Y

Original Research Paper

Spatio-Temporal Variations of Urban NO₂ Concentrations in China

Fangshu Gao*, Ying Peng**, Guoxiang Wang and Yi Qian*

College of Geography Science, Nanjing Normal University, 1st Wenyuan Road, Nanjing 210046, P. R. China **Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, 73st East Beijing Road, Nanjing 210008, P. R. China; *Corresponding authors: Yi Qian and Fangshu Gao

ABSTRACT

Nat. Env. & Poll. Tech. Website: www.neptjournal.com *Received:* 17-5-2013 *Accepted:* 4-7-2013

Key Words: Urban NO₂ concentration Spatial variation Temporal variation The spatial and temporal characteristics of NO₂ concentration were analysed from 31 cities in China over a six-year period (2003-2008). Urban NO₂ concentrations were in the range of 12-73 μ g/m³, and its pollution levels had an expanding trend in the temporal variations during 2003-2008. Spatially, there was an evident difference for urban NO₂ concentration in the three regions of the eastern, central and western China. NO₂ concentrations in cities had a significant reduction in eastern China, an obviously heightened trend in central China and a small heightened trend in western China due to the combined effects of industrial structural transformation and industrial upgrading, as well as control measures.

INTRODUCTION

China

Atmospheric nitrogen pollution is a very topical environmental issue, and captures the attention of policy makers (Curtis et al. 2006, Zhang et al. 2007, Shon & Kim 2011). NOx (NO + NO₂) has been one of the most important pollutants in urban areas for its massive discharge from motor vehicle exhaust and stationary sources such as electric utilities as well as the rapid development and industrialization (Zhang et al. 2007, Zhu et al. 2012, Ozkurt et al. 2013). Environmental NO₂ is also a potential risk factor for summer smog, acid rain and ischemic stroke and vascular dementia (Latza et al. 2009, Andersen et al. 2009, Li & Xin 2013).

Driven by rapid socioeconomic development and intensive energy use, NO₂ emissions and their impacts have evolved into a serious environmental challenge for China (Wang et al. 2009, Takashima et al. 2011). In China, energy consumptions and motor vehicles especially for private cars are increasing rapidly in recent years, which led to the swift growth of urban NO₂ emission. Moreover, NOx index was eliminated while NO, index was slacked in the "Ambient Air Quality Standards (GB3095-1996)", which also led to an increase of NO₂ concentration in cities (Chen & Yu 2010). NO₂ emission from thermal power plants added 2.4 million tons during the only six years (1999-2004), which was the sum of that in the 12 years (1987-1998). Beijing and regions of northeastern China were found to be the seriously polluted area-of the world (Richter et al. 2005). In the future 30 years, NO₂ emission would continue to grow steadily and become the largest country beyond USA under the present controlling measures which inflict suffering on the public health and the ecological environment development (Zhou et al. 2008, Fang et al. 2010, Loft et al. 2012, Li & Xin 2013).~

Under environmental pressure, China's government is attempting to shift its development mode from one dependent on intense fossil energy inputs with consequent high emissions to a more resource-efficient and environmentfriendly alternative (Wu 2009). However, current policies and regulations of NO₂ control are still not perfect in such a short history in China. Taking the NO₂ emission standard for example, the limit values are given only in power plants and motor vehicles (Zhang et al. 2008). Moreover, prior studies mostly concerned in local regions and a certain province (autonomous region, and municipality directly under the central government) for discussing the spatio-temporal variations and sources of NO₂/NOx (Zhang et al. 2007, Chen et al. 2010, Li & Wang 2011) and urban photochemical air pollution (Zhang et al. 1998, Li 2009). As for the national scale, more researchers used the satellite measure to map the spatial variations of tropospheric NO₂ columns (Richter et al. 2005, Zhang et al. 2007, Wang et al. 2009) while less applied the geo-statistical method to understand the spatial and temporal variations of NO₂ concentration. In this study, the long-term trends of urban NO₂ concentrations were evaluated, and its spatial distributions on a national scale were done by a geo-statistical method for NO₂/NOx control strategy establishment, further for the harmonious development of the national environment and economy.

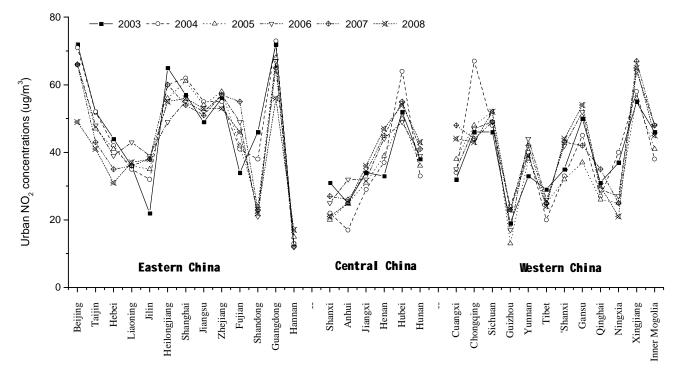


Fig. 1: Urban NO₂ concentration of 31 provinces (autonomous regions and municipalities directly under the Central Government, excluding Taiwan province, Hong Kong and Macao Special Administrative Region) during 2003-2008. Geographically, China is divided into the eastern (including Beijing, Tianjin, Hebei, Liaoning, Jilin, Heilongjiang, Shanghai, Jiangsu, Zhejiang, Fujian, Shandong, Guangdong, Hainan provinces), the central (including Shanxi, Anhui, Jiangxi, Henan, Hubei, Hunan provinces) and the western region (including Guangxi, Chongqing, Sichuan, Kunming, Tibet, 'Shanxi, Gansu, Ningxia, Xinjiang, Anner Mongolia autonomous region).

METHODS AND SOURCES OF DATA

Data sources: All the data of NO₂ concentrations (μ g/m³) and the number of population used in this study were from "The China Year Books" during 2004-2009. There is usually no data integrality for the index of NO₂ concentration is not monitored in urban monitoring standard, so urban NO₂ concentrations in the capital 31 cities (exclude Taiwan) replaced those in their corresponding provinces, as shown in Fig. 1.

Measurement of regional difference: Standard error (SE) is a common method to measure regional absolute difference. Due to the effect of the increasing population, a single SE does not to test completely regional differences of NO₂ concentrations. What is more, the two indexes above cannot be decomposed into formal between-group and within-group difference. Thus, all of the SE, coefficient of variation (CV) and Theil (1967) measures were used to analyse the difference in the study, and the relative formulates above are as follows:

$$SE = \sqrt{\sum_{j=1}^{n} (Y_j - \overline{Y})^2 / n}$$
 ...(1)

Where SE is the standard error, Y_j is urban NO₂ concentration of province j, \overline{Y} is the annual average, and n is the number of provinces.

$$CV = \sqrt{\sum_{i=1}^{n} \left(Y_{i} - \overline{Y} \right)^{2} / p_{i} / \overline{Y}} \qquad \dots (2)$$

Where *CV* is the weighted coefficient of variation, p_j is the proportion of province *j* in the whole nation population. Both Y_j and \overline{Y} are the same as the formula (1) above.

$$T = BT + WT = \sum_{i=1}^{m} G_i \log(G_i / p_i) + \sum_{i=1}^{m} G_i \sum_{j=1}^{i} (g_i / G_i) \qquad \dots (3)$$

Where *T* is the Theil, *BT* is the between-group Theil, *WT* is the within-group Theil, *m* is the number of the decomposed region, *t* is the number of provinces of *i* region, *Gi* is the proportion of region *i* of the whole nation NO₂ concentration, p_i is the proportion of province *j* in the whole nation population, g_i is the proportion of province *j* in region *i* of the whole nation NO₂ concentration, p_i is the proportion of province *j* in region *i* of the whole nation NO₂ concentration, p_i is the proportion of province *j* in region *i* of the whole nation population.

Exploratory spatial data analysis (ESDA): Exploratory

Table 1: Proportions and decompositions of differences in urban NO_2 concentrations using Theil index (2003-2008).

Т	WT	BT
0.4105	0.3935 (95.9%)	0.0170 (4.1%)
0.4179	0.3976 (95.2%)	0.0203 (4.8%)
0.3646	0.3488 (95.7%)	0.0158 (4.3%)
0.3411	0.3257 (95.5%)	0.0155 (4.5%)
0.3265	0.3115 (95.4%)	0.0151 (4.6%)
0.3186	0.3026 (95.0%)	0.0160 (5.0%)
	0.4179 0.3646 0.3411 0.3265	0.4105 0.3935 (95.9%) 0.4179 0.3976 (95.2%) 0.3646 0.3488 (95.7%) 0.3411 0.3257 (95.5%) 0.3265 0.3115 (95.4%)

Table 2: The number of categories I, II and III in different provinces.

	Ι	II	III	
2003	15	13	3	
2008	9	13	8	

spatial data analysis (ESDA) extends exploratory data analysis to spatial data for the process of spatially distributed visualization, the identification of atypical location or outliers, the representation of spatial association pattern, the identification of spatial cluster or hotspot, coldspot, and spatial regime as spatial heterogeneity. In this study, Getis-Ord Gi* was used to evaluate hotspots and coldspots spatial pattern from the observed data. The value of G* would be positive if the clustering happened in one observed data result or that value is more than mean and the value of G* would be negative if the clustering value which is less than mean happened in one observed data result (Joko-Prasetyo et al. 2012). The equation of G* statistic could be defined as in the equation (4).

$$G^* = \sum_{j=1}^{n} W_{ij}(d) Z_j / \sum_{j=1}^{n} Z_j \qquad \dots (4)$$

Based on this equation, is the element of spatial weight matrix and (d) is the element of distance inter spatial object.

RESULTS AND DISCUSSION

Temporal variations of urban NO₂ **concentration:** During 2003-2008, urban NO₂ concentrations were in the range of 12-73µg/m³ (Fig. 1), and the NO₂ level of 46.2% (\pm 5.4%) cities attained Grade I (0-40µg/m³) with a downward trend while others were in Grade II (40-80µg/m³) with an upward trend on the whole. Fig. 2 exhibits that the annual average concentrations of urban NO₂ vary from 39.8 to 42.4 µg/m³, and have a descending-ascending phase while both their *CV* and *SE* have a similarly descending trend. During 2003-2005, the annual average concentrations had the descending phase with a degree of 4.3%, and their *CV* and *SE* had the stock declining trend. And then, the annual average concentrations had the descending phase with a degree of 2.3%, while their *CV* and *SE* had the declining trend year by year during 2005-

2008. The results above indicated that urban NO_2 pollution had an expanding trend in the temporal variations during 2003-2008.

Spatial variations of urban NO₂ **concentration:** Based on the urban NO₂ concentrations and the numbers of population, the Theil indexes including WT and BT were calculated during 2003-2008 (Table 1). The T and BT reduced by 11.19% and 11.37% during 2003-2005 while by 12.61% and 13.24% during 2005-2008, respectively. The proportion of BT to T increased from 4.1% to 5.0% and that of WT to T reduced from 95.9% to 95.0%. As for the contributions of WT to T and BT to T, BT from eastern, central and western China contributed from 94.97% to 95.86% to T of the whole China. This also indicated that there was a visible descending trend for the regional differences of urban NO₂ concentrations due to their inner differences of urban eastern, central and western China.

Distributions of urban NO₂ concentration types: Based on the general trend of urban NO₂ concentration during 2003-2008 (Fig. 2), the NO₂ concentrations in the two years (2003 and 2008) were selected and its Getis-Ord Gi* values were calculated by the ESDA mode of ARCGIS software (ver. 9.3). And the natural breakpoint in descending order scores was used to classify the provinces as category I, II and III, as shown in Fig. 3. The number of category I, II and III in different provinces were also made statistically, as given in Table 2.

On the whole, there were a large number of provinces in category I degree and the smallest amount of provinces in category III degree (Table 2). The number of provinces in category I degree reduced from 15 in 2003 to 9 in 2008 with a high degree of 40%. The number of provinces in category II degree stayed the same (13), but the amount of provinces in category III degree increased from 3 in 2003 to 8 in 2008. The results showed that urban NO₂ pollution was rather serious with a severe "duality" in China, which due to the combined effects of industrial structural transformation and industrial upgrading (Yang 2008, Wang et al. 2009, Chen et al. 2012). Fu et al. (2011) found most polluting industries in central and western China were higher than that of the eastern regions during 2003-2008 and would bound to transfer to western regions in the future. Liu et al. (2012) also found power industry and agro-food processing industry had appeared to shift to the central and western regions since 2001 while paper products industry, non-metallic mineral products industry began to shift to the central region in 2006. Moreover, economic development model and industrial restructuring in China proved that the optimization of industrial structure was the guarantee of harmonious development of economic construction and environmental governance

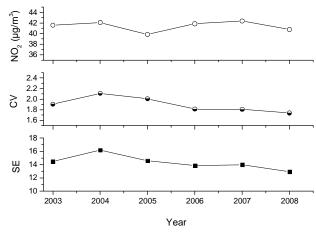


Fig. 2: Annual average values of urban NO₂ concentrations, their standard errors (*SE*) and coefficients of variation (*CV*) in China during 2003-2008.

(Zhang 2012). A research on industrial pollution in different industrial sectors showed that changes in industry structure among industrial sectors have a significant influence on pollution in China's high pollution intensity industries sectors and low and middle pollution intensity industries (Jiang 2010). The industrial pollution transferred from the eastern areas to central and western areas as well as industrial transfer in eastern areas, which may cause more serious NO₂ pollution and inevitably further deteriorate ecological environment in central and western areas while alleviate the NO₂ pollution of eastern China.

Regionally, the category I of urban NO_2 concentration appeared mainly in eastern China while other categories (II and III) concentrated mainly in western China (Fig. 3), which was in good agreement with the characteristics of NOx emission and reduction potential and the transfer of pollutionintensive industries in China (Yang 2008, Wang et al. 2010). Nowadays, with the entrance into WTO and the development of western regions, increasing overseas investments have been introduced to western regions, including some pollution-intensive industries refused by the eastern area. The moving eastward of industrial output and the westward of industrial contamination bring some pollution, and burdened the weakened biological environment in the west regions.

Compared with Fig. 3a and Fig. 3b, the degree of urban NO_2 concentration in Shanghai, Jiangsu and Zhejiang province of the Yangtze River Delta Economic Zone and Guangzhou province kept stable in the category I in 2003 and 2008, which indicated that urban NO_2 pollution was significant, though their regional economic development was in the rapid growth stage. The degree of NO_2 concentration in Hubei, Gansu and Sichuan province was also steady in the category I in 2003 and 2008,-which suggested that the

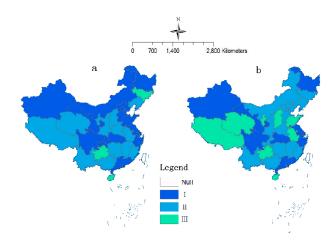


Fig.3: Degree distributions of urban NO₂ concentration in 2003 (a) and 2008 (b).

three provinces are in the relatively developed industry region of central-west China, environment protection must be enhanced and the ability of continuous development also is improved in undertaking industrial transformation. Moreover, the category II regions kept stable in Liaoning and Fujian province of eastern China, Henan, Hunan and Jiangxi province of central-China, and Yunnan, Guangxi and 'Shanxi province of western China, while the category III regions in Hainan province of eastern China and Guizhou province of western China.

Degree distributions of urban NO₂ concentration changed in some provinces between 2003 and 2008 (Fig. 3a, 3b). The degree of urban NO₂ concentration in the regions of Beijing, Tianjin and Hebei province (Jing-Jin-Ji region) changed from the category I in 2003 to II in 2008, which was attributed to the government policy for the Beijing 2008 Olympic Games. In order to heed the call for the Green Beijing Olympic Games, long-term effective-measures of controlling atmospheric pollution-such as controlling automobiles emission were taken by increasing investments in Beijing and its neighbouring regions of Tianjin and Hebei province, which led to the decline of regional NO₂ concentration (Yu et al. 2009, Li & Wang 2011). In addition, the degrees of urban NO₂ concentration had highly reduced in those provinces: the category in Shandong and Inner Mongolia from I in 2003 to III in 2008, in Chongqing from I to II, and in the regions of Anhui, Tibet, Ningxia and Qinghai from II to III (Figs. 3a, 3b).

Spatial distributions of urban NO₂ **concentrations:** In order to discuss the dynamic changes of urban NO₂ concentrations in a spatial scale, the two regions of the increased and decreased regions (Fig. 4) were done by the Manual Classify method in the ARCGIS software during the two stages

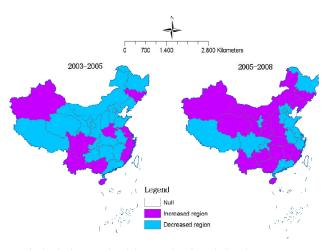


Fig.4: The increased and decreased regions during the two stages (2003-2005, 2005-2008).

sify method in the ARCGIS software during the two stages (2003-2005, 2005-2008) based on Fig. 1.

The number of increased provinces had an increasing trend during 2003-2008, as shown in Fig. 4. There were 12 and 17 provinces belonging to the increased regions during 2003-2005 and 2005-2008, respectively. As for regional distributions, the numbers of increased provinces in eastern, central and western China were six, one and five during 2003-2005 and four, five, eight during 2005-2008. That is to say, there had a significant reduction in eastern China, an obviously heightened trend in central China and a small heightened trend in western China. This result above, again, proved that the tendency towards worsening NO₂ pollution has been arrested in eastern China where the ability of sustainable development has been improved. But the regions of NO₂ concentration increase has been spreading and the regional ability of sustainable development needed further improvement in central and western China, which should be paid attention to industry, academe and government.~

Compared to Fig. 3 and Fig. 4, the NO₂ concentrations in Jiangsu, Hubei, Xinjiang, Gansu and Sichuan province were in both the category I and the increased regions, which indicated that regional NO₂ pollution was relatively serious. Though the NO₂ concentrations were in the category III in Hainan and Guizhou province and in the category II in those provinces of Liaoning, Fujian, Henan, Hubei, Jiangxi, Yunnan, Guangxi and Shanxi, all of these provinces were in the increased regions, which suggested that regional NO₂ pollution had a worsening tendency. There was an effective pollution treatment for NO₂ emission for regional NO₂ concentrations were declining in Heilongjiang, Zhejiang, Shanghai and Guangzhou provinces though the provinces were in the category I. What is more, regional NO₂ concentrations declined from the category II to category III in Shanxi, Tibet, Ningxia, Anhui, Qinghai province and from the category I to II in Beijing, Tianjin, Hebei, Shandong, Inner Mongolia and Chongqing. At the same time, all of the 11 provinces above were in the declined region. This indicated that NO_2 emission had a good control in the 11 provinces of China.

CONCLUSIONS

 NO_2 concentrations from 31 cities in China over a six-year period (2003-2008) were used for the study of temporal and spatial distribution. Urban NO_2 concentrations were in the range of 12-73µg/m³, and its pollution levels had an expanding trend in the temporal variations during 2003-2008. Spatially, there was a-visible descending trend for the regional differences of urban NO_2 concentrations due to their inner differences of urban NO_2 concentrations due to their inner differences of urban eastern, central and western China. NO_2 concentrations in cities had a significant reduction in eastern China, an obviously heightened trend in central China and a small-heightened trend in western China due to the combined effects of industrial structural transformation and industrial upgrading.

REFERENCES

- Andersen, Z.J., Kristiansen, L.C., Andersen, K.K., Olsen, T.S., Hvidberg, M., Jensen, S.S., Ketzel, M., Latza, U., Gerdes, S. and Baur, X. 2009. Effects of nitrogen dioxide on human health: Systematic review of experimental and epidemiological studies conducted between 2002 and 2006. International Journal of Hygiene and Environmental Health, 212: 271-287.
- Chen, S.S., Shu, J. and Xu, J.Z. 2010. Trophospheric NO₂ pollution analysis of some typical cities in China. Resources and Environment in the Yangtze Basin, 19(12): 1484-1490 (in Chinese).
- Chen, W., Liu, D.S. and Li, J. 2012. Research on countermeasures of environment pollution on industry transfer. Environmental Protection, 9: 48-50 (in Chinese).
- Chen, Y. and Yu, T. 2010. Trend analysis of NOx variation in Guangzhou city. Environmental Science and Management, 35: 47-49 (in Chinese).
- Curtis, L., Rea, W., Smith-Willis, P., Fenyves, E. and Pan, Y. 2006. Adverse health effects of outdoor air pollutants. Environment International, 32: 815-830.
- Fang, S.Z., Zhao, S.M., Li, F.R. and Yang, S.P. 2010. Current situation of emission models and emission listing of nitrogen oxides. Environmental Science Survey, 29(3): 4-7 (in Chinese).
- Fu, S.X., Zhang, W.B. and Zhang, K.Y. 2011. A study on transfer of polluting industries and their regional distribution based on total factor productivity. Finance and Economics, 11: 53-68 (in Chinese).
- Jiang, K. 2010. Decomposing and analyzing the environmental pollution of different type of pollution of industry based on the panel data of industrial from 1998 to 2005. Ecological Economy, 4: 96-99, 113 (in Chinese).
- Joko-Prasetyo, S.Y., Subannar, Winarko, E. and Daryono, B.S. 2012. Endemic outbreaks of brown planthopper (*Nilaparvata lugens* Stal.) in Indonesia using exploratory spatial data analysis. Journal of Computer Science Issues, 9: 162-171.
- Li, H.R. and Xin, X.Y. 2013. Nitrogen dioxide (NO₂) pollution as a potential risk factor for developing vascular dementia and its synaptic mechanisms. Chemosphere, 92: 52-58.
- Li, L.J. and Wang, Y. 2011. The characterization of $\rm NO_2$ pollution in Beijing based on satellite and conventional observation data. Acta Scientiae

Nature Environment and Pollution Technology

Vol. 12, No. 3, 2013

Circumstantiae, 31(12): 2762-2768 (in Chinese).

- Li, Y.X. 2009. Analysis on the reasons of the formation of nitrogen oxides pollution and photochemical smog of motor vehicle. Sci-Tech Information Development and Economy, 19(6): 167-169 (in Chinese).
- Loft, S., Sørensen, M., Tjønneland, A., Overvad, K. and Raaschou-Nielsen, O. 2012. Stroke and long-term exposure to outdoor air pollution from nitrogen dioxide: A cohort study. Stroke, 43: 320-325.
- Liu, Q.L., Wang, Q. and Li, P. 2012. Regional distribution changes of-pollution-intensive industries-in-China. Ecological-Economy, 1: 107-112 (in Chinese).
- Ozkurt, N., Sari, D., Akalin, N. and Hilmioglu, B. 2013. Evalution of the impact of SO₂ and NO₂ emmissions on the ambient air-quality in Çan-Bayramiç region of northwest Turkey during 2007-2008. Science of the Total Environment, 456-457: 254-266.
- Richter, A., Burrows, J.P., Nüß, H., Granier, C. and Niemeier, U. 2005. Increase in tropospheric nitrogen dioxide over China observed from space. Nature, 437: 129-132
- Shon, Z.H. and Kim, K.H. 2011. Impact of emission control strategy on NO₂ in urban areas of Korea. Atmospheric Environment, 45: 808-812.
- Takashima, H., Irie, H., Kanaya, Y. and Akimoto, H. 2011. Enhanced NO₂ at Okinawa Island, Japan caused by rapid air-mass transport from China as observed by MAX-DOAS. Atmospheric Environment, 45: 2593-2597.
- Theil, H. 1967. Economics and Information Theory. North Holland: Amsterdam.
- Wang, Y.Q., Jiang, H., Zhang, X.Y., Zhou, G.M., Yu, S.Q. and Xiao, Z.Y. 2009. Temporal-spatial distribution of tropospheric NO₂ in China using OMI satellite remote sensing data. Research of Environmental Sciences, 22(8): 932-937 (in Chinese).
- Wu, X.Q. 2009. Current conditions, problems and countermeasures-of NOx pollution control-in China. Science and Technology Industry of China,

8: 13-16 (in Chinese).

- Yang, Y. 2008. The comparative study to the eastern and western regions transfer of pollution-intensive industries in China. Ecological-Economy, 1: 285-288 (in Chinese).
- Yu, H., Wang, P.C., Zong, X.M., Li, X. and Lv, D.R. 2009. Variations of NO₂ vertical column densities in Beijing region during the 2008 Olympic Games based on satellite measurement. Bulletin of Science and Technology, 54(3): 299-304 (in Chinese).
- Zhang, C.Y., Wang, S.X., Xing, J., Zhao, Y. and Hao, J.M. 2008. Current status and future projections of NOx emissions from energy related industries in China. Acta Scientiae Circumstantiae, 28(12): 2470-2479 (in Chinese).
- Zhang, X.Y., Zhang, P., Zhang, Y., Li, X.J. and Qiu, H. 2007. The trend, seasonal cycle, and sources of tropospheric NO₂ over China during 1997-2006 based on satellite measurement. Science in China Series D: Earth Science, 150(2): 1877-1884 (in Chinese).
- Zhang, Y.F. 2012. Environmental protection, international trade and China's industrial upgrading road. Journal of Guangdong Business College, 27(5): 31-38 (in Chinese).
- Zhang, Y.H., Shao, K.S., Tang, X.Y. and Li, J.L. 1998. The study of urban photochemical smog pollution in China. Acta Scicentiarum Naturalium Universitatis Pekinesis, 34(2-3): 392-340 (in Chinese).
- Zhu, N., Li, H.Y., Han, M., Guo, L., Chen, L.Q., Yun, Y., Guo, Z., Li, G.K. and Sang, N. 2012. Environmental nitrogen dioxide (NO₂) exposure influences development and progression of ischemic stroke. Toxicology Letters, 214: 120-130.
- Zhou, W., Wang, X.S., Zhang, Y.H., Su, H. and Lu, K.E. 2008. Current status of nitrogen oxides related pollution in China and integrated control strategy. Acta Scientiarum Naturalium Universitatis Pekinensis, 44(2): 323-330 (in Chinese).