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SPATIO-TEMPORAL CHARACTERISTICS OF NO₂ IN PRD URBAN GROUP AND THE ANTHROPOGENIC INFLUENCES ANALYSIS BASED ON OMI REMOTE SENSING DATA

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Abstract: Spatio-temporal distribution characteristics and variation trends of tropospheric NO₂ in Pearl River Delta (PRD) urban group and its adjacent areas were analyzed from 2005 to 2013 based on remote sensing data from ozone monitoring instrument (OMI) satellite, and further explored the impact of human activities on NO₂. Compared with the ground observation data, the OMI NO₂ remote sensing data displayed high reliability. Due to active industrial production, high car ownership, great energy and power consumption, the average tropospheric NO₂ concentration (7.4×10^{15} molec/cm²) of PRD region is about 3 times of the adjacent areas. At the same time, the regional high pollution NO₂ in PRD region as a whole, the urban group effect is remarkable. Sinusoidal model can well fit the periodic variation of the NO₂ in PRD and adjacent areas. NO₂ concentration was highest in winter while lowest in summer. The concentration of NO₂ in PRD region is decreasing in recent 9 years, which has a significantly negative correlation with the second industry output and car ownership. This suggests that the nitrogen oxide emissions governance in PRD region had achieved initial results. The concentration of NO₂ increased significantly in the eastern and northern Guangdong Province, there are good positive correlations with the second industrial outputs and car ownerships, it is thus clear that industrial emissions and automobile exhausts are important sources of NO₂ in these regions. The concentration of NO₂ in western Guangdong area is stable.

Key words: Pearl River Delta urban group; NO₂; spatial and temporal distribution; anthropogenic influences

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1 INTRODUCTION

NO_x (including NO and NO₂) is the main pollutant gas in the troposphere and is directly harmful to the human body. Moreover, NO_x is an important precursor of O₃ and other secondary photochemical pollutants (such as PAN and secondary aerosol) (Crutzen^[1]), and is one of the important pollution sources forming acid rain, acid fog and photochemical smog (Ji et al.^[2]; Ding et al.^[3]; Wang et al.^[4]). The sources of NO_x in the

troposphere include artificial sources and natural sources. The artificial sources mainly refer to fossil fuel combustion (including industry, automobiles, airplanes, ships etc.) and biomass burning. Under natural conditions, the emission sources of NO_x are mainly lightning, ammonia (NH₃) oxidation, soil discharge and stratospheric transport (Lee et al.^[5]). The NO_x from human activities exceeds half of the total emissions of NO_x, especially in urban areas, where most of the NO_x emissions are from human activities (Beirle et al.^[6]). NO₂ is one of the important factors affecting climate change. The global average radiative forcing of the nitrate aerosol produced by NO₂ in the upper atmosphere is (-0.15 ± 0.19) W/m² (Stocker et al.^[7]). Therefore, acquiring the spatio-temporal distribution characteristics of NO₂ and analyzing the impact of human activities on them has an important significance.

Currently there are two ways to monitor NO₂: surface observation and satellite remote sensing. The former can acquire the all-weather NO₂ concentration information with higher precision, but it can only be conducted at limited surface stations and it is difficult to acquire the spatial distribution characteristics of NO₂ within a large range. Satellite remote sensing is widely adopted due to its advantages of wide observation and

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coverage area, good data uniformity and low cost. The satellite remote sensing data of NO_2 in the troposphere mainly come from the Global Ozone Monitoring Experiment (GOME/GOME-2), the SCanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY) and the Ozone Monitoring Instrument (OMI). Based on the satellite data, many scholars have analyzed the spatio-temporal distribution characteristics of NO_2 in the troposphere (Beirle et al.^[8]; Richter and Burrows^[9]; Zhang et al.^[10]; Zhang et al.^[11]; Wang et al.^[12]; Yu et al.^[13]; Wei et al.^[14]; Xiao et al.^[15]; Yao et al.^[16]; Li et al.^[17]), analyzed and verified the spatial allocation weight and the total amount of emissions^[6, 18-21] of NO_x emission sources, and studied the impacts of human activity on NO_2 concentration distribution and change tendencies^[10, 16, 17, 20-23].

Since China's reform and opening-up, fossil energy consumption has been steadily increasing as China's economy and society rapidly develop, and a great quantity of polluting gases (including SO_2 , NO_2 etc.) have been emitted into the atmosphere, causing long-term high tropospheric NO_2 concentrations in China urban areas which are rapidly increasing^[10, 21, 24]. As China's urbanization level increases and the size of its urban areas grows larger, inter-city industry relations and economic cooperation continues to consolidate. Three urban groups (the Beijing-Tianjin-Hebei Region, Yangtze River Delta and Pearl River Delta) have come into being, and new urban groups have also emerged. China's urban groups are rapidly developing, and the proportion of China's economy that they represent is growing greater and greater, making them the engines of China's economy. However the high-density population concentration and the high-intensity economic activities in urban groups far exceed the bearing capacity of the regional ecological environments, causing the regional environment and air quality to deteriorate sharply. As a consequence, highly-contaminated areas are formed and the haze weather becomes more frequent (Wu et al.^[25, 26]; Zhang et al.^[27]). The spatio-temporal distribution characteristics and change tendencies of NO_2 from urban groups have attracted the concern of government departments and the whole society, however research related to urban groups is still rare.

By taking the urban group in the Pearl River Delta (PRD) as an example, using OMI remote sensing data of tropospheric NO_2 vertical column as a basis, and performing comparative validation on the reliability of the satellite remote sensing data with surface observation data, this work carried out a statistical analysis on the spatio-temporal distribution characteristics and variation trends of the tropospheric NO_2 vertical column in the PRD from 2005 to 2013, and analyzed the impact of human activities on NO_2 concentration distribution and changes during that period. As a comparison, the paper studied the

spatio-temporal distribution of NO_2 and the impact of human activities in eastern Guangdong, northern Guangdong (N-GD) and western Guangdong (W-GD) which neighbor the PRD.

2 DATA AND METHODS

The OMI (Ozone Monitoring Instrument) carried by the AURA satellite was jointly developed by Holland, Finland and National Aeronautics and Space Administration (NASA). It has three detecting channels with the wavelength range of 270-500 nm (UV-1: 270-310 nm, UV-2: 310-365 nm, VIS: 365-500 nm), a spectral resolution of 0.42 nm (UV-1), 0.45 nm (UV-2) and 0.63 nm (VIS), a scanning width of 2600 km, a sub-satellite point resolution of 13×12 km (UV-2 and VIS channel) and 13×24 km (UV-1 channel). The OMI covers the entire earth once each day (Levelt and Noordhoek^[28]). Through data inversion, the OMI remote sensing data can help to acquire the concentration of multiple reactive trace gases throughout the world, such as O_3 , NO_2 , SO_2 and HCHO. Among these, the concentration of the tropospheric NO_2 vertical column (NO_2 molecular number concentration from the ground to the tropopause along the vertical direction per unit area) was obtained (Chance^[29]) through the inversion of Differential Optical Absorption Spectroscopy (DOAS) technology on the basis of the spectral information of the wave band 405-465 nm. In the current study, the monthly average value data of the tropospheric NO_2 vertical column acquired by OMI from 2005 to 2013 were used, and the spatial resolution of the data was $0.125^\circ \times 0.125^\circ$. The data product originated from the Koninklijk Nederlands Meteorologisch Instituut (KNMI, Royal Netherlands Meteorological Institute). Since its reversion accuracy is on a higher level ($\pm 0.7 \times (10^{15} \text{ molec/cm}^2)$) (Boersma^[30]), the data has been widely used in analytical study^[6, 13, 18-20, 23] on the spatio-temporal distribution characteristics and pollution sources of NO_2 .

Since 2004, the Guangdong Meteorological Service has gradually established a network of atmospheric composition observation stations in the PRD. Among these the main station is the Guangzhou Panyu Atmospheric Composition Observation Station of the China Meteorological Administration (GPACOS, station number: 59481), located in Dazhengangshan, Nancun Town, Panyu District, Guangzhou (its position is shown in the following Fig.1). The observation range of the main station covers the physical and chemical characteristics of aerosol and reactant gas characteristics. These observation data have been widely used in local atmospheric physics and atmospheric environment research^[25, 31-36]. The nitrogen oxide gas analyzer (EC9841) uses the chemiluminescence method to measure the surface NO_2 volume-ratio concentration value (unit: ppbv). The NO_2 surface observation data of the main station from 2007 to 2012 are selected and

processed them into monthly average data, then compared them to the satellite remote sensing NO₂ data in the local grids. It should be noted that the satellite remote sensing inversion can obtain the total amount of the NO₂ column (molecular number), but the data from EC9841 is the surface NO₂ concentration (volume ratio); these are not the same amount. When the vertical distribution weight function of NO₂ concentration remains unchanged, both have a linear relation. In areas with serious NO₂ pollution, the high NO₂ concentration value is mainly concentrated at the ground layer, and the weight of the ground layer in the vertical distribution is higher; at this point both amounts are comparable; therefore comparison and inspection can be performed between both amounts (Lin and Xu^[37]).

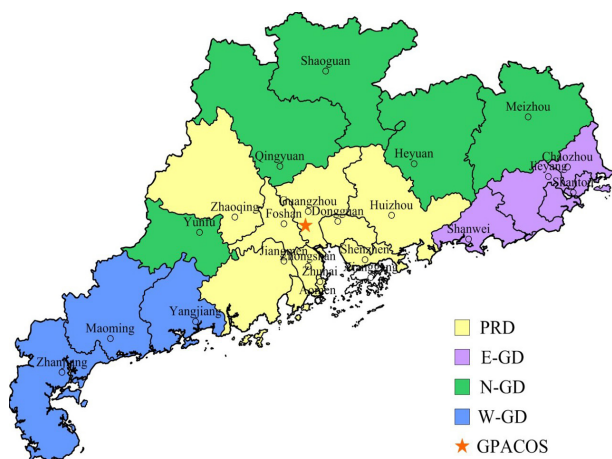


Figure 1. Regional division of Guangdong Province and location of GPACOS.

According to the historical *Guangdong Statistical Yearbook* issued by the Statistics Bureau of Guangdong Province, Guangdong Province is divided into four regions, namely the PRD, the eastern Guangdong region (E-GD, east wing), the northern Guangdong region (N-GD, mountainous area) and the western Guangdong region (W-GD, west wing) (see Fig. 1). Among these the PRD covers the following nine cities: Guangzhou, Shenzhen, Zhuhai, Foshan, Huizhou, Dongguan, Zhongshan, Jiangmen and Zhaoqing. The E-GD region covers the following four cities: Shantou, Shanwei, Chaozhou and Jieyang. The N-GD region covers the following five cities: Shaoguan, Heyuan, Meizhou, Qingyuan and Yunfu. The W-GD region covers the three following cities: Yangjiang, Zhanjiang and Maoming.

In order to study the impact of human activities on NO₂ concentration distributions and variation trends, this study adopted all social and economic development data from Guangdong province in 2012, and data related to the regional GDP, production output value of secondary industry and the amount of civilian vehicles from 2005 to 2012. All these data originated from the 2006-2013 Guangdong Statistical Yearbooks published by the

Statistics Bureau of Guangdong Province.

3 RESULTS AND ANALYSIS

3.1 Contrast test on satellite remote sensing data and surface observation data

A comparative analysis was performed on the monthly average value of NO₂ observed by the GPACOS and the monthly average tropospheric NO₂ column value acquired by the OMI from 2007 to 2012, the relation between them is shown in Fig.2 (the relevant coefficient is expressed as R). Comparison results show that the tropospheric NO₂ column concentration acquired by OMI and the surface NO₂ observation have good consistency, and shows significant correlation ($P < 0.01$), with a correlation coefficient of about 0.73. Fig. 3 provides the monthly average value over the 6 year (2007-2012) of the two datasets. It can be seen that the variation trends of the monthly average NO₂ concentration of the two datasets are almost consistent for these data over many years; the high NO₂ concentration values occurred in the winter, and low in summer. Therefore, the OMI remote sensing data of the tropospheric NO₂ column concentration used in this study has high reliability and can reveal the distribution and change characteristics of surface NO₂ concentration.

3.2 Spatio-temporal distribution characteristics

The average value of the tropospheric NO₂ column concentration data acquired by the OMI in the PRD and its surrounding areas (E-GD, N-GD and W-GD) from 2005 to 2013 is shown in Fig.4. According to the figure, the NO₂ column concentration value in the PRD was far higher than that of its adjacent areas; the average concentration value was 7.4×10^{15} molec/cm², which was approximately 3 times of that of the adjacent areas. The high NO₂ pollution areas in the PRD (with a concentration value exceeding 10×10^{15} molec/cm²) were mainly distributed in the downstream flat areas of the Pearl River, which is the exactly area in which the PRD urban group located. Among these, the central urban areas of Guangzhou and Foshan had the highest values throughout the whole province of Guangdong, and the NO₂ column concentration was 20×10^{15} molec/cm². Eight cities of the PRD region except Huizhou had high NO₂ pollution areas, and these high pollution areas are connected together and form a high pollution belt. This shows that the atmospheric pollution situation in the PRD is very severe, the atmospheric pollution in one city affects other cities, and the urban group effect is very pronounced. In the E-GD region, the average NO₂ column concentration was about 2.5×10^{15} molec/cm², and the high NO₂ column concentration of 4.5×10^{15} molec/cm² was distributed in the central urban area of Shantou. In the N-GD region, the average NO₂ column concentration was a bit higher than that of the eastern Guangdong region at about 2.8×10^{15} molec/cm², and the high NO₂ pollution areas were mainly distributed in the

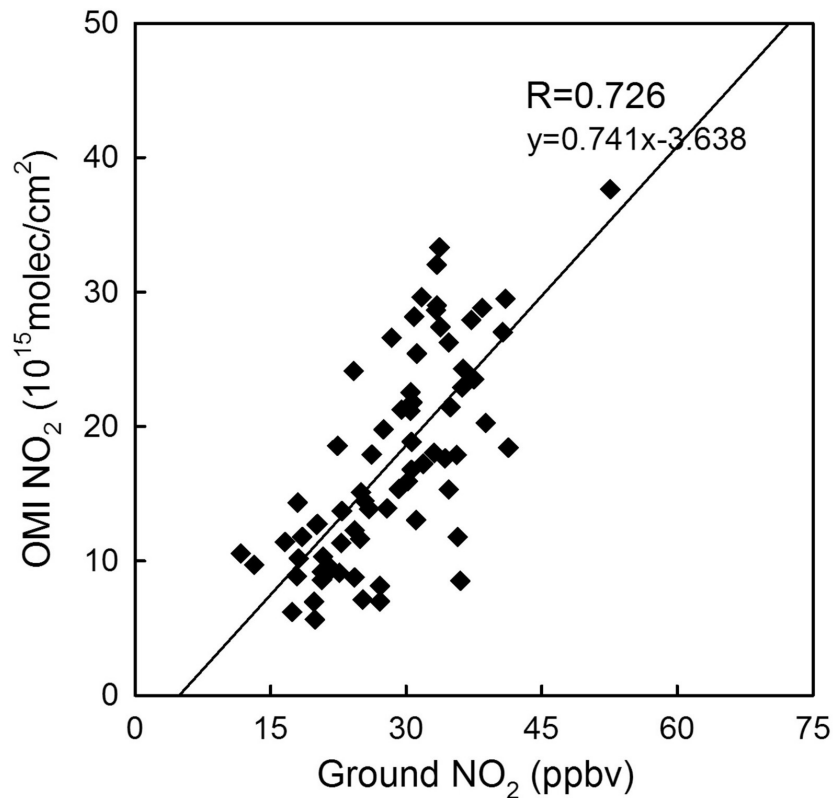


Figure 2. Comparison of average monthly NO₂ values from surface observation data and satellite remote sensing data 2007-2012.

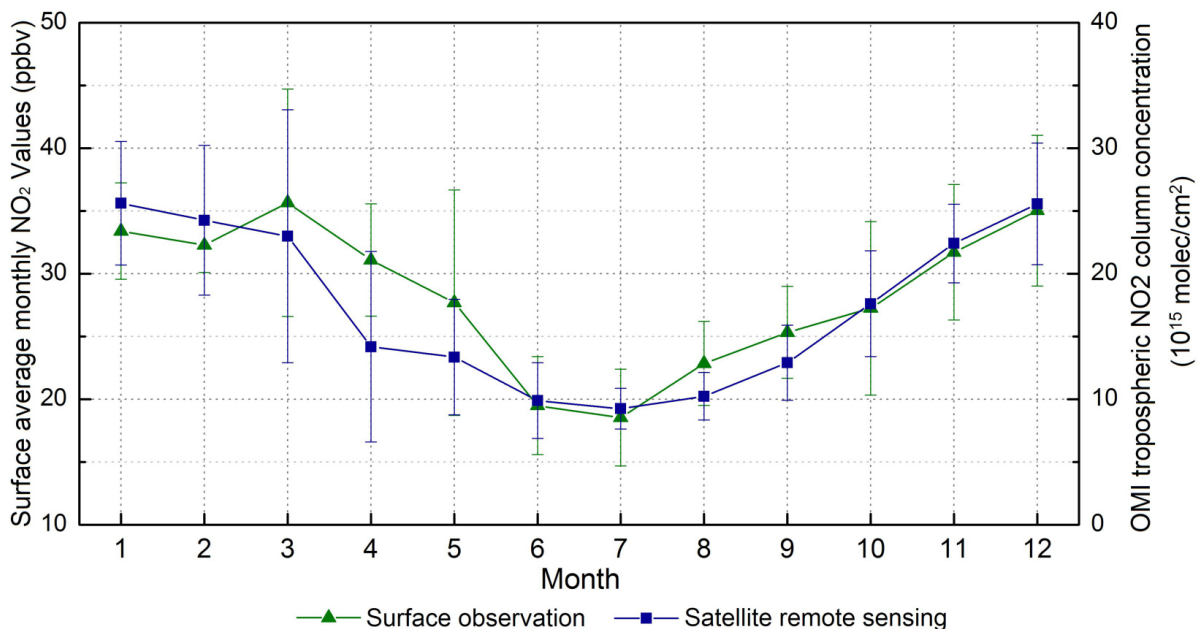


Figure 3. Comparison of 6-year monthly average NO₂ values from surface observation data and satellite remote sensing data.

areas close to Guangzhou and Zhaoqing, as well as Yingde of Qingyuan. In these areas, the NO₂ pollution concentration exceeded 5.0×10^{15} molec/cm². In the W-GD region, the local NO₂ column concentration was equal to that of the E-GD at about 2.5×10^{15} molec/cm². The high NO₂ pollution areas were mainly distributed in the urban areas of Maoming and Zhanjiang, and their NO₂ pollution concentration exceeded 3.0×10^{15}

molec/cm².

Figure 5 shows the monthly average value of tropospheric NO₂ column concentration obtained by OMI remote sensing data in the PRD and the adjacent areas over the past 9 years (2005-2013). It can be seen that the monthly average NO₂ column concentration value in the four areas had roughly the same change trends, where high values occurred in the winter and the

minimum value occurred in the summer. Because the lifetime of NO_x has significant relationship with the air temperature and solar radiation^[8, 10, 11], it is likely for high NO_2 pollution values to occur in winter with low temperatures and weak solar radiation. However, the natural NO_2 emission sources emit a relatively larger amount in summer and autumn (lightning processes, soil emissions etc.), but less in winter and spring. Under natural conditions, the NO_2 concentration shows a regular variation that it is on a high level in summer and autumn and on a low level in winter and spring^[10, 12, 38].

Additionally, under natural conditions, the yearly average value of tropospheric NO_2 column concentration was less than $1.0 \times 10^{15} \text{ molec/cm}^2$ ^[15, 17]. This demonstrates that the regular NO_2 variation in the PRD region and its adjacent areas was obviously different from the regular variations under natural conditions, and the tropospheric NO_2 concentrations in the four areas were obviously higher than those under natural conditions. This is mainly related to the large amounts of NO_x emitted by human activities in these areas.

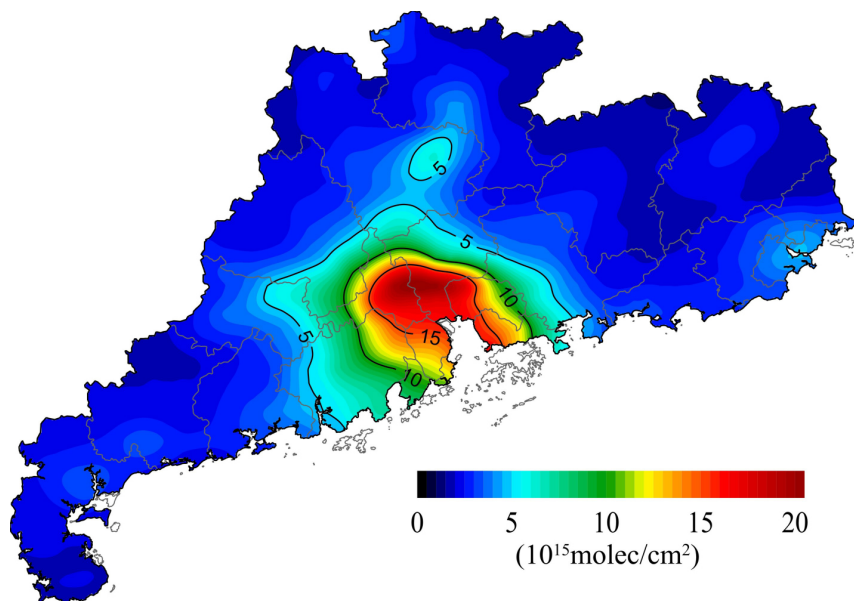


Figure 4. Distribution of tropospheric NO_2 column concentration values in the PRD and surrounding areas (E-GD, N-GD and W-GD region) from 2005-2013 according to OMI remote sensing data.

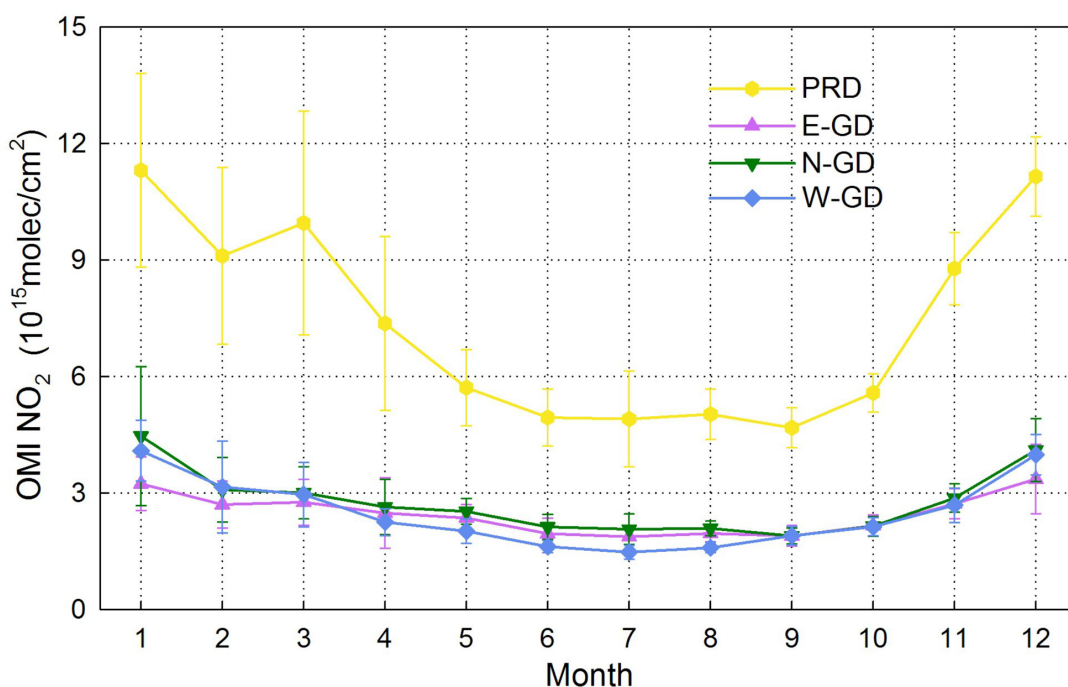


Figure 5. Monthly average OMI-sensed tropospheric NO_2 column concentration values in the PRD and adjacent areas (E-GD, N-GD, W-GD region) over the past 9 years (2005-2013).

3.3 Variation trend characteristics

In order to quantitatively analyze the variation trend characteristics of the tropospheric NO₂ column concentrations in the PRD and its adjacent areas, this study adopted the Sinusoidal model to fit the variation trend characteristics^[11, 12] in NO₂ concentration with time; the fitting formula is presented in detail as follows:

$$y = A_t + B_t x + C_t \sin\left(2\frac{\pi}{D_t}x + E_t\right) \quad (1)$$

where y is the monthly average value of the tropospheric NO₂ column concentration within the computing area; x is the month; t is the different region; A_t, B_t, C_t, D_t and E_t are the model parameters; $A_t + B_t x$ is

the variation trend line; $12 \cdot B_t / A_t$ is the relative change rate of the annual average value (annual growth rate); $C_t \sin\left(2\frac{\pi}{D_t}x + E_t\right)$ can describe the periodic change characteristics of the monthly average value of NO₂ column concentration (where C_t is the change amplitude, D_t is the cycle, and E_t is the peak value or valley value).

Based on Eq. (1), the monthly and annual average tropospheric NO₂ column concentrations in the PRD and its adjacent area was fitted, the fitting result is shown in Fig. 6, and the corresponding fitting curve parameters are shown in Table 1.

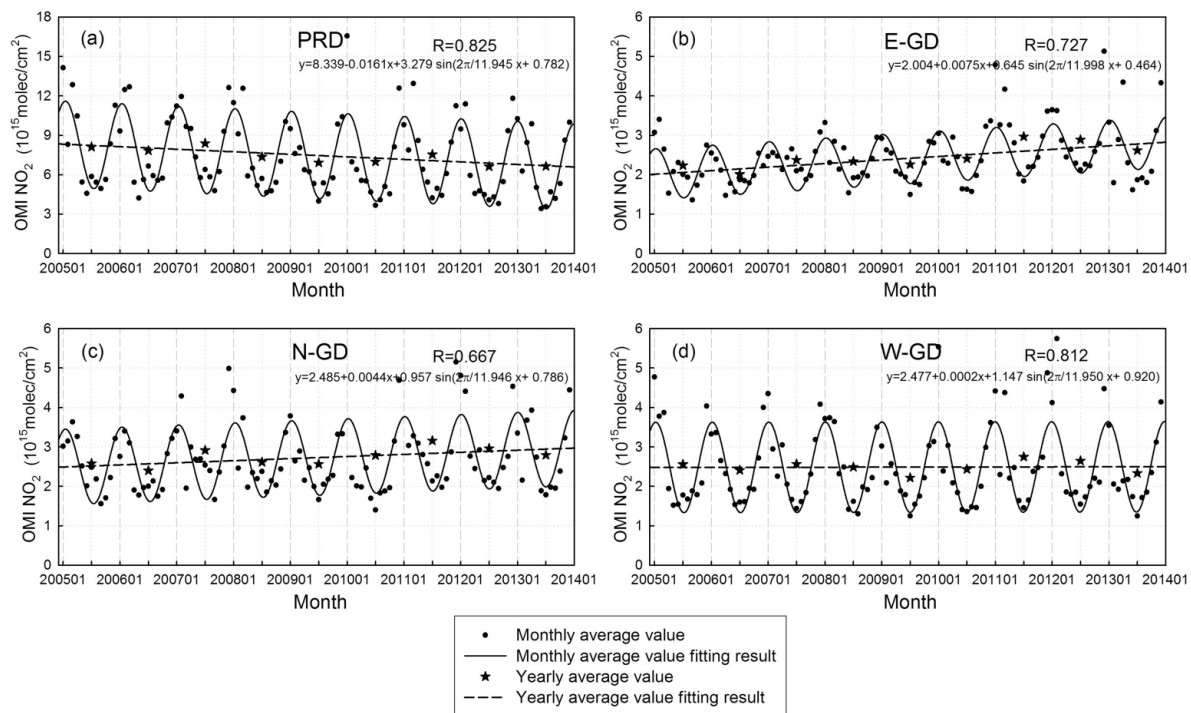


Figure 6. Sinusoidal model fitting result for tropospheric NO₂ column concentrations in the PRD and its adjacent areas from 2005-2013.

Table 1. Sinusoidal model fitting parameters for PRD and its adjacent areas.

Area	A_t	B_t	C_t	D_t	E_t	R	Annual growth rate (%)
PRD	8.339	-0.0161	3.279	11.945	0.782	0.825**	-2.32
E-GD	2.004	0.0075	0.645	11.998	0.464	0.727**	4.49
N-GD	2.485	0.0044	0.957	11.946	0.786	0.667**	2.12
W-GD	2.477	0.0002	1.147	11.950	0.920	0.812**	0.10

Note: ** means that it is of significant correlation on the level of 0.01.

According to the fitting results, the quarterly periodic change characteristics in the PRD and its adjacent areas were significant, the cycle averaged 12 months; high values occurred in December and January, and low values occurred in June and July. The variation amplitude of the monthly average value in the PRD was far larger than that of its adjacent areas, and the amplitude was about 3.3×10^{15} molec/cm². The change amplitude of the monthly average value in the E-GD

region was minimum, the amplitude was 0.6×10^{15} molec/cm². The PRD and W-GD fitting results were better, and the correlation coefficient was above 0.80. However the E-GD and N-GD region had a little less fitting results, and the correlation coefficient was approximately 0.70. The correlation of the 4 regions passed the significance test at the level of 0.01. Since there was significant cloud cover over the PRD and its adjacent areas, the OMI NO₂ inversion was directly

affected^[37], resulting in a huge variation in the quantity of valid grid point data, and causing a certain difference between the satellite remote sensing value and the curve fitting value. However as a whole, the sinusoidal model fitting curve can better reveal the variation characteristics over time for the tropospheric NO₂ column concentrations in the PRD and its adjacent areas. The fitting result can also be used for verifying the total NO_x emissions and their change trends in the PRD and its adjacent areas.

According to the variation trend of the annual average of the troposphere NO₂ column concentration from 2005 to 2013, the annual average value of NO₂ concentration in the PRD was gradually decreased by about 2.3% per year. This means that the NO₂ pollution degree in the PRD decreased year after year, and such a conclusion was consistent with the statistical result that the total number of haze days in the PRD were tending to decrease (Liu et al.^[39]). The annual average value of the tropospheric NO₂ column concentration in the areas surrounding the PRD was completely different from that of the PRD. Among these, the NO₂ concentration in the E-GD region greatly increased, and its annual growth rate was about 4.5%; the NO₂ concentration in the N-GD region increased gradually, and its annual growth rate was about 2.1%; but the NO₂ concentration in the W-GD region has remained unchanged over the past 9 years. It can be seen from these results that the NO₂ pollution in the E-GD region was increasing rapidly, the NO₂ pollution in the N-GD region was gradually increasing each year, but the NO₂ pollution in the W-GD region remained at the original level.

3.4 Analysis of the impact of human activities

In order to study the impact of human activities on NO₂ concentration distribution characteristics and variation tendencies in the PRD and its adjacent areas, this study adopted and analyzed all social and economic development data of Guangdong province from 2012, as well as data related to the production output value of secondary industry and the amount of civilian vehicles from 2005 to 2012. A comparative analysis on the NO₂ concentration distribution and its annual average value with these data was then carried out.

According to 2013 Guangdong Statistical Yearbook, in 2012, 3/4 of the primary energy consumption of Guangdong came from fossil energy, among which raw coal and raw oil were respectively 49% and 27%. The secondary industry of Guangdong consumed 99% or more of the raw coal, with the manufacturing industry accounting for 31% of the raw coal consumption, and the electricity, fuel gas and water production & supply industry accounting for 68% of the raw coal consumption. From these statistics it can be seen that the secondary industry was the mostly important artificial NO_x pollution source. Correspondingly, the production value of the secondary industry can reveal the NO_x pollution generated in its

production process. Additionally, the traffic & transport industry, warehousing industry and postal service industry of Guangdong consumed approximately 60% of oil products. Therefore mobile sources on the roads are also one of the important artificial NO_x pollution sources. The total amount of civilian vehicles can reveal the NO_x pollution caused by motor vehicle exhaust emissions.

The statistical data related to the PRD and its adjacent areas in 2012 are shown in Table 2. The statistical results show that the area of the PRD is only 30% of the total area of Guangdong province, but the local permanent resident population was 54% of that of Guangdong Province, and the local GDP, secondary industry production value and gross industrial output value account for almost 80% of that of Guangdong Province. The comprehensive energy consumption and electricity consumption of the PRD are respectively 72% and 77% of that of Guangdong Province; the total amount of civilian vehicles in the PRD was more than 80% of that of Guangdong Province. Additionally the thermal power plants in Guangdong province are widely distributed in the PRD^[4, 40]. From all of these facts it can be seen the population density of the cities in the PRD was huge, the local economy and industrial production and activities were huge, and the local energy and electric power consumption, quantity of automobiles and corresponding raw coal and oil consumption were all on a high level. The PRD consumed a huge amount of fossil energy and emitted a huge amount of polluting gases (including SO₂, NO_x etc.), which caused the tropospheric NO₂ column concentration in the PRD to be higher than that of its adjacent areas (shown by Fig.4); therefore the PRD is one of the four high pollution areas of China.

The relation between the annual average value of the tropospheric NO₂ column concentration and the production value of secondary industry in the PRD and its adjacent areas from 2005 to 2012 is shown in Table 3. According to the statistical results, the annual average value of the tropospheric NO₂ column concentration and the production value of secondary industry in the PRD had an obvious negative correlation relationship ($P < 0.05$) with a correlation coefficient of -0.73; which showed the production value of secondary industry in the PRD was gradually increasing, but the NO₂ concentration was gradually decreasing. According to the statistical data for 2012 (Table 2), per unit of GDP energy consumption in the PRD was lower than that of its adjacent three areas. Such conclusions show that the production value of secondary industry in the PRD is gradually increasing, but the NO₂ concentration per unit of production value of secondary industry was gradually decreasing, which caused the annual average NO₂ value (total NO₂ emissions) to decrease year after year. This demonstrates that the local governments and enterprises in the PRD focused on improving and optimizing

industrial structure and production efficiency and actively boosted industrial transformation and upgrade. Therefore, the NO₂ pollution emissions from the local secondary industry and the pollution degree of NO_x were reduced. However in E-GD and N-GD, the annual average NO₂ value had a positive correlation relationship with the production value of secondary

industry, and correlation coefficients of 0.86 and 0.67, respectively. When the production value of secondary industry increased, the annual average NO₂ value increased as well. To some extent, these show that the secondary industry in E-GD and N-GD are the major contributor to NO₂ pollution. The correlation coefficient of W-GD was only 0.49, which was at a low level.

Table 2. Statistical data of PRD and its adjacent areas during 2012

	PRD	E-GD	N-GD	W-GD
Area of land(sq.km)	54754 (30.5%)	15462 (8.6%)	76751 (42.7%)	32644 (18.2%)
Year-end permanent population(10,000 person)	5689.6 (53.7%)	1709.7 (16.1%)	1556.9 (14.7%)	1637.8 (15.5%)
Gross regional domestic product (GDP) (RMB 0.1 billion)	47779.6 (79.1%)	4138.9 (6.9%)	4638.4 (7.7%)	3821.8 (6.3%)
Production value of secondary industry (RMB 0.1 billion)	22084.6 (79.2%)	2272.5 (8.2%)	1921.6 (6.9%)	1585.7 (5.7%)
Value of gross output (RMB 0.1 billion)	20731.7 (79.8%)	2119.1 (8.2%)	1735.4 (6.7%)	1390.2 (5.3%)
Comprehensive energy consumption (10,000 t standard coal)	25313.7 (71.9%)	2707.9 (7.7%)	3201.0 (9.1%)	3968.0 (11.3%)
Electricity consumption (0.1 billion/kw.h)	3397.9 (77.0%)	376.6 (8.5%)	232.6 (5.3%)	406.3 (9.2%)
Unit GDP energy consumption (tonnage standard coal/RMB10,000)	0.530	0.654	1.038	0.683
Quantity of civilian vehicles (10,000 vehicles)	532.5 (81.4%)	46.2 (7.1%)	40.6 (6.2%)	34.4 (5.3%)

Note: The figures in the brackets are the percentages to the total quantity of Guangdong province.

Table 3 shows the correlation between the quantity of civilian vehicles and the annual average tropospheric NO₂ column concentration from 2005 to 2012. The statistical results show that there is a significant negative correlation ($P < 0.05$) relationship between the annual average NO₂ concentration value and the quantity of civilian vehicles in the PRD, and the correlation coefficient was -0.75. This shows that in recent years, the annual average NO₂ concentration value (total NO₂ emissions) tended to decrease while the quantity of civilian vehicles in the PRD was increasing. Obviously, the NO₂ emitted from automobiles in the PRD was decreasing year after year. Such a result could not be realized without the stricter administer of local governments, such as upgrading motor vehicle exhaust emission standards, improving oil quality, phasing out Yellow-labeled trucks, vigorously developing public transport and supporting and encouraging the use of clean-energy motor vehicles. However in E-GD and N-GD, the variations of the annual average tropospheric NO₂ column concentrations had positive correlation relationships with the variations the quantity of civilian vehicles, where the correlation coefficients were respectively 0.89 and 0.73. It can be seen that the

Table 3. Correlation coefficient of annual average NO₂ concentration, production value of secondary industry and quantity of civilian vehicles in the PRD and its adjacent areas 2005-2012.

	Production value of secondary industry	Quantity of civilian vehicles
PRD	-0.727*	-0.752*
E-GD	0.863**	0.886**
N-GD	0.672	0.725*
W-GD	0.489	0.454

increases of NO₂ concentration had close relationships with the increase of the number of automobiles in E-GD and N-GD, and motor vehicle exhaust emissions are one of the main NO₂ pollution sources. The correlation coefficient in W-GD was only 0.45, which was at a low level.

4 CONCLUSIONS

Based on the tropospheric NO₂ column concentration data of OMI from 2005 to 2013, this paper studied the spatial and temporal distribution and variation tendency characteristics of NO₂ in the PRD and its adjacent three areas (E-GD, N-GD and W-GD), and analyzed the impact of human activities on NO₂, with the following conclusions reached:

(1) The tropospheric NO₂ column concentration data of OMI had good consistency with the surface NO₂ observation value; the OMI NO₂ remote sensing results had high reliabilities, and can reveal the surface NO₂ distribution and variation characteristics.

(2) The average tropospheric NO₂ column concentration (7.4×10^{15} molec/cm²) in the PRD was almost 3 times of that in the adjacent areas, and the local high pollution areas in the PRD are connected together, where the atmospheric pollution in one city affected that in other cities, and the urban group effect was pronounced. The NO₂ concentrations in E-GD, N-GD and W-GD was similar. The variation tendencies of the monthly average NO₂ column concentrations in the PRD and its adjacent areas were comparatively consistent; high values occurred in winter, low values occurred in summer, and the concentration level was apparently higher than that under natural conditions. Such phenomenon are mainly related to the emissions from human activities.

(3) The sinusoidal model fitting curve can well reveal the change characteristics of the tropospheric NO₂ column concentration variation over time in the PRD and its adjacent areas, and the fitting result can be used for verifying the NO₂ emissions and variation trends in the PRD and its adjacent areas. The NO₂ column concentration in the PRD had a tendency to decrease year after year, the NO₂ column concentration in E-GD was growing rapidly, the growth of the NO₂ column concentration in N-GD was significant, but the one in W-GD remained at a comparatively stable level.

(4) The huge urban population density, high economic and industrial activity, great number of automobiles and huge fossil energy consumption in the PRD caused the local tropospheric NO₂ column concentration to be far higher than that of its adjacent areas, and thence the PRD is a high NO₂ pollution area. The production value of secondary industry and total quantity of automobiles in the PRD was increasing year after year, but the local NO₂ concentration showed a tendency to decrease; both showed obvious negative correlations. This conclusions reveal that the actions of local governments in the PRD for controlling and preventing motor vehicle exhaust emissions and boosting local industrial transformation and upgrade have produced preliminary results. The NO₂ concentration in E-GD and N-GD had good positive correlations with the local production values of secondary industry and total quantity of automobiles. This demonstrates that the secondary industry and motor vehicles in E-GD and N-GD contributed too much to the NO₂ pollution. The correlation between the NO₂ concentration in W-GD and the local production value of secondary industry and total quantity of automobiles was not on a high level.

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