Space borne tropospheric nitrogen dioxide (NO_2) observations from 2005-2020 over the Yangtze River Delta (YRD), China: variabilities, implications, and drivers

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Abstract

Nitrogen dioxide (NO_2) is mainly affected by local emission and meteorology rather than long-range transport. Accurate acknowledge of its long-term variabilities and drivers are significant for understanding the evolutions of economic and social development, anthropogenic emission, and the effectiveness of pollution control measures on regional scale. In this study, we quantity the long-term variabilities and the underlying drivers of NO_2 from 2005 to 2020 over the Yangtze River Delta (YRD), one of the most densely populated and highly industrialized city clusters in China, using OMI space borne observations and the multiple linear regression (MLR) model. We have compared the space borne tropospheric results to the surface in-situ data, yielding correlation coefficients of 0.8 to 0.9 over all megacities within the YRD. As a result, the tropospheric NO_2 column measurements can be used as representatives of near-surface conditions, and we thus only use ground-level meteorological data for MLR regression. The inter-annual variabilities of tropospheric NO_2 vertical column densities (NO_2 VCD_{trop}) from 2005 to 2020 over the YRD can be divided into two stages. The first stage was from 2005 to 2011, which showed overall increasing trends with a wide range of (1.91 ± 1.50) to $(6.70 \pm 0.10) \times 10^{14}$ molecules/cm$^2\cdot$yr^{-1} ($p<0.01$) over the YRD. The second stage was from 2011 to 2020, which showed over all decreasing trends of (-6.31 ± 0.71) to $(-11.01 \pm 0.90)\times 10^{14}$ molecules/cm$^2\cdot$yr^{-1} ($p<0.01$) over each of the megacities. The seasonal cycles of NO_2 VCD_{trop} over the YRD are mainly driven by meteorology (81.01% - 83.91%) except during winter when anthropogenic emission contributions are pronounced (16.09% - 18.99%). The inter-annual variabilities of NO_2 VCD_{trop} are mainly driven by anthropogenic emission (69.18% - 81.34%) except for a few years such as 2018 which are partly attributed to meteorology anomalies (39.07% - 91.51%). The increasing trends in NO_2 VCD_{trop} from 2005 to 2011 over the YRD are mainly attributed to high energy consumption associated with rapid economic growth which causes

significant increases in anthropogenic NO_2 emission. The decreasing trends in NO_2 VCD_{trop} from 2011 to 2020 over the YRD are mainly attributed to the stringent clean air measures which either adjust high energy industrial structure toward low energy industrial structure or directly reduce pollutant emissions from different industrial sectors.

Keywords: OMI; nitrogen dioxide; Emissions; Meteorology; Multiple linear regression model

1. Introduction

As a major tropospheric pollutant, nitrogen dioxide (NO_2) not only threatens human health and crop growth but also involves in a series of atmospheric photochemical reactions (Yin et al., 2019;Wang et al., 2011;Geddes et al., 2012). NO_2 is a crucial precursor in the formation of ozone (O_3), particulate matter (PM), acid rain, and photochemical smog in the troposphere (Yin et al., 2021a;Lu et al., 2019a;Lu et al., 2019b;Sun et al., 2018). Since severe NO_2 pollution increases the risk of respiratory disease and is highly associated with mortality (Meng et al., 2021;MacIntyre et al., 2014;Tao et al., 2012), many countries take the NO_2 level as an important pollution indicator of air quality (Xue et al., 2020). The sources of tropospheric NO_2 are mainly from anthropogenic emission through high temperature combustions, like transportation (vehicles, ships, and airplanes) and industrial facilities (petrochemicals and power plants) (Zheng et al., 2018b;Chi et al., 2021;van Geffen et al., 2015). Additional minor sources of NO_2 are attributed to natural emissions from the biogeochemical reactions in soil, volcanic eruption, and lightning (Bond et al., 2001;Zhang et al., 2003;Lu et al., 2021). The dominant sink of tropospheric NO_2 is attributed to a chemical destruction which first converts NO_2 into nitric acid (HNO_3) and peroxyacetyl nitrate (PAN) which then are by dry or wet deposition (Browne et al., 2013). Due to a short lifetime of a few hours, tropospheric NO_2 is heavily affected by local emission and meteorology rather than long-range transport (Kim et al., 2015;Cheng et al., 2012).

Many scientists have used a suite of active and passive observation technologies onboard ground-based, vehicle-based, ship-based, airborne, or space borne platforms to assess the temporal-spatial variabilities of NO_2 and identify their driving forces in different regions around the globe (Richter et al., 2005;Jiang et al., 2018;Liu et al., 2018;Zhang et al., 2021;Schreier et al., 2015;Shaiganfar et al., 2017). Among all observation technologies and platforms, space borne remote sensing observations have their unique features. By validating with ground-based remote sensing or balloon observations, space borne observations can provide global NO_2 dataset with a reasonable accuracy. Typical space borne instruments include the SCIAMACHY, GOME, OMI, and TROPOMI, which have been widely used in scientific investigations of global nitrogen cycle, O_3 formation regime, and regional pollution & transport, quantification of NO_2 emissions from biomass burning regions, megacities, and industrial facilities, and validation of shipborne observations and atmospheric chemical transport models (CTMs) (Richter et al., 2005;Bechle et al., 2013;Boersma et al., 2011;Ghude et al., 2009;Lamsal et al., 2008). Using space borne observations to derive long term trends of NO_2 and their drivers not only provides valuable information for evaluation of regional emissions, but also improves our understanding of atmospheric evolutions. (Richter et al., 2005) first investigated the inter annual variabilities of tropospheric NO_2 vertical column densities (NO_2 VCD_{trop}) from space with GOME and SCIAMACHY observations during 1996-2004. (Richter et al., 2005) found substantial reductions in NO_2 VCDs over some areas of Europe and the USA, but a highly significant increase of about 50%—with an accelerating trend in annual growth rate—

over the industrial areas of China. In a subsequent study, (Ghude et al., 2009) found the same phenomenon as those of (Richter et al., 2005) with GOME and SCIAMACHY observations from 1996 to 2006, which disclosed that NO_2 VCD_{trop} showed increasing trends over the rapidly developing regions (China: 11 ± 2.6%/year, South Asia: 1.76 ± 1.1%/year, Middle East Africa: 2.3 ± 1 %/year) and decreasing or level-off trends over the developed regions (US: -2 ± 1.5%/year, Europe: 0.9 ± 2.1%/year). With multiple satellite platforms including GOME, SCIAMACHY, OMI, and GOME-2, (Hilboll et al., 2013) also found 5% to 10% yr^{-1} of increasing trends for NO_2 VCD_{trop} over eastern Asia during 1996 to 2011 . With the OMI observations, (Lamsal et al., 2015) have quantified the NO_2 trend from 2005 to 2013 over the US and (Krotkov et al., 2016) have investigated the NO_2 trends over different countries for the period of 2005–2014.

Along with the great advances in social and economic development in recent decades, air quality in China has changed dramatically (Sun et al., 2020;Sun et al., 2021c;Yin et al., 2020;Yin et al., 2021c;Yin et al., 2021d). China has implemented a series of clean air measures in different stages to tackle air pollution across China. One of the landmark clean air measures could be the Action Plan on the Prevention and Control of Air Pollution implemented in 2013, which launched many stringent measures to improve air quality across China. These measures include the reduction of air pollutant emissions, the adjustment of industrial structure and energy mix, the establishment of early-warning systems and monitoring for air pollution, and other compulsive policies (China State Council, 2013). Both space borne and ground-based observations have witnessed the effectiveness of these successful policies. The OMI NO_2 VCD_{trop} have been decreased by 21% from 2011 to 2015 over 48 cities of China (Liu et al., 2017). The national averaged surface NO_2 recorded by the China National Environmental Monitoring Center (CNEMC) network has significantly decreased from (16.68 ± 4.82) ppbv in 2013 to (11.29 ± 3.25) ppbv in 2020 (Lin et al., 2021).

In this study, we use NO_2 VCD_{trop} from 2005-2020 provided by OMI to comprehensively evaluate the long-term trends, implications, and underlying drivers of NO_2 over the Yangtze River Delta (YRD, including Anhui, Jiangsu, Shanghai, and Zhejiang Provinces, Table S1). In addition to anthropogenic emission, meteorology also drives NO_2 variability by affecting emissions, transport, chemical production, and scavenging. The relationships of NO_2 against meteorological variables are complex and are region and time dependent. In present work, we separate the contributions of meteorology and anthropogenic emission to the NO_2 variability by multiple linear regression (MLR) model over the major cities (Hefei, Nanjing, Suzhou, Shanghai, Hangzhou, Ningbo) within the YRD. As one of the three most densely populated and highly industrialized city clusters in China, the YRD has long been identified as a key region for air pollution mitigation. This study can not only improve our understanding of temporal spatial NO_2 evolutions in the atmosphere but also provides valuable information for future clean air policy. We introduce detailed descriptions of OMI and ground-level NO_2 products in section 2.1, and meteorological fields in section 2.2. The method for separating contributions of meteorology and anthropogenic emission is presented in section 2.3. Sections 3.1 and 3.2 analyze the temporal-spatial variabilities of tropospheric NO_2 from 2005 to 2020 over the YRD on provincial and megacity levels, respectively. A comparison between the OMI NO_2 product and the ground-level measurements is performed in section 3.3. We discuss the implications and underlying drivers of the variabilities of tropospheric NO_2 from 2005 to 2020 over the YRD in section 4. We conclude this study in section 5.

2. Data and method

2.1 Observation data

2.1.1 OMI NO_2 product

OMI is a hyperspectral atmospheric composition detection instrument onboard the National Aeronautics and Space Administration (NASA) Aura Earth Observing System (EOS) satellite launched in July, 2004 (Boersma et al., 2007). The EOS satellite flies over a low-Earth orbit at an altitude of about 710 km. The local overpass time (LT) of OMI satellite is about 13:30 in early afternoon. The retrieval micro window for NO_2 VCDs lies in between 405 nm and 465 nm with a spectral resolution of about 0.5nm (Marchenko et al., 2015). The spatial resolution of OMI measurements is 13 × 24 km^2 at nadir. OMI provides observations of O_3, NO_2, SO_2, aerosol, cloud, HCHO, BrO, and OClO with nearly daily global coverage (Levelt et al., 2006). The daily LV3 data product of NO_2 VCD_{trop} data (GES DISC; http://disc.sci.gsfc.nasa.gov, last accessed: 1 September 2021) which is a gridded data with a 0.25° × 0.25° spatial resolution are used in this study. The NO_2 VCD_{trop} are calculated by Stratosphere–troposphere separation (STS) scheme proposed by numerous previous studies (Bucsela et al., 2013;Lamsal et al., 2014;Goldberg et al., 2017). The STS scheme first subtract the stratospheric NO_2 slant column densities (SCDs) from the total NO_2 SCDs and then it divides the resulting tropospheric NO_2 SCDs by the tropospheric air mass factor (AMF). The formulation for calculating NO_2 VCD_{trop} is as follow:

$$VCD_{trop} = \frac{SCD_{total} - SCD_{strat}}{AMF_{trop}} \tag{1}$$

where AMF is defined as the ratio of the SCD to the VCD (Solomon et al., 1987),

$$AMF_{trop} = \frac{SCD_{trop}}{VCD_{trop}} \tag{2}$$

The tropospheric AMF are calculated by NO_2 profiles simulated by the Global Modeling Initiative (GMI) chemistry transport model with the horizontal resolution of 1° × 1.25° (Rotman et al., 2001). Separation of stratospheric and tropospheric columns is achieved by the local analysis of the stratospheric field over unpolluted areas (Bucsela et al., 2013). The OMI NO_2 VCD_{trop} dataset has been used in many studies to investigate O_3 formation regime and regional pollution & transport (Lin et al., 2010;Zhang et al., 2017;Duncan et al., 2013;Liu et al., 2016). In this study, only the LV3 data product collected with cloud radiance fractions of less than 30% is used (Streets et al., 2013).

2.1.2 Ground level NO_2 data

We extract ground level NO_2 data over the YRD from the China National Environmental Monitoring Center (CNEMC) network (http://www.cnemc.cn/en/, last access: November 26, 2021). The CNEMC network has operated more than 3000 monitoring sites that almost cover all major cities over China by 2020. The CNEMC datasets have been used in many studies for evaluation of regional atmospheric pollution & transport (Li et al., 2021;Lu et al., 2019a;Lu et al., 2020;Sun et al., 2021a;Yin et al., 2021a;Zhao et al., 2016;He et al., 2017). As one of the six key atmospheric pollutants (CO, SO_2, NO_2, PM_{10}, O_3, and $PM_{2.5}$) routinely measured by the CNEMC network, ground level NO_2 measurements at 188 sites in 40 cities over the YRD are available since 2014. In this study, comparisons between the OMI NO_2 data product and the ground level NO_2 measurements are only performed over 6 key megacities, i.e., Shanghai, Nanjing, Hangzhou, Suzhou, Ningbo, and Hefei, within the YRD. The population, geolocation, the number of measurement site, and data range of each city are summarized in Table 1. The number of measurement site in each city ranges from 8 to 11, the altitude ranges from 3 to 50 m (above sea level, a.s.l.), and the population ranges from 0.9 to 2.5 million. All ground level NO_2 data at each station are measured by active differential absorption ultraviolet (UV) analyzers. We use a data quality control method following previous studies to remove unreliable NO_2 data (Lu et al., 2019a;Lu et al., 2020;Sun et al., 2021a;Yin et al., 2021a). Specifically, we first convert all hourly measurements into *Z* scores, we then remove the measurement if its Z score meets one of the following rules: (1) Z_i is larger or smaller than the previous value Z_{i-1} by 9 ($|Z_i - Z_{i-1}| > 9$); (2) The absolute value of Z_i is greater than 4 ($|Z_i| > 4$); (3) the ratio of the *Z* value to the third-order center moving average is greater than 2 ($\frac{3Z_i}{Z_{i-1}+Z_i+Z_{i+1}} > 2$), where *i* represents the i^{th} hourly measurement data. After removing OUTLIERS with above filter criteria, we finally average NO_2 data at all measurement sites in each city to form a city representative NO_2 dataset.

2.2 Meteorological fields

We obtain meteorological fields during 2005-2020 from the second Modern-Era Retrospective analysis for Research and Applications (MERRA-2) (Gelaro et al., 2017). This dataset is produced by the NASA Global Modeling and Assimilation Office (https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/, last accessed: 1 August, 2021) with a spatial resolution of 0.5° × 0.625°, temporal resolutions of 1 h for boundary layer height and surface meteorological variables, and 3 h for other variables. Previous studies have verified that meteorological fields provided by MERRA-2 match well with the meteorological parameters observed by Chinese weather stations (Song et al., 2018;Carvalho, 2019;Wang et al., 2017;Kishore Kumar et al., 2015;Zhou et al., 2017). In order to match OMI observations which are available at about 13:30 LT, the average for meteorological data is only performed between 13:00 and 14:00 LT.

2.3 Multiple linear regression (MLR) model

We establish a multiple linear regression (MLR) model to quantify the contributions of meteorology and anthropogenic emission to the long-term variabilities of NO_2 VCD_{trop} during 2005-2020 over the YRD. Similar MLR methodologies have been used in previous studies to estimate the contributions of meteorology and emission to the variabilities of O_3 and $PM_{2.5}$ in North America, Europe and China (Li et al., 2019;Li et al., 2020;Xu et al., 2011;Zhai et al., 2019;Zhao and Wang, 2017). The meteorological parameters used in our MLR model are elaborated in Table 2.

In order to highlight the variabilities of NO_2 VCD_{trop}, we follow the method of previous studies and calculate NO_2 VCD_{trop} anomalies ($\boldsymbol{y}_{anomaly}$) by subtracting a reference value ($\boldsymbol{y}_{reference}$) from all tropospheric NO_2 observations ($\boldsymbol{y}_{individual}$) (Hakkarainen et al., 2016;Hakkarainen et al., 2019;Mustafa et al., 2021). The formulation of this method is expressed as:

$$\boldsymbol{y}_{anomaly} = \boldsymbol{y}_{individual} - \boldsymbol{y}_{reference} \quad (3)$$

In this study, we take the average of all NO_2 VCD_{trop} from 2005 to 2020 (i.e., the 16-year mean) as the reference value. The MLR model for each city is explained as:

$$\boldsymbol{y} = \beta_0 + \sum_{k=1}^{11} \beta_k \boldsymbol{x}_k \quad (4)$$

where $\boldsymbol{y}$ are the regression result for monthly OMI NO_2 VCD_{trop} anomalies, β_0 is the intercept, and $\boldsymbol{x}_k$ ($k \in [1, 11]$) are the meteorological variables. The regression coefficients β_k are calculated by nonlinear least squares fitting. This MLR model finds the optimal regression result by minimizing the sum of squares of the fitting residual and then solves regression coefficients β_k by the following equation:

$$\beta_k = (\sum \boldsymbol{x}_k \boldsymbol{x}_k^T)^{-1}(\sum \boldsymbol{x}_k \boldsymbol{y}_k) \tag{5}$$

The regression results $\boldsymbol{y}$ represent the meteorology induced contributions to the variabilities of NO_2 VCD_{trop}. Since both soil and lighting NO_x are meteorology dependent, the effects of soil and lighting NO_x on NO_2 variability are also attributed to meteorology contribution. The difference $\boldsymbol{y}'$ between the monthly OMI NO_2 VCD_{trop} anomalies $\boldsymbol{y}_{anomaly}$ and $\boldsymbol{y}$ calculated as equation (6) represents the portion that cannot be explicitly explained by the meteorological influence.

$$\boldsymbol{y}' = \boldsymbol{y}_{anomaly} - \boldsymbol{y} \tag{6}$$

By subtracting the meteorological influence from the total NO_2 amounts, the $\boldsymbol{y}'$ is referred to as the aggregate contribution of anthropogenic emission. Positive $\boldsymbol{y}$ and $\boldsymbol{y}'$ indicate that meteorology and anthropogenic emission cause NO_2 VCD_{trop} above the reference value (i.e., the 16-year mean), respectively. In contrast, negative $\boldsymbol{y}$ and $\boldsymbol{y}'$ indicate that meteorology and anthropogenic emission cause NO_2 VCD_{trop} below the reference value, respectively.

Since the meteorological parameters listed in Table 2 differ in units and magnitudes, which could lead to unstable performance of the model. Therefore, we normalized all meteorological parameters via equation (7) before using them in regression. This normalization pre-processing procedure can also speed up the convergence of the MLR model.

$$\boldsymbol{z}_k = \frac{\boldsymbol{x}_k - \boldsymbol{u}_k}{\boldsymbol{\sigma}_k} \tag{7}$$

where $\boldsymbol{u}_k$ and $\boldsymbol{\sigma}_k$ are the average and 1σ standard deviation (STD) of $\boldsymbol{x}_k$, and $\boldsymbol{z}_k$ is the normalized value for parameter $\boldsymbol{x}_k$.

3. Temporal-spatial variabilities of NO_2 VCD_{trop} over the Yangtze River Delta

3.1 Variabilities at provincial level

We present the temporal-spatial distribution of the annual averaged NO_2 VCD_{trop} over the YRD from 2005 to 2020 in Figure 1. The major pollution areas for NO_2 VCD_{trop} over the YRD are located in the south of Jiangsu Province and north of Zhejiang Province. In addition, NO_2 pollution in eastern Anhui Province showed an increasing trend during 2005-2013 and became one of the major pollution areas within YRD during 2010-2013. The amplitudes of NO_2 VCD_{trop} over the YRD showed large year to year variabilities from 2005 to 2020 but spatial extensions of the major pollution areas are almost constant over years. Among all the pollution areas, the heaviest pollution regions are uniformly located in the densely populated and highly industrialized megacities such as Shanghai, Nanjing, Suzhou, Hangzhou, Ningbo, and Hefei.

The annual means and seasonal cycles of NO_2 VCD_{trop} over the YRD during 2005-2020 at Province or municipality level, i.e., Anhui Province, Jiangsu Province, Zhejiang Province, and Shanghai municipality, are presented in Figure 2. The NO_2 VCD_{trop} over each province are calculated by averaging all observations within the boundary of each province. For seasonal variability, clear seasonal features over the whole YRD region and each province are observed (Figure 2a): (1) high levels of NO_2 VCD_{trop} occur in late winter to spring and low levels of NO_2 VCD_{trop} occur in later summer to autumn; (2) the 1σ STDs in late winter to spring are larger than those in later summer to autumn; and (3) seasonal cycles of NO_2 VCD_{trop} over Jiangsu, Zhejiang and the whole YRD region show bimodal patterns, i.e., two seasonal peaks occur around March and December or January, and one seasonal trough occurs around September; but these over Anhui shows a unimodal pattern and don't have the peak around March. The NO_2 VCD_{trop} present a maximum monthly mean value of (1.93 ± 0.21), (2.40 ± 0.25), (1.61 ± 0.16), and (1.91 ± 0.16) × 10^{16} molecules/cm^2 in January or December over Anhui, Jiangsu, Zhejiang, and the whole YRD region, respectively. The minimum monthly mean values over Anhui, Jiangsu, Zhejiang and the whole YRD region occur in July, with values of (0.35 ± 0.05), (0.83 ± 0.07), (0.57 ± 0.06), and (0.39 ± 0.01) × 10^{16} molecules/cm^2, respectively.

Except for a few anomalies such as the year-to-year decrease in 2005-2006, and the increases in 2016-2017 and 2018-2019, the overall inter annual variabilities of NO_2 VCD_{trop} over the YRD can be divided into two stages (Fig. 2b). The first stage was from 2005 to 2011, which showed overall increasing trends in NO_2 VCD_{trop} over the YRD. During 2005 to 2009 of this stage, change rates of NO_2 VCD_{trop} were less pronounced, where the 2009 relative to 2005 levels have only increased by (0.33 ± 0.02)×10^{15} (3.96 ± 0.25) %, (1.05 ± 0.11)×10^{15} (8.55 ± 0.08) %, and (0.46 ± 0.03)×10^{15} molecule/m^2 (5.05 ± 0.32) % over Anhui, Jiangsu and the whole YRD region, respectively, and leveled off over Zhejiang. However, NO_2 VCD_{trop} in 2011 relative to 2009 showed significantly increments of (2.88 ± 0.23)×10^{15} (33.78 ± 2.70) %, (3.81 ± 0.32)×10^{15} (29.01 ± 2.45) %, (2.08 ± 0.18)×10^{15} (27.97 ± 2.43) %, (2.10 ± 0.19)×10^{15} molecule/m^2 (21.59 ± 1.95) % over Anhui, Jiangsu, Zhejiang and the whole YRD region, respectively. The second stage was from 2011 to 2020, which showed overall decreasing trends in NO_2 VCD_{trop} over the YRD. The total decrements over Anhui, Jiangsu, Zhejiang and the whole YRD region in 2020 relative to 2011 are (4.91 ± 0.39)×10^{15} (41.48 ± 3.30) %, (4.82 ± 0.31)×10^{15} (43.25 ± 2.72) %, (3.78 ± 0.36)×10^{15} (40.47 ± 4.12) %, (4.82 ± 0.35)×10^{15} molecule/m^2 (43.26 ± 3.07) %, respectively.

We have followed the methodology of (Li et al., 2020)) and used the linear regression model to estimate the inter annual trends of NO_2 VCD_{trop} over the YRD (Table 3). During 2005-2011, inter annual trends of NO_2 VCD_{trop} over the YRD region and each province spanned a wide range of (1.74 ± 0.72) ×10^{14} molecules/cm^2·yr^{-1} (p=0.02) to (5.94 ± 1.01)×10^{14} molecules/cm^2·yr^{-1} (p<0.01), indicating a regional representative of each dataset. In contrast, inter annual trends of NO_2 VCD_{trop} over the YRD region and each province from 2011 to 2020 varied over (-4.86 ± 0.49) to (-8.16 ± 0.82) ×10^{14} molecules/cm^2·yr^{-1} (p<0.01). For the aggregate trends during 2005-2020, NO_2 VCD_{trop} over the whole YRD region and each province are negative. The largest and lowest decreasing trends are observed in Jiangsu and Anhui, with values of (-1.92 ± 0.30)×10^{14} molecules/cm^2·yr^{-1} (p<0.01) and (-0.92 ± 0.26)×10^{14} molecules/cm^2·yr^{-1} (p<0.01), respectively.

3.2 Variabilities at megacity level

The annual means and seasonal cycles of NO_2 VCD_{trop} over the major megacities within YRD during 2005-2020 are presented in Figure 3. Similar to the derivation of provincial level NO_2, NO_2 VCD_{trop} over each megacity are calculated by averaging all observations within the boundary of each megacity. The results show that the amplitudes and variabilities of NO_2 VCD_{trop} at megacity level are basically coincident with those at the corresponding provincial levels. Overall, the amplitudes and 1σ STDs of NO_2 seasonal cycles in cold seasons are larger than those in warm seasons, and the inter annual NO_2 variabilities at megacity level can also be divided into two stages, i.e., an overall increasing stage during 2005-2011 and a decreasing stage during 2011-2020. As a result, it is feasible to select these major megacities as representatives for mapping the drivers of NO_2 variabilities over the YRD.

Specifically, megacity level of NO_2 VCD_{trop} show seasonal maxima in December and seasonal minima in July. Seasonal maxima over Hefei, Shanghai, Nanjing, Suzhou, Hangzhou, and Ningbo are (2.03 ± 0.15), (2.80 ± 0.23), (2.62 ± 0.25), (2.66 ± 0.16), (1.83 ± 0.18), and (2.27 ± 0.21)×10^{16} molecules/cm^2, and seasonal minima are (0.34 ± 0.04), (0.83 ± 0.11), (0.58 ± 0.06), (0.62 ± 0.05), (0.32 ± 0.02), and (0.38 ± 0.03)×10^{16} molecules/cm^2, respectively. The seasonal maxima are on average (82.27 ± 2.34) %, (67.19 ± 1.56) %, (71.06 ± 2.32) %, (83.33 ± 3.05) %, (77.62 ± 2.89) %, and (70.84 ± 2.76) % higher than the seasonal minima over respective megacity. As commonly observed, the seasonal variability of NO_2 VCD_{trop} with respect to their annual means spanned a wide range of −55.1% to 103.5% depending on season and measurement time (Figure 3a).

The NO_2 VCD_{trop} in all megacities show the maximum values in 2011, where the maximum values over Hefei, Shanghai, Suzhou, Ningbo, Nanjing and Hangzhou are (1.41 ± 0.25), (2.18 ± 0.23), (1.81 ± 0.17), (1.39 ± 0.12), (1.88 ± 0.18) and (1.19 ± 0.14)×10^{16} molecules/cm^2, respectively (Figure 3b). In terms of the increments relative to the 2005 levels, Hefei and Shanghai from 2005 to 2011 have the largest and lowest increments of (5.37 ± 0.51)×10^{15} molecules/cm^2 (61.77 ± 5.87) % and (2.62 ± 0.27)×10^{15} molecules /cm^2 (14.68 ± 1.51) %, respectively. The increments over other cities varied over (3.31 ± 0.32)×10^{15} molecules /cm^2 (31.20 ± 3.02) % to (5.21 ± 0.41)×10^{15} molecules/cm^2 (38.40 ± 3.02) %. In terms of the decrements relative to the 2011 levels, Shanghai and Hangzhou from 2011 to 2020 have the largest and lowest decrements of (9.77 ± 0.82)×10^{15} molecules/cm^2 (46.89 ± 3.94) and (5.28 ± 0.45)×10^{15} molecules/cm^2 (45.43 ± 3.87) %, respectively. The decrements over other cities are also evident and varied over (6.33 ± 0.58)×10^{15} molecules/cm^2 (45.53 ± 4.18) % to (9.05 ± 0.98)×10^{15} molecules/cm^2 (48.12 ± 5.21) %. A few anomalies are also observed in some megacities and are in good agreement with the corresponding provincial levels. For example, NO_2 VCD_{trop} over Hefei and Suzhou had increased by (0.09 ± 0.01)×10^{15} molecules/cm^2 (0.77 ± 0.09) % and (0.80 ± 0.07)×10^{15} molecules/cm^2 (4.90 ± 0.43) % in 2013 relative to 2012 levels, respectively. In addition, NO_2 VCD_{trop} over Hefei, Shanghai, Nanjing, Hangzhou, and Suzhou had increased by (0.65 ± 0.12)×10^{15} (8.41 ± 1.55) %, (0.35 ± 0.02)×10^{15} (2.66 ± 0.15) %, (0.86 ± 0.18)×10^{15} (8.16 ± 1.71) %, (0.55 ± 0.08)×10^{15} (8.68 ± 1.26) %, and (0.29 ± 0.05)×10^{15} molecules/cm^2 (2.52 ± 0.43) % in 2019 relative to 2018 levels, respectively.

The inter annual trends of NO_2 VCD_{trop} during 2005-2011 over all cities are positive and span a wide range of (1.91 ± 1.50) to (6.70 ± 0.10)×10^{14} molecules/cm^2·yr^{-1} ($p<0.01$) (Table 4). In contrast, the inter annual trends of NO_2 VCD_{trop} during 2011-2020 over all cities are negative. The largest and lowest decreasing trends are observed in Nanjing and Hangzhou, with values of (-11.01 ± 0.90) and (-6.31 ± 0.71)×10^{14} molecules/cm^2·yr^{-1} ($p<0.01$), respectively. For the aggregate trends during 2005-2020, NO_2 VCD_{trop} over all cities are negative. The largest and lowest decreasing trends are observed in Shanghai and Hefei, with values of (-4.58 ± 0.43)×10^{14} molecules/cm^2·yr^{-1} ($p<0.01$) and (-0.30 ± 3.43)×10^{14} molecules/cm^2·yr^{-1} ($p=0.385$), respectively.

3.3 Comparisons with the CNMEC data

In order to investigate if satellite column measurements can represent the near surface variabilities, we have compared the OMI NO_2 VCD_{trop} data over the 6 megacities within the YRD with the ground level NO_2 data provided by the CNMEC (Figure 4). The comparisons over all megacities were performed on monthly basis between June 2014 and December 2020. Ground level NO_2 concentrations were taken as the average of all CNMEC stations in each city. The NO_2 VCD_{trop} values were taken as the average of all OMI observed grids within the scope of each city. Considering the overpass time of OMI is at about 13:30 LT, we only average the ground level NO_2 data between 13:00 and 14:00 LT for comparison, which ensures that the temporal differences between the CNMEC and OMI dataset are all within ± 30 minutes. With these rules, there are over 700 matching samples in each city available for comparison.

Correlation plots of OMI NO_2 VCD_{trop} data against the CNMEC ground level NO_2 measurements are shown in Figure 4. The results show that the NO_2 variabilities observed by OMI and the CNMEC are in good agreement over all megacities, with correlation coefficients (r^2) of 0.88, 0.81, 0.89, 0.88, 0.86 and 0.83 for Hangzhou, Hefei, Nanjing, Ningbo, Shanghai, and Suzhou, respectively. The discrepancies between OMI and CNMEC data can be mainly attributed to their differences in temporal-spatial resolutions. OMI averages NO_2 concentration at about 13:30 LT over a large coverage due to its relatively coarse spatial resolution (Wallace and Kanaroglou, 2009;Zheng et al., 2014). The CNMEC data represent the averaged point concentrations between 13:00 and 14:00 LT around the measurement site. NO_2 is a short lifetime species and is characterized by large temporal-spatial variabilities. Any temporal-spatial inhomogeneity in NO_2 concentration could affect the comparison (Meng et al., 2010;Wallace and Kanaroglou, 2009). Considering above differences, the correlations of the two datasets over all megacities are satisfactory. The tropospheric NO_2 column measurements can be used as representatives of near-surface conditions. As a result, to simplify calculations, we only use ground-level meteorological data for MLR regression.

Over polluted atmosphere, the NO_2 column measurements can be used as representative of near-surface conditions because tropospheric NO_2 has a vertical distribution that is heavily weighted toward the surface (Kharol et al., 2015;Zhang et al., 2017;Duncan et al., 2016;Duncan et al., 2013;Kramer et al., 2008). Many studies have taken advantage of this favourable vertical distribution of NO_2 to derive surface emissions of NO_2 from space (Silvern et al., 2019;Boersma et al., 2009;Streets et al., 2013;Anand and Monks, 2017;Lu et al., 2015;Ghude et al., 2013;Cooper et al., 2020). Meanwhile, the use of NO_2 column measurements to explore tropospheric O_3 sensitivities has been the subject of several past studies, which disclosed that this diagnosis of O_3 production rate (PO_3) is consistent with the findings of surface photochemistry (Jin et al., 2017;Jin and Holloway, 2015;Sun et al., 2018;Yin et al., 2021b;Souri et al., 2017;Sun et al., 2021b;Jin et al., 2020;Choi and Souri, 2015;Schroeder et al., 2017;Baruah et al., 2021).

4 Implications and drivers

We incorporate the 11 meteorological parameters listed in Table 2 into the MLR model to fit the time series of monthly averaged NO_2 VCD_{trop} from 2005 to 2020 over the 6 megacities within the YRD (Figure S1). Correlation plots of the MLR regression results and the satellite tropospheric NO_2 data are shown in Figure 5. The results show that the MLR model can well reproduce the seasonal variabilities of tropospheric NO_2 VCDs over each city with correlation coefficients of 0.85 to 0.90. We separate the contributions of meteorology and anthropogenic emission to the NO_2 variability over the 6 megacities with the methodology described in section 2.3. Figure 6 shows monthly averaged tropospheric NO_2 VCDs along with the meteorological-driven contributions and the anthropogenic-driven contributions in each city. Figure 7 is the same as Figure 6, but the statistics are based on annual average.

4.1 Drivers of seasonal cycles of NO_2 VCD_{trop}

As shown in Figure 6 for all megacities, the seasonal variabilities of meteorological contributions are consistent with those of NO_2 VCD_{trop} except the period from February to March, and the anthropogenic contributions varied around zero throughout the year except in December and February. This means that the seasonal variabilities of tropospheric NO_2 over the YRD are mainly determined by meteorology (81.01% - 83.91%) and also influenced by anthropogenic emission in December and February. Meteorological contributions are larger than zero in winter and lower than zero in summer, indicating that meteorology increases NO_2 level in winter and decreases NO_2 level in summer. This contrast in meteorological contribution is associated with the seasonal cycle of temperature. Similarly, anthropogenic contributions are larger than zero in December and lower than zero in February, representing anthropogenic emission increases NO_2 level in December and decreases NO_2 level in February. The enhanced anthropogenic contributions in December are mainly attributed to more extensive anthropogenic activities such as residential heating in megacities in this period which usually results in more anthropogenic NO_2 emission due to the increase in energy and fuel consumptions. The decreased anthropogenic contributions in February are attributed to the Spring Festival. We elaborate the analysis as below.

As shown in Figure S2, the vast majorities of meteorological contributions over all megacities are from temperature and additional minor contributions over some cities such as Nanjing, Shanghai, and Suzhou are attributed to relative humidity, pressure, or surface incoming shortwave flux (SWGDN) (Agudelo–Castaneda et al., 2014;Parra et al., 2009). Significant negative correlations between temperature and NO_2 VCD_{trop} are observed in all megacities (Figure S3, Table 5). Higher temperature tends to decrease NO_2 VCD_{trop} and vice versa. This is because higher temperature conditions could accelerate the chemical reaction that destructs NO_2 in the troposphere (Pearce et al., 2011;Yin et al., 2021a). In addition, surface pressure shows high positive and both surface relative humidity and SWGDN show negative correlations with NO_2 VCD_{trop}, but their contribution levels are much lower than the temperature. All other meteorological variables only have weak correlations with NO_2 VCD_{trop} (Table 5).

In all cities except Hefei, there is a significant increase in NO_2 level from February to March. The maximum and minimum increments occur in Shanghai and Nanjing, with values of $(3.28 \pm 0.29)\times10^{15}$ molecules/cm^2 (16.37 ± 1.45) % and $(0.47 \pm 0.05)\times10^{15}$ molecules/cm^2 (2.60 ± 0.28) %, respectively. In contrast, the meteorological contributions show decreased change rates in the same period. As a result, this increase in NO_2 level from February to March could be attributed to anthropogenic emission rather than meteorology. Indeed, anthropogenic contributions show significant increases of (3.95 ± 0.32) to $(6.53 \pm 0.55)\times10^{15}$ molecules/cm^2 over all megacities from February to March. The most important festival in China-the Spring Festival-typically occurs in February, when a large number of migrants in megacities return to their hometowns for holiday and most industrial productions are shut down, which could cause significant reductions in anthropogenic emission. In March, these migrants get back to work and all industrial enterprises resumed productions, which could cause a rebound in anthropogenic emission. The seasonal maxima of NO_2 in March are not observed in Hefei is because the anthropogenic emission induced increases are offset by meteorology induced decreases.

2020 is a special year compared to all other years, when a large-scale lockdown occurred in February and some regional travel restrictions occasionally occurred in other seasons across China due to COVID-19 disease. In the comparison, we removed all NO_2 measurements in 2020 to eliminate the influence of COVID-19. The monthly averaged NO_2 VCD_{trop} from 2005 to 2019 along with the meteorological contributions and the anthropogenic contributions in each city are shown in Figure S4. Figure S5 and Figure S6 are the same as Figure 2 and Figure 3, respectively, but for 2011 to 2019. We obtained the same conclusion as that from Figure 6, indicating the drivers of seasonal cycles of NO_2 VCD_{trop} deduced above are consistent over years.

4.2 Drivers of inter annual variabilities of NO_2 VCD_{trop}

As shown in Figure 7 for all megacities, the inter annual variabilities of anthropogenic contributions are in good agreement with those of NO_2 VCD_{trop}, indicating inter annual variabilities of NO_2 VCD_{trop} are mainly driven by anthropogenic emission. The same as those of NO_2 VCD_{trop}, the inter annual anthropogenic contributions over each city can also be divided into two stages, i.e., an overall increasing stage during 2005–2011 and a decreasing stage during 2011-2020. For the first stage (2005-2011), anthropogenic contributions account for 84.72%, 92.96%, 93.52%, 79.06%, 97.12%, and 90.21% of the increases in NO_2 VCD_{trop}, while meteorological contributions account for 15.28%, 7.04%, 6.48%, 20.94%, 2.88%, and 9.79% over Hangzhou, Hefei, Nanjing, Ningbo, Shanghai, and Suzhou, respectively. The annual averaged meteorological contributions over each city varied around zero in all years except few anomalies in some years. For example, meteorological contributions over all cities are larger than zero in 2005 and 2011 but lower than zero after 2014. Pronounced anomalies include the enhancements occurred in 2011 in all cities and the decrements in 2015 over Suzhou, in 2018 over Hangzhou, and in 2016 over other cities. All these anomalies in meteorological contributions are highly correlated with temperature anomalies (Figure S7). As shown in Figure S8 and S9, the temperature in all cities is lower than the reference value (i.e., the 16-year mean) in 2005 and 2011 and larger than the reference value after 2014. As a result, in addition to anthropogenic emission, the NO_2 enhancements in 2011 are partly attributed to the lower temperature in this year. Meanwhile, higher temperature in YRD region in recent years favors the decrease in NO_2 VCD_{trop}. For the second stage (2011-2020), anthropogenic contributions account for 70.15 %, 65.22 %, 66.97 %, 73.45 %, 74.43 %, and 73.84 % of the decreases in NO_2 VCD_{trop}, while meteorological contributions account for 29.85%, 34.78%, 33.03 %, 26.55 %, 25.57 %, and 26.16 % over Hangzhou, Hefei, Nanjing, Ningbo, Shanghai, and Suzhou, respectively.

Since anthropogenic NO_2 emissions are highly related to economic and industrial activities (Lin and McElroy, 2011;Russell et al., 2012;Vrekoussis et al., 2013;Guerriero et al., 2016), to understand the inter annual variabilities of NO_2 VCD_{trop}, we have investigated the inter annual variabilities of Gross Domestic Product (GDP) over the YRD from primary sector, secondary sector and tertiary sector (http://www.stats.gov.cn/, last accessed: 1 August, 2021) from 2005 to 2020. The primary sector includes agriculture, forestry, animal husbandry, and fishery; The secondary industry includes mining, manufacturing, power, heat, gas and water production and supply, and construction; The tertiary industry, namely the service industry, refers to all industries excluded the primary industry and the secondary industry. The secondary industry is more related to energy and fuel consumptions, and it thus dominates the anthropogenic NO_2 emission. Figure S10 shows the time series of GDP over the YRD from 2005 to 2020 and Figure S11 is the same as Figure S10 but for year-to-year increment, i.e., the increase in GDP at a given year relative to its previous year. The results show that the GDP of each province within the YRD increased over time starting from 2005 but the relative contribution of each industry sector is different from year to year. The primary sector-related GDP is relatively constant, but both the secondary sector and tertiary sector related GDPs show significant increasing trends from 2005 to 2020.

During 2009 to 2011, the GDPs have increased significantly by 198.45, 483.86, 656.40, and 327.05 billion yuan over Shanghai, Zhejiang, Jiangsu, and Anhui, where the secondary sector contributions account for 46.50%, 53.64%, 48.99%, and 60.34% respectively. Before 2011, much of China's economic growths still rely on the high-carbon fossil energy system and efforts to control atmospheric pollution were relatively small. These significant increases in GDP could cause significant increases in anthropogenic NO_2 emission. After 2011, China has implemented a series of clean air measures to tackle air pollution across China. These measures include the reduction of industrial pollutant emissions, the adjustment of industrial structure and energy mix, and other compulsive policies (China State Council, 2013). (Zheng et al., 2018a) have estimated China's anthropogenic emission trends from 2010 to 2017 with the bottom-up emission inventory. (Zheng et al., 2018a) found that, as the consequence of clean air measures, anthropogenic NO_x emission across China during 2010–2017 have been decreased by 17%. In Figure S12, we further analyzed the variabilities of NO_x emissions over the YRD region from 2008 to 2017 by category provided by the Multi-resolution Emission Inventory for China (MEIC) inventory, including motor vehicle emissions, major industrial emissions, resident emissions and power emissions (http://meicmodel.org, last accessed: February 25, 2022) (Li et al., 2017;Zheng et al., 2018a). The results show that the decreases in Tro_NO_2 over the YRD during 2011 to 2013 are attributed to the reductions of industrial and power emissions, during 2013 to 2014 are mainly attributed to the reductions of motor vehicle emissions and power emissions, and after 2014 are attributed to the reductions of motor vehicle emissions, power emissions and industrial emissions.

Although the total GDPs over all megacities are still increasing over time after 2011, much of these increases are from the tertiary sector, indicating the effectiveness of the adjustment of industrial structure and energy mix. The largest anthropogenic NO_2 producer from the tertiary sector is attributed to the transportation industry including such as traffic and cargo transport, etc. Chinese government had implemented stringent restrictions on vehicle exhaust emissions after 2011 (Ministry of Ecology and Environment of the People's Republic of China, 2016, 2011). For example, Chinese government implemented the fourth and the fifth national motor vehicle pollutant emissions standards in 2011 and 2018, respectively, which mandate 30% and 60% reductions in vehicle NO_x emissions relative to the third national standard (Ministry of Ecology and Environment of the People's Republic of China, 2007, 2018). These stringent measures could significantly reduce anthropogenic NO_2 emission from the tertiary sector. Overall, the decreasing trends in NO_2 VCD_{trop} from 2011 to 2020 over all megacities within the YRD are mainly attributed to the stringent clean air measures in this period which either adjust high energy industrial structure toward low energy industrial structure or directly reduce pollutant emissions from different industrial sectors.

5 Conclusions

In this study, we have quantified the long-term variabilities and the underlying drivers of NO_2 VCD_{trop} from 2005-2020 over the Yangtze River Delta (YRD) by OMI LV3 NO_2 data product and MLR regressions. The major pollution areas for NO_2 VCD_{trop} over the YRD are located in the south of Jiangsu Province and north of Zhejiang Province. In addition, NO_2 pollution in eastern Anhui Province showed an increasing trend during 2005-2013 and became one of the major pollution areas within YRD during 2010-2013. The amplitudes of NO_2 VCD_{trop} over the YRD showed large year to year variabilities from 2005 to 2020 but spatial extensions of the major pollution areas are almost constant over years. Among all the pollution areas, the heaviest pollution regions are uniformly located in the densely populated and highly industrialized megacities such as Shanghai, Nanjing, Suzhou, Hangzhou, Ningbo, and Hefei. For six megacities the space borne tropospheric results have been compared to surface in-situ data, yielding correlation coefficients between 0.8 and 0.9.

Clear seasonal features and inter annual variabilities of NO_2 VCD_{trop} over the YRD region are observed. Overall, the amplitudes and 1σ STDs of NO_2 seasonal cycles in cold seasons are larger than those in warm seasons, and the inter annual NO_2 variabilities at megacity level can be divided into two stages, i.e., an overall increasing stage during 2005-2011 and a decreasing stage during 2011-2020. We have used the MLR regressions to quantify the drivers of NO_2 VCD_{trop} from 2005 to 2020 over all megacities within the YRD. The seasonal cycles of NO_2 VCD_{trop} over the YRD are mainly driven by meteorology (81.01% - 83.91%) except in winter when anthropogenic emission contributions are also pronounced (16.09% - 18.99%). The inter annual variabilities of NO_2 VCD_{trop} are mainly driven by anthropogenic emission (69.18% - 81.34%) except in few years such as 2018 which are partly attributed to meteorology anomalies (39.07% - 91.51%).

The increasing trends in NO_2 VCD_{trop} from 2005 to 2011 over the YRD are mainly attributed to high energy consumption associated with rapid economic growth which cause significant increases in anthropogenic NO_2 emission. The decreasing trends in NO_2 VCD_{trop} from 2011 to 2020 over the YRD are mainly attributed to the stringent clean air measures in this period which either adjust high energy industrial structure toward low energy industrial structure or directly reduce pollutant emissions from different industrial sectors. This study can not only have improved our knowledge with respect to long term evolutions of economic and social development, anthropogenic emission, and the effectiveness of pollution control measures over the YRD, but also have positive implications for forming future clean air policies in the important region.

Code and data availability. Surface NO_2 measurements over the YRD are from http://www.cnemc.cn/en/. The OMI LV3 tropospheric NO_2 satellite data can be obtained from https://acdisc.gesdisc.eosdis.nasa.gov/data/Aura_OMI_Level3/. The Chinese economic data can be obtained from http://www.stats.gov.cn/. All other data are available on request of the corresponding author (Youwen Sun, ywsun@aiofm.ac.cn).

Author contributions. HY designed the study and wrote the paper. YS supervised and revised this paper. JN, MP, and CL provided constructive comments.

Competing interests. None.

Acknowledgements. This work is jointly supported by the National Key Research and Development Program of China (No.2019YFC0214802), the Youth Innovation Promotion Association, CAS (No.2019434), and the Sino-German Mobility programme (M-0036).

Table 1. Geolocation, the number of measurement site, and population for the 6 megacities within the YRD. Population statistics are based on the seventh nationwide population census in 2020 provided by National Bureau of Statistics of China.

City	Latitude	Longitude	Number of sites	Altitude (m)	Population (million)
Hangzhou	30.29	120.15	11	41.7	1.19
Hefei	31.85	117.25	10	29.8	0.94
Ningbo	29.87	121.55	9	5.1	0.94
Nanjing	32.04	118.77	9	8.9	0.93
Shanghai	31.23	121.47	10	4.5	2.49
Suzhou	31.30	120.62	8	3.5	1.28

Table 2. Meteorological parameters used in the MLR model.

Parameters	**Description**	**Unit**
T_{2m}	2m air temperature	°C
U_{10m}	10m zonal wind	m/s
V_{10m}	10m meridional wind	m/s
PBLH	Planetary boundary layer height	m
TCC	Total cloud area fraction	unitless
Rain	Rainfall	kg·m^2/s
SLP	Sea level pressure	Pa
SWGDN	Surface incoming shortwave flux	W/m^2
RH_{2m}	2m Relative humidity	%
TROPH	Tropospheric layer Height	m

Table 3. Inter annual trends of NO_2 VCD_{trop} over each province within the YRD and the whole YRD region during 2005 to 2011, 2011 to 2020 and 2005 to 2020.

Province	**Annual trend (10^{14} molecule/m^2)**		
	2005-2011	**2011-2020**	**2005-2020**
YRD	3.69 ± 0.78 ($p<0.01$)	-6.18 ± 0.52 ($p<0.01$)	-1.54 ± 0.23 ($p<0.01$)
Anhui	4.40 ± 0.89 ($p<0.01$)	-5.93 ± 0.58 ($p<0.01$)	-0.92 ± 0.26 ($p<0.01$)
Jiangsu	5.94 ± 1.01 ($p<0.01$)	-8.16 ± 0.82 ($p<0.01$)	-1.92 ± 0.30 ($p<0.01$)
Zhejiang	1.74 ± 0.72 ($p=0.02$)	-4.86 ± 0.49 ($p<0.01$)	-1.41 ± 0.22 ($p<0.01$)

Table 4. Inter annual trends of NO_2 VCD_{trop} over each city within the YRD during 2005 to 2011, 2011 to 2020 and 2005 to 2020.

Province	**Annual trend (10^{14} molecule/m^2)**		
	2005-2011	**2011-2020**	**2005-2020**
Hangzhou	4.07 ± 1.03 ($p<0.01$)	-6.31 ± 0.71 ($p<0.01$)	-1.41 ± 0.30 ($p<0.01$)
Hefei	6.70 ± 0.11 ($p<0.01$)	-6.73 ± 0.78 ($p<0.01$)	-0.30 ± 3.43 ($p=0.385$)
Nanjing	6.50 ± 1.25 ($p<0.01$)	-11.01 ± 0.90 ($p<0.01$)	-2.19 ± 0.39 ($p<0.01$)
Ningbo	3.79 ± 1.16 ($p<0.01$)	-7.16 ± 0.81 ($p<0.01$)	-2.51 ± 0.35 ($p<0.01$)
Shanghai	1.91 ± 1.50 ($p=0.204$)	-9.91 ± 0.97 ($p<0.01$)	-4.58 ± 0.43 ($p<0.01$)
Suzhou	5.84 ± 0.12 ($p<0.01$)	-7.16 ± 0.81 ($p<0.01$)	-2.32 ± 0.35 ($p<0.01$)

Table 5. Correlations of monthly averaged observations against each meteorological parameter from 2005 to 2020.

City	Correlations									
	T_{2m}	U_{10m}	V_{10m}	PBLH	TCC	Rain	SLP	SWGDN	RH_{2m}	TROPH
Hangzhou	-0.81	-0.11	-0.40	-0.43	-0.63	-0.34	0.84	-0.51	-0.78	0.28
Hefei	-0.84	0.02	-0.48	-0.51	-0.57	-0.39	0.83	-0.69	-0.77	0.25
Nanjing	-0.86	0.07	-0.47	-0.45	-0.56	-0.59	0.86	-0.63	-0.83	0.38
Ningbo	-0.84	0.39	-0.71	-0.14	-0.70	-0.47	0.86	-0.54	-0.82	0.07
Shanghai	-0.82	0.59	-0.65	0.08	-0.66	-0.45	0.83	-0.56	-0.83	0.32
Suzhou	-0.87	0.35	-0.59	-0.60	-0.67	-0.59	0.87	-0.72	-0.82	0.45

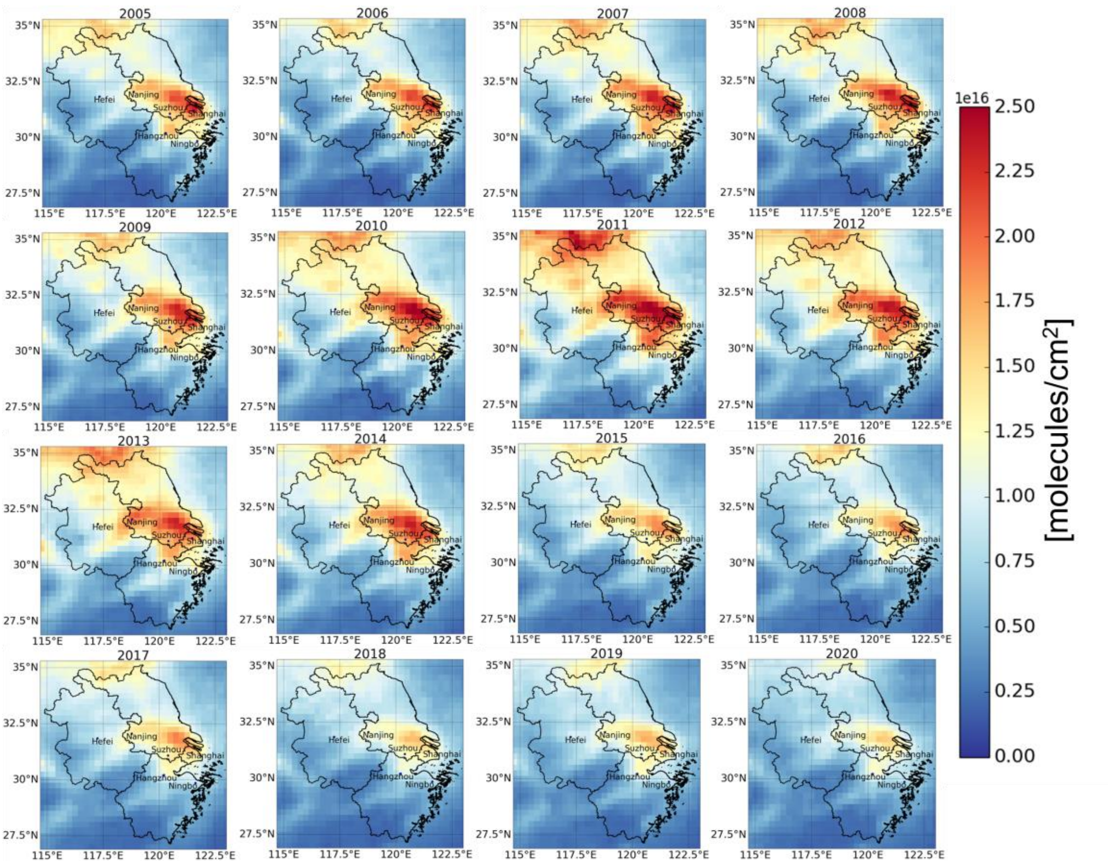

Figure 1. Temporal-spatial variabilities of NO_2 VCD_{trop} provided by OMI satellite over the YRD from 2005 to 2020. The three provinces (Anhui, Jiangsu, Zhejiang) and six key megacities (Hefei, Nanjing, Suzhou, Shanghai, Hangzhou, Ningbo) are marked.

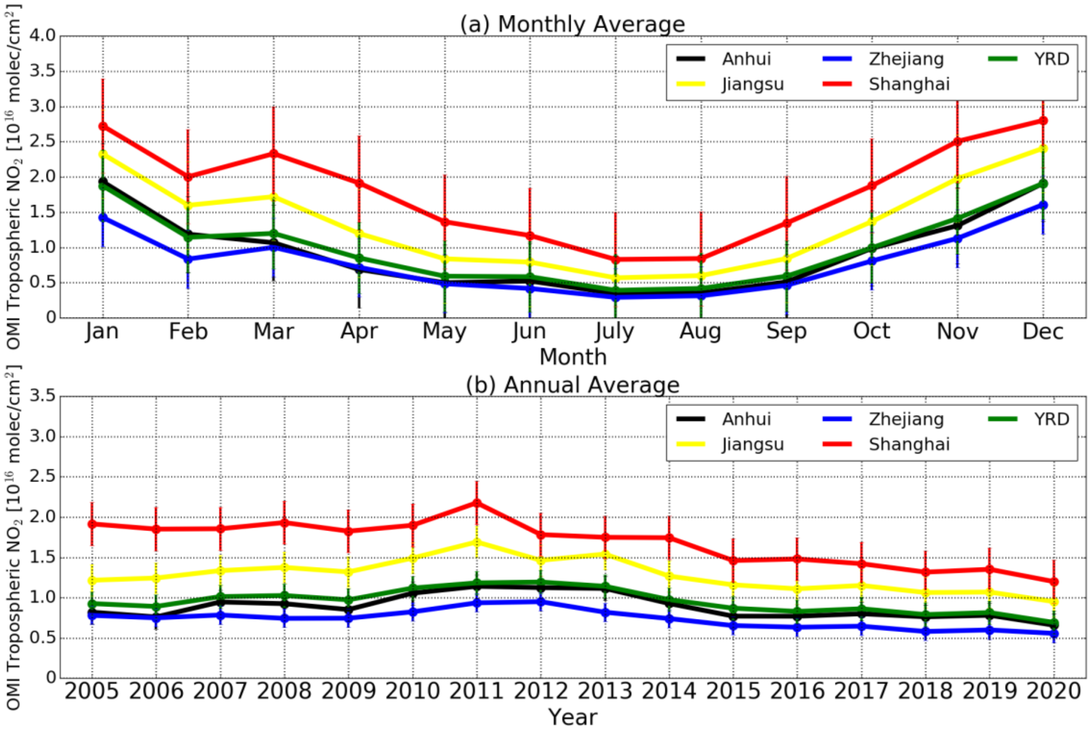

Figure 2. (a) Monthly averaged NO_2 VCD_{trop} over the whole YRD region (green dots and lines), Anhui Province (black dots and lines), Zhejiang Province (blue dots and lines), and Jiangsu Province (yellow dots and lines). (b) Same as (a) but for annual average. The vertical error bar is 1σ standard variation (STD) within that month or year.

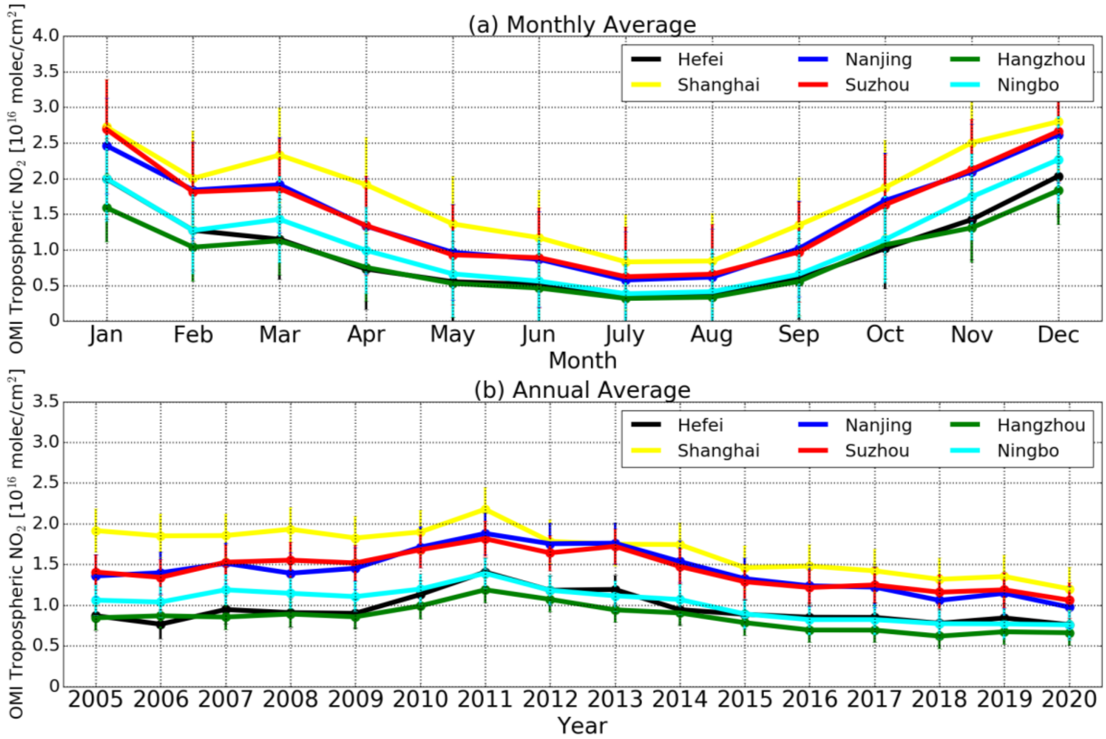

Figure 3. (a) Monthly averaged NO_2 VCD_{trop} over Hefei (black dots and lines), Nanjing (blue dots and lines), Shanghai (yellow dots and lines), Suzhou (red dots and lines), Hangzhou (green dots and lines), and Ningbo (cyan dots and lines). (b) Same as (a) but for annual average. The vertical error bar is 1σ standard variation within that month or year.

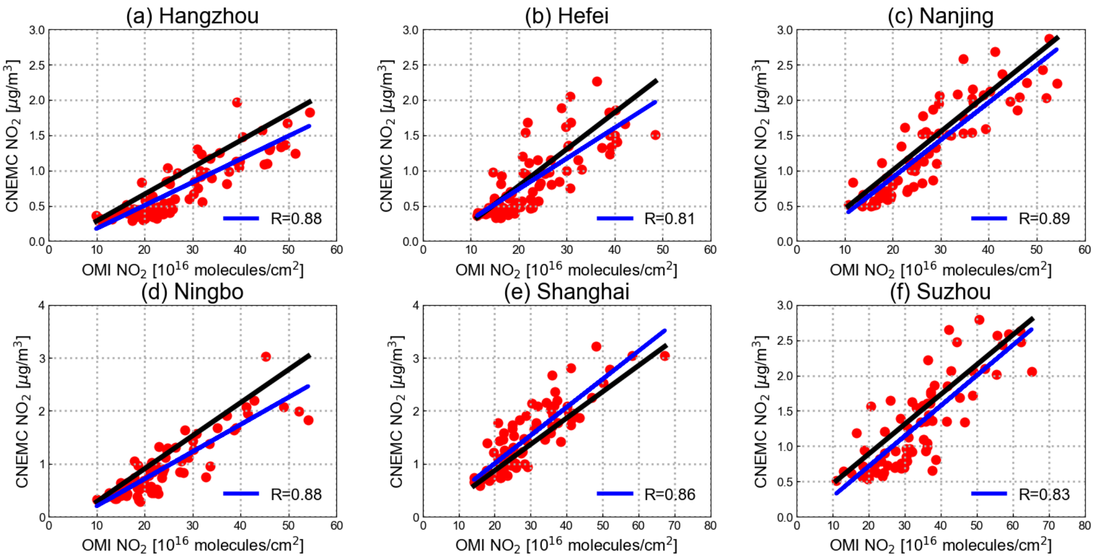

Figure 4. Correlation of OMI NO_2 VCD_{trop} against ground-level observations data over Hefei, Nanjing, Shanghai, Suzhou, Hangzhou and Ningbo. We fitted both datasets directly without uniform their units, which does not affect the investigation with respect to the agreement of the two datasets in terms of variabilities. Blue lines are linear fitted lines and black lines are one to one line.

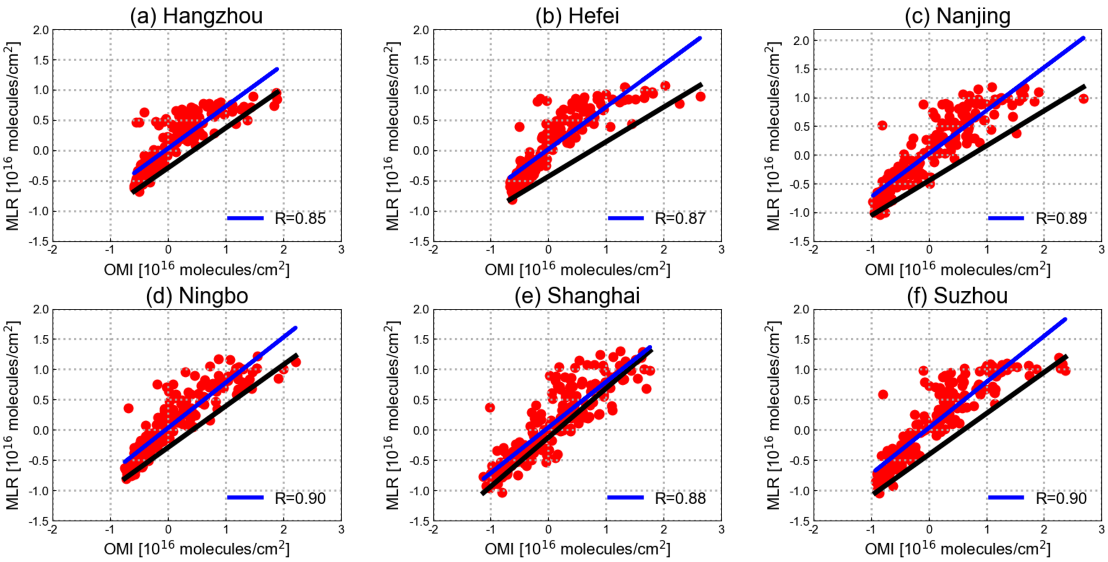

Figure 5. Correlations of OMI NO_2 VCD_{trop} against the MLR model results over Hefei, Nanjing, Shanghai, Suzhou, Hangzhou, and Ningbo. Blue lines are linear fitted lines and black lines are one to one line.

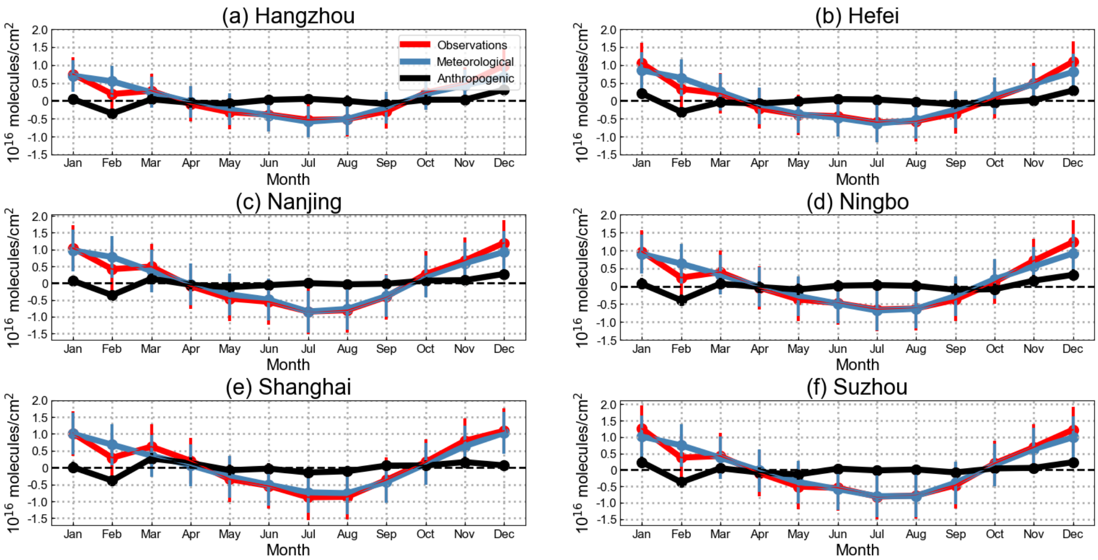

Figure 6. Monthly averaged NO_2 VCD_{trop} (red dots and lines) along with the meteorological-driven portions (blue dots and lines) and the anthropogenic-driven portions (black dots and lines) over each city within the YRD. The vertical error bar is 1σ standard variation (STD) within that month.

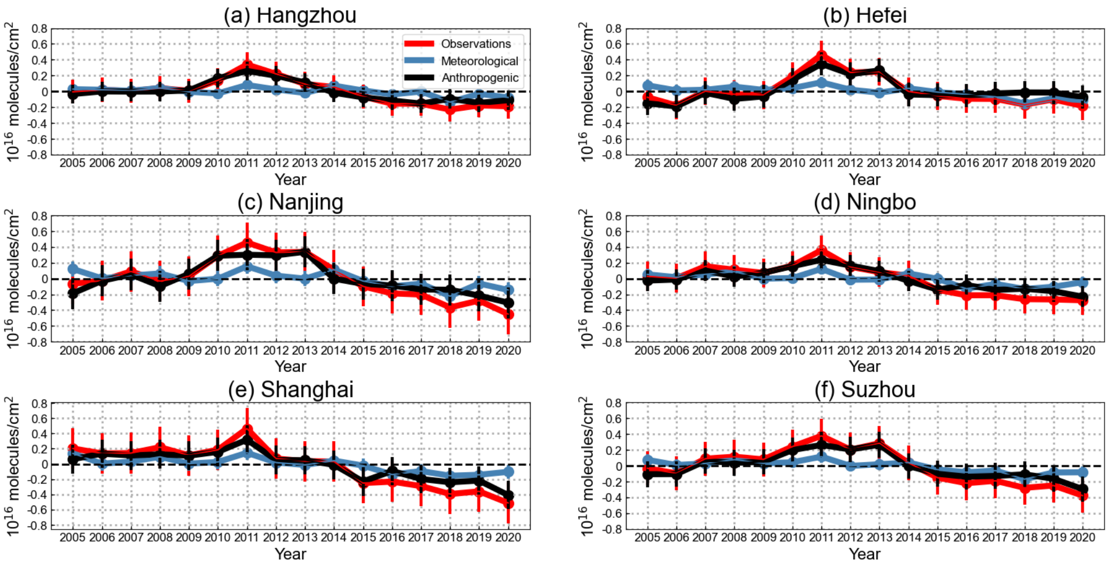

Figure 7. The same as Figure 6 but for annual average.

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