figure 5a,c), and they were lesser sources when using top-down NO$_X$ emissions (Figure 5f–h). These results indicate great achievements in emission reductions due to the implementation of strict emissions control measures. By contrast, cities in central Shanxi province exhibited more NO$_X$ emissions when using top-down NO$_X$ emissions than when using MEIC-2017 emissions. This result means that TY and its surrounding cities to the north need to strengthen their emission reduction management.

A city that exhibits a high NO$_2$ concentration does not necessarily severely emit NO$_X$. Moreover, the wind field makes NO$_2$ flow into and accumulate in that city, which also leads to heavy NO$_2$ pollution. For example, BJ exhibits a high NO$_2$ concentration during the two periods; however, it is a NO$_X$ sink. In addition, the regional distribution of sources and sinks agrees with that of the NO$_X$ emissions. Satellite observations can be used to qualitatively analyze source distribution and strength without updating the bottom-up emissions. The distribution of sources in the regional transport flux is affected by the wind field. When the wind speed is low, and the wind direction frequently changes, the transport flux agreement with bottom-up NO$_X$ emissions improves. These findings are considerably important for implementing air quality control measures during special events.
Figure 5. Four-month averaged tropospheric NO\textsubscript{2} regional transport flux (region with positive value indicates source of NO\textsubscript{2}, and negative value indicates sink of NO\textsubscript{2}) derived from MEIC-2017, top-down NO\textsubscript{X} emissions, and their differences. (a) Calculated NO\textsubscript{2} flux in W\textsubscript{2018} with MEIC-2017 inventory. (b) Same as (a) except with top-down W\textsubscript{2018} inventory. (c) Calculated NO\textsubscript{2} flux in W\textsubscript{2019} with MEIC-2017 inventory. (d) Same as (c) except with top-down W\textsubscript{2018} inventory. (e) Same as (c) except with top-down W\textsubscript{2019} inventory. (f) Difference between (a) and (b) ((b) minus (a)). (g) Difference between (c) and (d) ((d) minus (c)). (h) Difference between (c) and (e) ((e) minus (c)).

3.3. Assessment of City Boundary Transport Around BJ

To determine the effectiveness of emissions control measures on each city level, the relative contributions of local NO\textsubscript{X} emissions and city boundary transport fluxes must be quantified. Figure 6 represents the NO\textsubscript{X} transport fluxes surrounding BJ, the calculation method of which is derived from [35]. BJ is a large NO\textsubscript{X} sink in both periods. Zhangjiakou (ZJK) and BD, which are upwind of BJ, input NO\textsubscript{X} into BJ (the NO\textsubscript{X} transported through the outer boundary is a mixture of different sources and does not solely stem from the neighboring cities). BJ simultaneously exports NO\textsubscript{X} to the downwind cities, such as Chengde (CD), TJ, and LF. Figure 6a,c demonstrate the transport fluxes between BJ and its surrounding 5 cities during W\textsubscript{2018} and W\textsubscript{2019}, which are simulated using MIEC-2017 (same MEIC-2017 NO\textsubscript{X} emissions but different meteorology). In that case, the difference (Figure 6c minus Figure 6a) in the boundary transport flux of BJ is due to the meteorology change between the two periods. In W\textsubscript{2019}, BJ exhibits enhanced NO\textsubscript{X} input from
surrounding cities by 597 g/s (13.2%) compared to that of W_2018. The unfavorable meteorology in W_2019 can account for the 13.2% NO\textsubscript{X} concentration enhancement. Figure 6b exhibits the cities boundary transport flux between BJ and its surrounding cities during W_2018, which is the same as Figure 6a except that it uses the top-down W_2018 inventory. Figure 6d is the same as Figure 6c except that it uses the top-down W_2018 inventory. Comparing Figures 6d and 6b, i.e., the weather of W_2019 compared to that of W_2018, demonstrated an increase in the BJ input flux by 503 g/s (12.4%), which also reflects the adverse meteorological conditions in W_2019 causing NO\textsubscript{X} concentration enhancement. Figure 6e is the same as Figure 6d, except that the top-down W_2019 inventory was used. Comparing Figures 6e and 6d, the NO\textsubscript{X} emissions in top-down W_2019 compared to that in the top-down W_2018, for the BJ input were reduced by 1202 g/s (26.3%), which also reflects the favorable emissions control in W_2019 for NO\textsubscript{X} concentration reduction. Comparing Figures 6b and 6a, the boundary transport flux simulated using MEIC-2017 overestimates the NO\textsubscript{2} inflow flux in BJ by 443 g/s (10.9%) during W_2018. Comparing Figures 6e and 6c demonstrates that the boundary transport flux simulated and calculated using MEIC-2017 overestimates the NO\textsubscript{2} inflow flux in BJ by 1379 g/s (51.6%) during W_2019. Comparing Figures 6e and 6b reflects that the input of NO\textsubscript{2} from cities surrounding BJ exhibits a decrease in NO\textsubscript{X} emissions of 699 g/s (17.2%) from W_2018 to W_2019. This result is caused by the co-effects of meteorology and emissions changes. The favorable contribution of the emission reduction policy is 26.3%, while the unfavorable contribution of weather is approximately 13%.

Figure 6. Four-month averaged tropospheric NO\textsubscript{X} transport fluxes across city boundary for BJ. (a) Calculated NO\textsubscript{2} flux in W_2018 with MEIC-2017 inventory. (b) Same as (a) except with top-down W_2018 inventory. (c) Calculated NO\textsubscript{2} flux in W_2019 with MEIC-2017 inventory. (d) Same as (c) except with top-down W_2018 inventory. (e) Same as (c) except with top-down W_2019 inventory. Red number means net flux is inflow the NO\textsubscript{X}. Blue number means net flux is outflow NO\textsubscript{X}. Unit of flux is g/s.
Figure 7 displays the NO\textsubscript{X} emissions and net fluxes of NO\textsubscript{X} and NO\textsubscript{2} from W\textsubscript{2018} to W\textsubscript{2019}. As illustrated in Figure 7a, compared with W\textsubscript{2018}, BJ NO\textsubscript{X} emissions remain unchanged (both use MEIC-2017), while NO\textsubscript{X} input to BJ increased by 13.2\% (increase of 597 g/s) due to unfavorable meteorological conditions. BJ total NO\textsubscript{X} increment (including NO\textsubscript{X} emissions and NO\textsubscript{X} net flux) increased by 4.9\% during W\textsubscript{2019}. After the inventory adjustment, the top-down inventory demonstrates that, in W\textsubscript{2019}, NO\textsubscript{X} emissions and transport in BJ decreased by 20.4\% (reduction of 1386 g/s) and 17.2\% (reduction of 699 g/s), respectively. BJ total NO\textsubscript{X} increment decreased by 19.2\% compared to that of W\textsubscript{2018} (Figure 7b).

In the two periods, BJ NO\textsubscript{X} local emissions using MEIC-2017 were overestimated by 11.8\% and 40.5\%; and the NO\textsubscript{X} transport flux was underestimated by 10.9\% and 51.6\%. Based on the top-down inventory, the proportions of emissions in the two winters were 62.5\% and 61.5\%, and the proportions of the city boundary transport fluxes were 37.5\% and 38.5\%.

4. Discussion

The assimilation of satellite observations in atmospheric chemical transport models is vital for improving the accuracy of air pollutant simulations. This study derived optimized NO\textsubscript{X} emissions based on the TROPOMI/WRF-Chem relative difference using a mass balance approach. The pollutant distribution from TROPOMI is significantly impacted by meteorological conditions [66–68]. Thereby, the accuracy of this assimilated method (top-down emissions) decreases when strong transport on NO\textsubscript{2} occur. The Inversion errors and model-based simulation errors relating local NO\textsubscript{2} columns to local emissions constitute the uncertainty of top-down emissions. The overall error of the top-down emissions over NCP areas is estimated at ~50\% [16].

Our results have taken COVID-19 lockdown and spring festival into account, which shows dramatic reductions (~50\%) in NO\textsubscript{X} emissions over January 2020. Our results comply with the literature, [69] indicated that concentrations of NO\textsubscript{2} decreased by ~45.1\% observed by ground-based observations during January 2020 compared with that in January 2019. According to Figure 3, the slope of “Prior” is greater than 1, and the slope of TROPOMI Posterior is equal to 1. It means that the simulated NO\textsubscript{2} before assimilation is higher than the observation and more accurate after calibration.
Several studies have found that accurate representation of noonday NO\textsubscript{2} columns from highly localized sources requires a high model resolution. Since NO\textsubscript{X} emissions show a strong variation on the 20 × 20 km\textsuperscript{2} scale applied in this study. Increasing the model resolution can better represent these local gradients, thus improving the simulation of NO\textsubscript{2} concentrations [17,70,71].

The relative proportion of NO\textsubscript{X} transport in BJ is about 40%. Similar results have been obtained in studies using a comparable regional model [65]. During the Asia-Pacific Economic Cooperation Forum week, non-local emissions contributed to 41.3% of the total PM\textsubscript{2.5} concentration in BJ, emphasizing the correctness of strict and synergistic emissions control for BJ and its surrounding provinces.

Future work can be continued on the change of ozone and PM\textsubscript{2.5} affected by the NO\textsubscript{X} emission variety after TROPOMI assimilation.

5. Conclusions

By optimizing NO\textsubscript{X} emissions with TROPOMI observations, sufficient NO\textsubscript{2} simulation performance can be achieved, and the mean relative error can be reduced by 45%. Cities with high NO\textsubscript{X} emissions exhibit a pronounced decline (by 25–58%) after inventory adjustment. Particularly in January 2020, NO\textsubscript{X} exhibited a reduction of approximately 50% in BTH compared with that in January 2019. For BJ, in W\textsubscript{2018} and W\textsubscript{2019}, the bottom-up NO\textsubscript{X} emissions were overestimated by 11.8% and 40.5% when compared to the top-down NO\textsubscript{X} emissions, respectively. According to the top-down inventory, compared with those of W\textsubscript{2018}, NO\textsubscript{X} emissions in BJ decreased by 20.4% during W\textsubscript{2019}. A city with decreased NO\textsubscript{2} concentration may be due to not only reduced NO\textsubscript{X} emissions but also lower NO\textsubscript{2} inflows. We found that NO\textsubscript{X} emissions can account for approximately 60% of the NO\textsubscript{X} concentration, and the remaining 40% is caused by regional transport.

Moreover, by analyzing the regional horizontal transport flux, we can identify the major sources and the strengths of NO\textsubscript{X} emissions without an immediately updated bottom-up inventory. This approach can be used to evaluate the effectiveness of implementing emission control measures, particularly during special events.

Transport fluxes depend on wind speed and emissions. This NO\textsubscript{X} inflow and outflow fluxes can be calculated according to the city boundaries transport. In W\textsubscript{2018} and W\textsubscript{2019}, the BJ NO\textsubscript{X} input fluxes using MEIC-2017 were overestimated by 10.9% and 51.6%, respectively. The top-down inventory indicated that the major NO\textsubscript{X} input and output directions and their specific NO\textsubscript{X} flux values for BJ, which were the NO\textsubscript{X} inflows from ZJK and BD and outflows to CD, TJ, and LF during the wintertime. Compared with W\textsubscript{2018}, the NO\textsubscript{X} input flux decreased by 17.2% (−699 g/s) in BJ during W\textsubscript{2019}, which resulted from negative meteorological conditions (+503 g/s) and positive NO\textsubscript{X} emission controls (−1202 g/s). These results illustrate the effectiveness of reducing local emissions through emissions controls.

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Figure S1: Monthly NO\textsubscript{X} emissions. (a–b) Taken from the MEIC-2017. (e–h) Top-down NO\textsubscript{X} emissions derived from TROPOMI NO\textsubscript{2} VCDs in W\textsubscript{2018}. (i–l) Same as (e–h) except in W\textsubscript{2019}. Figure S2: Difference in surface NO\textsubscript{X} emissions between MEIC-2017 and top-down NO\textsubscript{X} emissions (a–h); (i–l) Change in surface NO\textsubscript{X} emissions between top-down W\textsubscript{2018} and top-down W\textsubscript{2019}. Figure S3: Monthly averaged tropospheric NO\textsubscript{2} VCDs simulated from MEIC-2017 (a–h) and top-down NO\textsubscript{X} emissions (i–t). Note that, (a–d) and (i–l) using the same meteorological condition (W\textsubscript{2018}), while the different NO\textsubscript{X} inventories. (e–h) and (m–t) also using the same meteorological condition (W\textsubscript{2019}), while the different NO\textsubscript{X} inventories. Figure S4: Variation in monthly averaged tropospheric NO\textsubscript{2} VCDs simulated from MEIC-2017 and top-down NO\textsubscript{X} emissions. Figure S5: Monthly averaged tropospheric NO\textsubscript{2} regional transport flux drive from MEIC-2017 (a–h) and top-down NO\textsubscript{X} emissions (i–t). Note that, (a–d) and (i–l) using the same meteorological condition (W\textsubscript{2019}), while the different NO\textsubscript{X} inventories. (e–h) and (m–t) also using the same meteorological condition (W\textsubscript{2019}), while the different NO\textsubscript{X} inventories. Figure S6: Variation in monthly averaged tropospheric NO\textsubscript{2} regional transport flux drive from MEIC-2017 and top-down NO\textsubscript{X} emissions. Note
that, (a–d) using the meteorological condition in W_2018, while (e–l) using the same meteorological condition (W_2019).

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**Conflicts of Interest:** The authors declare no conflict of interest.

**Abbreviations**
The following abbreviations are used in this manuscript:

- **“2 + 26” cities**
  - BJ Beijing
  - TJ Tianjin
  - SJZ Shijiazhuang
  - TS Tangshan
  - BD Baoding
  - LF Langfang
  - CZ Cangzhou
  - HS Hengshui
  - HD Handan
  - XT Xingtai
  - TY Taiyuan
  - YQ Yangquan
  - ChangZ Changzhi
  - JC Jincheng
  - JN Jinan
  - ZB Zibo
Another two neighboring cities in the north of Beijing

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