Spatial Distribution and Temporal Trend of Tropospheric NO₂ over the Wanjiang City Belt of China

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We utilize the tropospheric NO₂ columns derived from the observations of Ozone Monitoring Instrument (OMI) onboard AURA to analyze the spatial distributions and temporal trends of NO₂ in Wanjiang City Belt (WCB) of China from 2005 to 2016. The aim of this study is to assess the effect of industrial transfer policy on the air quality in WCB. Firstly, we used the surface in situ NO₂ concentrations to compare with the OMI-retrieved tropospheric NO₂ columns in order to verify the accuracy of the satellite data over the WCB area. Although it is difficult to compare the two datasets directly, the comparison results prove the accuracy of the OMI-retrieved tropospheric NO₂ columns in cities of WCB. Then, the spatial distributions of the annual averaged tropospheric NO₂ total columns over Anhui Province show that NO₂ columns were considerably higher in WCB than those in other areas of Anhui. Also, we compared the spatial distributions of the total NO₂ columns in 2005 through 2010 and in 2011 through 2016 and found that the total NO₂ columns in WCB increased by 19.9%, while the corresponding value increased only 13.9% in other Anhui areas except the WCB area. Furthermore, the temporal variations of NO₂ columns show that although the NO₂ columns over WCB and Anhui increased significantly from 2005 to 2011, they decreased sharply from 2011 to 2016 due to the strict emission reduction measures in China. Finally, the HYSPLIT model was used to analyze the origins of NO₂ and transport pathways of air masses in a typical city, Ma’anshan city.

1. Introduction

Nitrogen dioxide (NO₂) is a reactive, short-lived atmospheric trace gas with both natural and anthropogenic sources. Major sources of NO₂ are fossil fuel combustion, biomass burning, soil emissions, and lightning [1]. NO₂ is a toxic air pollutant on the condition of high concentration and plays an important role in tropospheric chemistry as a precursor of tropospheric ozone and secondary aerosols [2]. Observations of the spatiotemporal variations of NO₂ form the basis of understanding the spatial distributions and temporal trends of NO₂.

Many techniques and methods have been successfully used in monitoring atmospheric NO₂ based on surface in situ measurements, remote sensing from satellite sensors, and ground-based instruments [3–8]. Although the in situ measurements and remote sensing from ground-based instruments show high accuracy and precision, their usefulness in determining the spatiotemporal distributions of trace gases is limited due to their sparse spatial and temporal coverage. Space-based measurements provide information on NO₂ distributions at a large scale and over areas where in situ and ground-based systems cannot be easily deployed [9].

A series of sun-synchronous satellites were launched with spectrometers, which allowed scientists to observe the global distribution of several important tropospheric trace gases including NO₂, SO₂, and O₃. Satellite observations make it easy to understand the spatiotemporal variations of atmospheric NO₂ [10–13]. Lamsal et al. examined the seasonal variation in lower tropospheric NO₂ by the observation of the OMI, in situ surface measurements, and a global GEOS-Chem model [9]. Ul-Haq et al. applied the
linear regression model for tropospheric NO\textsubscript{2} and the anthropogenic NO\textsubscript{x} emissions using OMI data [14]. Gu et al. used the NO\textsubscript{2} columns observed from OMI and the Community Multiscale Air Quality (CMAQ) model to derive the ground-level NO\textsubscript{2} concentrations in China [15]. Sharma et al. presented the temporal variations of surface NO\textsubscript{2} during 2012 to 2014 at an urban site of Delhi, India [16]. Varotsos et al. found a progressive increase of mean values of NO\textsubscript{2}/NO\textsubscript{x} versus NO\textsubscript{2} when the level of NO\textsubscript{x} increases in Athens, Greece [17].

Han et al. used the tropospheric NO\textsubscript{2} columns observed from OMI to compare with the bottom-up emissions of NO\textsubscript{2} derived from the CMAQ model and three emission inventories over East Asia [18]. Liu et al. [19] analyzed the NO\textsubscript{x} emission trends and the major reasons for changes over China from satellite observations and the Multi-resolution Emission Inventory for China (MEIC). The emissions derived from the bottom-up method based on the inventory and the top-down method based on OMI observations showed good agreement. Lambsal et al. used aircraft and surface in situ measurements as well as ground-based remote sensing data to validate the OMI retrieval of tropospheric NO\textsubscript{2} [20]. Ialongo et al. compared the OMI NO\textsubscript{2} total columns with the ground-based remote sensing data collected by the Pandora spectrometer to evaluate the satellite data product at high latitudes [21]. Tong et al. utilized OMI observations and Air Quality System (AQS) data to study the long-term NO\textsubscript{x} trends over eight large US cities [22]. McLinden et al. combined OMI observations with a regional-scale air quality model to monitor the air quality of the Canadian Oil Sands [23]. Kim et al. used three regression models in conjunction with OMI tropospheric NO\textsubscript{2} columns to estimate the surface NO\textsubscript{2} volume mixing ratio in five cities of South Korea [24].

Tropospheric NO\textsubscript{2} vertical columns obtained from satellite instruments have been widely used to study NO\textsubscript{x} pollutions over China [25–27]. Lin found that the anthropogenic emissions are the dominant source of NO\textsubscript{2} over East China [28]. Understanding global and regional distributions and temporal trends of the pollution gases provides a basis for development of mitigation strategies. Most studies focus on the air quality of North China Plain, Pearl River Delta, and Yangtze River Delta [28, 29], which are the economic development centers of China and have regional heavy pollutions. But we pay little attention to mideastern China. Mideastern China is experiencing significant socioeconomic changes following the national industrial transfer strategies. Excessive development in a limited number of regions tends to be unsustainable because of limited resources. So industrial transfer is performed from the coastal areas to the inland areas [30]. Wanjiang City Belt (WCB) was established in January 2010 by National Development and Reform Commission (NDRC) of China to make the industrial transfer from the Yangtze River Delta and other mega-regions to Anhui Province [31]. Anhui is located in the mideastern region of China.

The aim of this study is to describe the spatial distributions and temporal trends of tropospheric NO\textsubscript{2} based on satellite observations in twelve years in Anhui, in order to assess the effect of industrial transfer policy on the air quality in WCB. This paper is organized as follows. Firstly, the materials and methods used are described in Section 2. The area of Wanjiang City Belt, satellite data, surface in situ data, and HYSSPLIT model used in the analysis are introduced. Secondly, results and discussion are presented in Section 3. Comparisons of satellite data with surface in situ data for NO\textsubscript{2} in WCB are made in Section 3.1. The spatial distributions of the annual averaged tropospheric NO\textsubscript{2} total columns in Anhui Province are shown in Section 3.2. The variation of the total tropospheric NO\textsubscript{2} columns before and after establishment of the WCB is discussed in Section 3.3. Also, the seasonal variations of tropospheric NO\textsubscript{2} are analyzed in Section 3.4. Finally, conclusions are presented in Section 4.

2. Materials and Methods

2.1. The Introduction of Wanjiang City Belt. Industrial transfer is one of the important national strategies in China. The construction of WCB is the first approved demonstration area for industrial transfer on the national level [32]. WCB comprises 59 counties in Anhui Province along the Yangtze River, including Anqing, Chaohu, Chizhou, Chuzhou, Hefei, Ma’anshan, Tongling, Wuhu, Xuancheng, Jin’an District, and Shucheng County of Lu’an [33]. Figure 1 shows the location of Anhui Province, and the green area represents WCB.

Anhui Province has diverse topography, as shown in Figure 2. The north of Anhui belongs to the North China Plain, while the north-central areas are part of the Huai River Plain. The two regions are flat with dense population. The south of the province is characterized by uneven topography. The Yangtze River runs through the south of Anhui between the Dabie Mountains and a series of hills.

2.2. Satellite Data. OMI is an ultraviolet/visible spectrometer aboard the NASA’s EOS Aura satellite. The instrument provides information on trace gases, such as ozone (O3), sulfur dioxide (SO\textsubscript{2}), and nitrogen dioxide (NO\textsubscript{2}), and other pollutants retrieved from the spectral region between 270 and 500 nm [34]. EOS Aura circles in a polar synchronous orbit with a 98.2° inclination to the equator, at an altitude of around 705 km. The overpass times are about 13:45 mean local solar time [11, 34].

In the present study, we collect the OMI-retrieved tropospheric NO\textsubscript{2} columns from Royal Netherlands Meteorological Institute (KNMI) DOMINO v2.0 products from 2005 to 2016, which are available at http://www.temis.nl/airpollution/no2col/no2regioom/month_v2.php. The spatial resolution is 0.125 × 0.125° latitude-longitude, which has been widely used for scientific applications [21, 35, 36]. We used the monthly mean data to analyze the spatial distribution and temporal trends.

2.3. Surface In Situ Data. The Chinese Ministry of Environmental Protection issued “construction scheme of National Environmental Monitoring Network (in cities at the
Figure 1: The location of (a) Anhui Province and (b) Wanjiang City Belt (WCB).

Figure 2: Digital elevation model (DEM) of Anhui (m).
prefecture level and above) during the Twelfth Five-Year Plan” in 2012. 1436 monitoring stations have been set up in 338 cities in China since then. These surface monitoring stations provide the concentrations of NO2, SO2, PM10, CO, O3, and PM2.5 and visibility. Chinese National Environmental Monitoring Center (CNEMC) is responsible for publishing the near-real-time data collected from all monitoring stations publicly. The ground-level NO2 concentrations are mainly obtained by a nitrogen oxide analyzer based on the gas-phase chemiluminescence method. The surface in situ data of NO2 are only accessible from 2015 to 2016, so we use the surface data in the two years. The temporal resolution of the ground-level NO2 concentrations is one datum per hour in each monitoring station.

2.4. HYSPLIT Model. In this study, we used the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model developed by National Oceanic and Atmospheric Administration (NOAA) to simulate the back trajectories of air mass [37]. The HYSPLIT model is a complete system, which has been extensively used in calculation of air mass trajectories, atmospheric transport, and dispersion. The model is often used to locate the origin of air masses and build the relationships between source and receptor by back trajectory analysis [38]. The input for the HYSPLIT model is the Global Data Assimilation System (GDAS) meteorological data, which are available at the GDAS website (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1).

3. Results and Discussion

3.1. Comparison of Satellite Data with Surface Data. We used the surface in situ data to compare with the satellite data in order to verify the accuracy of the OMI-retrieved tropospheric NO2 columns. The ground-level NO2 concentrations observed by the CNEMC stations in 2015 and 2016 were utilized. The satellite data were extracted corresponding to the data grid in which the monitoring stations are located. We collected the surface data from 13:00 to 14:00 everyday, as this time period coincides with the OMI overpass local time. Figure 3 shows the selected CNEMC stations in Anhui Province.

The monthly averaged data from each CNEMC station in Anhui are compared with satellite data. Figure 4 shows the comparison results of the two datasets. From Figure 4, the two data show almost the same variation trend of NO2 in each city. The Pearson linear correlation coefficients of the data for each area are high, as listed in Table 1. We used the 2-tailed test to test the statistical significance of the
Figure 4: Continued.
correlation coefficient, and the correlation is significant at the 0.05 level. The two datasets have different spatial scale representativeness and surface sensitivity, so it is unreasonable to compare the two datasets directly. However, our comparison results prove the accuracy of the OMI-retrieved tropospheric NO\textsubscript{2} columns in the WCB area.

3.2. Spatial Distributions of Tropospheric NO\textsubscript{2} in WCB.
Figure 5 plots the spatial distributions of the annual averaged tropospheric NO\textsubscript{2} total columns over Anhui throughout the years from 2005 to 2016. It is found that the NO\textsubscript{2} columns were considerably higher in WCB than those in other areas of Anhui. The annual averaged NO\textsubscript{2} column reached 811 × 10\textsuperscript{15} molec./cm\textsuperscript{2} in the WCB region from 2005 to 2016, while the annual averaged NO\textsubscript{2} column was 733 × 10\textsuperscript{15} molec./cm\textsuperscript{2} in other areas of Anhui during the same period. The p value is 0.09221 when the two-sample t test is used. Figure 6 is the plot of the histogram of the annual averaged tropospheric NO\textsubscript{2} total columns in each city of Anhui Province. As can be seen from Figure 6, the highest NO\textsubscript{2} columns appeared in the Ma’anshan city, where iron and steel industry is the major industry with high emission of pollutions. The spatial distributions of annual averaged tropospheric NO\textsubscript{2} columns in this province agree with the results of satellite-retrieved NO\textsubscript{2} emissions in eastern China in other studies [1, 10].

3.3. Temporal Trends of Tropospheric NO\textsubscript{2} in WCB. The WCB region is established in 2010, so we compared the tropospheric NO\textsubscript{2} columns in this region before and after the year of 2010. Figure 7 shows the spatial distributions of the total NO\textsubscript{2} columns in 2005 through 2010 and in 2011 through 2016 as well as the difference between the two periods. The difference between the two periods represents the change in the total NO\textsubscript{2} columns. The total NO\textsubscript{2} columns in WCB increased from 531 × 10\textsuperscript{15} molec./cm\textsuperscript{2} to 637 × 10\textsuperscript{15} molec./cm\textsuperscript{2}, and the relative increase rate is about 19.9% between the two periods. The total NO\textsubscript{2} columns in other Anhui areas except the WCB area increased from 494 × 10\textsuperscript{15} molec./cm\textsuperscript{2} to 563 × 10\textsuperscript{15} molec./cm\textsuperscript{2}, and the relative increase rate is about 13.9%. It is clear that the total columns in WCB increased more than those in other areas, which may result from the construction of the WCB and the policy of industrial transfer from eastern coastal areas to inland areas. Furthermore, the fraction of tropospheric NO\textsubscript{2} columns in WCB to the total tropospheric NO\textsubscript{2} columns in Anhui is up to 59.3% in 2016, while this value is 56.6% in 2011 (Figure 8). The increased fraction of tropospheric NO\textsubscript{2} columns after the year of 2011 in WCB also reflects the effect of the construction of WCB on the air quality.

Furthermore, the temporal variations of NO\textsubscript{2} columns are studied. The NO\textsubscript{2} columns of Anhui and WCB from 2005 to 2016 are plotted in Figure 9. Fortunately, it is found that the NO\textsubscript{2} columns over WCB and Anhui increased significantly from 2005 to 2011 and then decreased sharply from 2011 to 2016. The statistical significance of all the linear fits in Figure 9 is determined using the correlation coefficient and the correlation is significant at the 0.05 level. The two datasets have different spatial scale representativeness and surface sensitivity, so it is unreasonable to compare the two datasets directly. However, our comparison results prove the accuracy of the OMI-retrieved tropospheric NO\textsubscript{2} columns in the WCB area.
Figure 5: Annual averaged OMI-retrieved NO$_2$ columns ($\times 10^{13}$ molec./cm$^2$) from 2005 to 2016 in Anhui Province.

Figure 6: Annual averaged OMI-retrieved NO$_2$ columns ($\times 10^{13}$ molec./cm$^2$) from 2005 to 2016 of cities in Anhui Province.
was confirmed by using the $t$ test and $F$ test, at the 95% confidence level. The recent decrease trend reflects the impact of emission control measures and policies taken by the government. It is well known that the new Ambient Air Quality Standard has been implemented since 2012. It is a stricter air quality standard than the previous standard, especially for NO$_2$ and fine particles in the atmosphere [39, 40].

### 3.4. Seasonal Variations of Tropospheric NO$_2$ in WCB

Seasonal variations of tropospheric NO$_2$ were analyzed. In Anhui, spring includes March, April, and May, summer comprises June, July, and August, autumn includes September, October, and November, while winter comprises December, January, and February. Figure 10 displays the tropospheric NO$_2$ columns in different seasons during the 12 years. It is apparent that the highest NO$_2$ column occurred in winter, followed by autumn and spring, while summer had the lowest NO$_2$. Also, the seasonal variation shows the same trend during all twelve years. This seasonal trend may be due to the combined effect of the emission source, sink, and weather conditions. Emissions from power plants increase due to domestic heating in winter. In addition, the weather of winter is characterized by lower temperature and more overcast days than that of other seasons, which results in the reduction of the photochemical reaction of NO$_2$ with volatile organic compounds (VOCs) [41].
Figure 11 illustrates the time series of the annual averaged tropospheric NO$_2$ columns for each city in Anhui Province from 2005 to 2016. It can be seen from Figure 11 that Ma’anshan, Bengbu, Huainan, and Chuzhou showed the maximum NO$_2$ level in 2011. Bozhou, Lu’an, Fuyang, Xuancheng, Hefei, Huangshan, Chizhou, and Anqing displayed the maximum NO$_2$ level in 2012. Suzhou, Huaibei, Xuanche, Hefei, Wuhu, and Tongling showed the maximum NO$_2$ level in 2013. As mentioned earlier, the new Ambient Air Quality Standard of China has been implemented since 2012. The effect of Ambient Air Quality Standard often lags behind the policy itself, so some cities in Anhui Province reached their maximum of NO$_2$ columns in 2012 or 2013.

We used the HYSPLIT model to analyze the origins of NO$_2$ and transport pathways of air masses in the typical city of WCB, Ma’anshan city. Ma’anshan city is in the west of Nanjing area and about 40 km from the center of Nanjing city, whereas Nanjing is one of the industrial centers of Yangtze Delta.
We performed the cluster analysis of the 24 h air mass back trajectories starting at 500 m for the full year of 2015. Figure 12 shows five major types of backward trajectory clusters for different seasons in Ma’anshan in 2015. During the spring, summer, and autumn, air masses are mainly from the eastern regions, so the high emissions of NO\textsubscript{2} in the Nanjing area may influence the concentration of atmospheric NO\textsubscript{2} in the Ma’anshan area. In the winter, the prevailing wind is from north (>50%), where the tropospheric NO\textsubscript{2} columns are relatively low. This means that the high level of NO\textsubscript{2} in Ma’anshan in winter is not from the transport but caused by the local emissions. The high level of tropospheric NO\textsubscript{2} in the Ma’anshan area results from the rapid industrial development and the increase of vehicles on the road.

4. Conclusions

Atmospheric nitrogen dioxide plays an important role in tropospheric chemistry and air quality. Satellite observations have great potential for understanding the spatial distributions and temporal variations in atmospheric NO\textsubscript{2} on
Data Availability

The Ozone Monitoring Instrument (OMI) data used to support the findings of this study are available at the Royal Netherlands Meteorological Institute (KNMI) repository (http://www.temis.nl/airpollution/no2col/no2regioomimonth_v2.php). The meteorological data used to support the findings of this study are available at Global Data Assimilation System (GDAS) repository (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1). The surface in situ data used to support the findings of this study are available at the Chinese National Environmental Monitoring Center (CNEMC) repository (http://www.cnemc.cn/).

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors’ Contributions

Yu Xie and Wei Wang conceived, designed, and performed the experiments. Qinglong Wang provided valuable comments in revising the manuscript.

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