## Spatio-temporal Distributions of Tropospheric NO<sub>2</sub> over Oases in Taklimakan Desert, China

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**Abstract:** Soil biogenic NO emission is one of the most important sources of atmospheric nitrogen oxides ( $NO_x$ ) worldwide. However, the estimation of soil source, especially in arid areas presents large uncertainties because of the substantial lack of measurements. In this study, we selected the Ruoqiang oases on the southeastern edge of the Taklimakan Desert, China as the study area and applied Ozone Monitoring Instrument (OMI) NO<sub>2</sub> retrievals (DOMINO v2.0, 2005–2011) to investigate the spatial distribution and seasonal variations in tropospheric NO<sub>2</sub> vertical column density (VCD). High NO<sub>2</sub> VCDs were observed over the oases (farmlands and natural vegetation), with the highest value obtained during summer, and lowest during winter. Pulses were observed during spring. We conducted in-situ measurements in June 2011 in Milan oasis and employed ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) instruments to validate satellite NO<sub>2</sub> retrievals. The findings are as follows: 1) in the study area soil biogenic NO emission is the dominant source of tropospheric NO<sub>2</sub>; 2) oases (farmlands) are hotspots of tropospheric NO<sub>2</sub>, and a higher increase in tropospheric NO<sub>2</sub> is found in oases from winter to summer; and 3) enhancement of soil biogenic NO emission due to soil managements is predictable. Given the rapid agricultural development in the southern Uygur Autonomous Region of Xinjiang, researches on soil biogenic NO emission and increase in tropospheric NO<sub>x</sub> should be given more importance.

Keywords: NO<sub>2</sub> vertical column density; ozone monitoring instrument (OMI); soil biogenic NO emission; arid area; agricultural management

**Citation:** Qi Yue, Spatio-temporal distributions of tropospheric NO<sub>2</sub> over oases in Taklimakan Desert, China. *Chinese Geographical Science*, doi: 10.1007/s11769-014-0696-z

### 1 Introduction

Atmospheric NO<sub>x</sub> (NO and NO<sub>2</sub>) is a key catalyst in the oxidized cycles of atmospheric carbon monoxide, methane and volatile organic compounds. Ambient concentrations determine whether ozone is generated or destroyed in the troposphere (Chameides *et al.*, 1992). Soil biogenic NO<sub>x</sub> emission is estimated to range from 6.6 Tg N/yr to 9.6 Tg N/yr globally, contributing 10%–40% of the total sources (Denman *et al.*, 2007). Soil NO<sub>x</sub> emissions were estimated to be less than 6% of

annual anthropogenic emissions in the eastern China (Lin, 2012). However, in rural (non-industrialized) areas, soil biogenic emissions can be the dominant source. Existing models in which soil sources were not considered have been found to underestimate atmospheric  $NO_x$ in remote areas (Ma *et al.*, 2006). The Taklimakan Desert is located in the southern Uygur Autonomous Region of Xinjiang and has a typical arid climate. Similar to any part in China, Xinjiang has undergone rapid development in recent decades. Reclamation, intensive irrigation and increased fertilizer application may have

Received date: 2013-02-26; accepted date: 2013-05-16

Foundation item: Under the auspices of German Research Foundation and Max Planck Society (No. MA 4798/1-1), National Natural Science Foundation of China (No. 31070384)

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profoundly influenced the regional environment and human lives. Increased soil biogenic NO emission may be attributed to increased agricultural activity. Sufficient soil moisture, high soil temperature, and regular nutrient supply are among the optimal conditions for soil biogenic NO emission. Managed (fertilized) soils in arid area are proven to emit NO twofold to fivefold (Bouwman et al., 2002a; 2002b). Moreover, water-induced pulses of biogenic NO emissions from arid areas have been observed in many studies (Ghude et al., 2010). Increased NO<sub>x</sub> emissions are associated with increased levels of available nitrogen, which promotes biological activity. However, such an increase in available nitrogen also presents negative consequences such as changes in atmospheric composition, alteration of species composition, and ecosystem eutrophication (Galloway et al., 2004).

Arid areas comprise nearly 40% of the total land surface of the Earth (Harrison and Pearce, 2000). The Taklimakan Desert covers an area of  $5 \times 10^5$  km<sup>2</sup> (approximately 6.1% of the arid and hyper-arid land in the world). However, in-situ measured NO emission data from arid areas, particularly from the Taklimakan Desert, are substantially lacking because of the remote location. Therefore, we applied the results of the satellite observation (Ozone Monitoring Instrument, OMI), which provides long-term (2005–2011) daily measurement in pixels down to 13 km × 24 km to examine the spatial distribution and seasonal variation of tropospheric NO<sub>2</sub>. We further investigated the effects of the recent agricultural development on soil biogenic NO emission by analyzing the agricultural statistical data. Given the uncertainties in satellite observations, we applied ground-based Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) measurements in June 2011 to validate the satellite retrievals. This study could provide useful information to study the spatio-temporal variations of the tropospheric NO<sub>2</sub> and its soil sources over arid area.

### 2 Materials and Methods

#### 2.1 Study area

The Taklimakan Desert is located between Tianshan Mountains and the Kunlun Mountains in Uygur Autonomous Region of Xinjiang (Fig. 1a), remote from any ocean (the nearest distance to the North Sea is 2500 km). The geographical location of the desert determines the regional climate as a cold desert climate (BWk), according to Koeppen-Geiger classification (Koeppen, 1931; Mamtimin, 2005). We selected the agriculturally managed Ruoqiang oases, including Waxxari oasis, Ruoqiang oasis, and Milan oasis on the southeastern edge of the Taklimakan Desert, and the area surrounding the oases as our study area (Figs. 1a, 1b) to study the soil sources of tropospheric NO<sub>x</sub>, for that the Ruoqiang oases area is far from other anthropogenic NO<sub>x</sub> sources, with the nearest oases and farms located more than 150 km

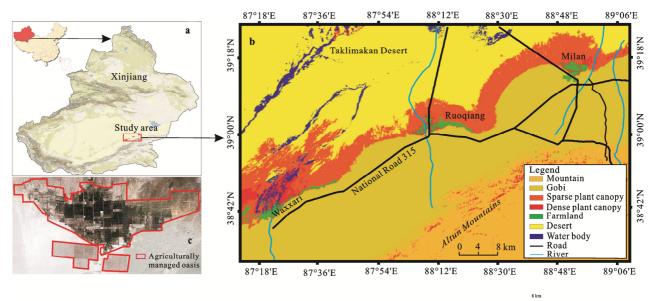


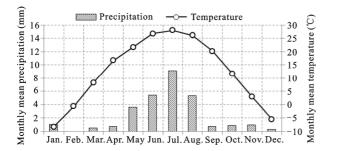
Fig. 1 a, location of study area in Xinjiang; b, land use classification in study area; c, map of Milan oasis

away, where soil biogenic emission is considered the dominant source of tropospheric  $NO_x$ . In-situ observations were carried out in the Milan oasis in June 2011 (Fig. 1c). Monthly mean precipitation and monthly mean air temperature observed at Ruoqiang meteorological station for the last seven years (2005–2011) (China Meteorological Data Sharing Service System, http://www.cma.gov.cn/2011qxfw/2011qsjgx/) were calculated (Fig. 2).

#### 2.2 Satellite retrieved NO<sub>2</sub>

Satellite remote sensing provides data for the vertical column density (VCD) of NO<sub>2</sub> in the troposphere. With a global coverage and a long-term dataset, this technology can be used to examine specific remote arid areas. Satellite retrievals have been rarely used to examine the soil source of NO<sub>x</sub> over arid areas despite arid soil NO emission being an important contributor for atmospheric NO<sub>x</sub>. Challenges hinder the use of the tool: observing very weak atmospheric absorptions of NO<sub>2</sub> over arid area and solely from a soil source is difficult; in addition, given that satellite retrievals are area-average information, the soil source must be separated from other sources that contribute to the column results (Jaegle et al., 2005). Although soils are biogenically emitting NO, the NO emission can be determined by measuring the tropospheric  $NO_2$ , which spans a longer lifetime ranging from hours to days, given its atmospheric fate (NO is rapidly oxidized by ambient  $O_3$  into  $NO_2$ ).

Remote sensing for atmospheric trace gases has recently been developed in spatio-temporal resolution. The OMI was launched onboard the Aura satellite on July 15, 2004. As is one of the most advanced satellite-borne instruments, the OMI provides a dataset of daily tropospheric NO<sub>2</sub> VCD with a spatial resolution down to 13 km  $\times$  24 km at nadir (Levelt *et al.*, 2006).



**Fig. 2** Monthly mean of precipitation and temperature observed at Ruoqiang meteorological station during 2005–2011

The OMI tropospheric  $NO_2$  columns available at www.tenis.nl (DOMINO v2.0) (Boersma *et al.*, 2007; 2011) are used in this study.

Ground-based Multi-Axis Differential Optical 2.3 **Absorption Spectroscopy (MAX-DOAS) observations** Given the relatively coarse spatial resolution and temporal sampling, as well as the known potential uncertainties of satellite observations, satellite data products should be validated to corresponding ground-based observations. Many studies on satellite data validation have been conducted over the eastern China, reporting a significant increase in tropospheric NO<sub>x</sub> especially for the urban area (Chen et al., 2009). However, groundbased observations for satellite data validation over the vast arid area in the western China have rarely been performed. In this study, we compared ground-based MAX-DOAS observation results with satellite retrievals. The in-situ measurement was conducted from June 1 to June 20 in 2011 in the Milan oasis (39°15 'N, 88°53'E).

The mini MAX-DOAS instrument is a fully automated, lightweight spectrometer (13 cm  $\times$  19 cm  $\times$  14 cm) designed for spectral analysis of scattered sunlight (Platt and Stutz, 2008). The obtained spectra are analyzed using the DOAS method, with WinDOAS software (Fayt and van Roozendael, 2001). The spectra analysis results in a differential slant column density (dSCD), calculated as the difference between the SCDs of measured spectrum and that of the Fraunhofer reference spectrum. For most cases, stratospheric absorption is the same in all spectra. Thus we eliminated the stratospheric information by subtracting the dSCD measured at 90° from those measured at small elevation angles (15° and 20°) and determine the DSCD<sub>trop</sub>. Then we applied geometrical approximation of air mass factor (AMF) shown in Equation (1). The VCD is calculated using the geometrical approximation of AMF as shown in Equation (2) (Shaiganfar et al., 2011).

$$4MF_{\alpha} = \frac{1}{\sin(\alpha)} \tag{1}$$

$$VCD_{\alpha} = \frac{DSCD_{\text{trop}}}{DAMF_{\alpha}} = \frac{DSCD_{\text{trop}}}{\frac{1}{\sin(\alpha)} - \frac{1}{\sin(90)}} = \frac{DSCD_{\text{trop}}}{\sin(\alpha)^{-1} - 1}$$
(2)

where  $AMF_{\alpha}$  indicates the air mass factor for a certain observation angle of the MAX-DOAS observation,  $\alpha$ represents the observation angle of the MAX-DOAS instruments, and  $DAMF_{\alpha}$  is the difference between the geometrical approximation of  $AMF_{\alpha}$  and  $AMF_{90}$ . The  $VCD_{\alpha}$  is the vertical column density measured and calculated at the angle of  $\alpha$ , which equals to the results of the  $DSCD_{trop}$  (tropospheric differential slant column density) divided by the  $DAMF_{\alpha}$ .

### **3** Results and Discussion

# 3.1 Spatial distribution of tropospheric NO<sub>2</sub> over Ruoqiang oases

Figure 3 shows the average NO<sub>2</sub> VCD over Ruoqiang oases from 2005–2011 under cloud-free days (cloud fraction < 0.3), and the centers of the three oases are shown in the figure. Compared with the land use classification map (Fig. 1b), high NO<sub>2</sub> VCDs were located over the oases (farmland) and regions covered by natural vegetation, and low values were found over the desert, Gobi desert, and Mountain area. In Ruoqiang County, the industrial facility (Luobupo Potassium Salt Co.) is located 200 km northeast from the study area. Biomass burning has been prohibited since 2009. Therefore, fossil fuel and biomass combustion are not considered as the major sources of atmospheric NO<sub>x</sub>.

# 3.2 Seasonal variations of tropospheric NO<sub>2</sub> over Ruoqiang oases

We calculated the monthly mean values of NO<sub>2</sub> VCDs over Ruoqiang oases which were the highest during summer (June, July, and August) and the lowest during winter (December, January, and February) (Fig. 4). Pulses were observed during spring (March, April, and May). By contrast, the monthly mean values of tropospheric NO<sub>2</sub> VCDs over Urumqi (43°51'N, 87°37'E; the capital city of Xinjiang; a highly polluted urban area) were the highest during winter because of the heating season (when a large amount of fossil fuels is consumed) and the lowest during summer. The observed results of NO<sub>2</sub> VCD are also affected by the atmospheric lifetime of NO<sub>2</sub>, which is longer during winter (low OH concentration) and shorter during warmer seasons. Therefore, in the study area, soil biogenic NO emission is regarded as the most important contributor to tropospheric NO<sub>2</sub>.

The most important factors which influence soil biogenic NO emission include soil moisture, soil temperature, and nutrient content. The soils in the study area are mostly irrigated by the snowmelt runoff from the Altun Mountains. During spring, the conditions for soil biogenic NO emission are improved, with the increase in temperature, snowmelt runoff, and nutrient accumulation during winter, therefore, the 'pulses' occurred. Many field and laboratory experiments have also found pulses of biogenic NO emissions following rain on dry soils. The first rains (irrigation) activate water-stressed nitrifying bacteria, leading to the consumption of accumulated nitrogen and release large amounts of NO as by product. Increased  $NH_4^+$  and C availability caused by death and mineralization of microbial biomass, as well as the accumulation of soil NO<sub>2</sub><sup>-</sup> during the dry season is consistent with occurrence of smaller N-gas pulses following subsequent wettings (Davidson, 1992). In summer, the conditions for soil biogenic NO emission continue to be favorable, and tropospheric NO<sub>2</sub> VCDs

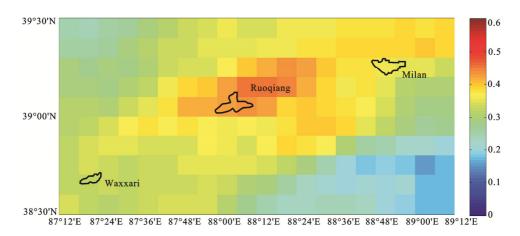
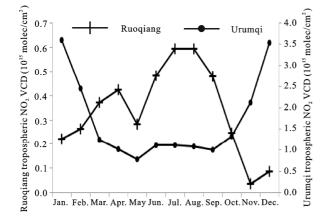


Fig. 3 Tropospheric NO<sub>2</sub> vertical column density (VCD) (10<sup>15</sup> molec/cm<sup>2</sup>) over Ruoqiang oases during 2005–2011

remain high. During winter, the soils are poorly irrigated, and the temperature may decrease to lower than  $0^{\circ}$ C, thereby limiting NO emission from the soil.

Figure 5 shows tropospheric  $NO_2$  VCDs of different seasons over the Ruoqiang oases surrounding desert. Tropospheric  $NO_2$  VCDs are higher in summer than in winter. Meanwhile, the increase in  $NO_2$  VCDs is greater in the oases where agricultural activity is more than in other land-use type areas.

The observed seasonal variation and water-induced enhancement of tropospheric  $NO_2$  columns over Ruoqiang oases are consistent with soil biogenic NO emission. Therefore, soil biogenic NO emission is the dominant source for tropospheric  $NO_2$  in the study area, significantly influenced by soil water and temperature.



**Fig. 4** Seasonal variations of tropospheric NO<sub>2</sub> vertical column density (VCD) over Ruoqiang oases and Urumqi during 2005–2011

# 3.3 Potential effects of agricultural activities on soil NO emissions

The uncertainties in estimates of soil biogenic NO emission mostly result from complex or even unknown emission-related mechanisms and the lack of available measurements. A review by Meixner and Yang (2006) identified only 17 field and laboratory flux measurements of NO from limited locations in arid regions (annual precipitation below 450 mm). Arid and semi-arid soils were found to release considerable amounts of NO as soon as they are wetted. According to Galbally (2008), the six reviewed field studies on NO emissions (with the average emission ranging from  $0.06 \text{ ng}(\text{N})/(\text{m}^2 \cdot \text{s})$  to 3.5 ng(N)/(m<sup>2</sup> \cdot \text{s})) in semi-arid and arid zones indicated that the semi-arid and arid lands could be major contributors to global soil NO emissions (Feig et al., 2008). Soil moisture is a critical factor determining the rate of NO emission (Hartley and Schlesinger, 2000). In arid areas, water-induced soil NO<sub>x</sub> emissions multiplied by the area of arid regions result in large contributions of soil NO<sub>x</sub> emissions to the global budget (Scholes et al., 1997). The results of Jaegle et al. (2004) indicated that in Africa, the enhancement of soil NO<sub>x</sub> emission during the rainy season and the rain-induced pulses lasted from one to three weeks, affecting  $3 \times 10^6$  km<sup>2</sup> of semi-arid sub-Saharan savanna. The microbial activity controlling soil biogenic NO emissions and consumption is influenced by various factors such as soil moisture, soil temperature,

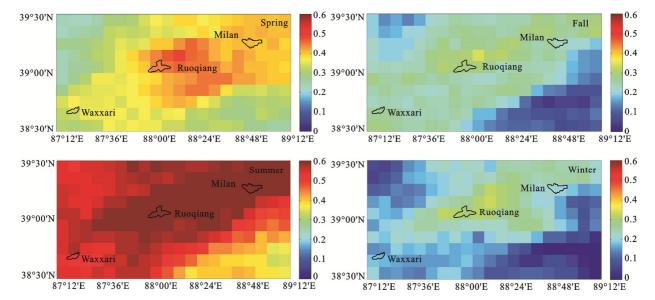


Fig. 5 Tropospheric NO<sub>2</sub> vertical column density (VCD)  $(10^{15} \text{ molec/cm}^2)$  in different seasons over Ruoqiang oases during 2005–2011

and soil nutrient status. These factors are influenced by agricultural activities (Hudman *et al.*, 2010).

According to the results from Gu and He (2004), 90% of the oases area in the southern Xinjiang are agriculturally managed. In Ruogiang County, cotton is one of the three primary crops (the other two are wheat and corn), and Ruoqiang County is also famous for the jujube fruit products. Since 2005, the agricultural GDP (gross domestic product) has increased by 1090.15% from  $2339.51 \times 10^4$  USD to  $27843.72 \times 10^4$  USD (Table 1). The increases can be attributed to the enlargement of sown area and efficient management (irrigation and fertilization). According to the results in statistical vearbook of Xinjiang during the period of 2005–2011, the sown area in Ruoqiang oases was enlarged by 229.71%, from  $3.71 \times 10^3$  ha to  $12.21 \times 10^3$  ha. The irrigated area was enlarged by 142.33%, from  $3.00 \times 10^3$ ha to  $7.27 \times 10^3$  ha. The amount of N-fertilizer application in Ruoqiang oases was increased by 277.42%, from 886.00 t to 3344.00 t.

Assuming that soils in arable lands exhibit the same NO emission rate, the enlargement of sown area increased the soil biogenic NO emission by about 229.71%. Furthermore, in arid areas irrigation provides favorable conditions for soil biogenic NO emission. The 'pulses' of soil NO emission observed globally are caused by natural or anthropogenic irrigation. Despite the difficulty in quantifying the impact of irrigation, an enlargement of 142.33% of irrigation area has undoubtedly activated microbes and increased NO emission from the soils. Nitrogen fertilizers are intensely applied in the study area (Ma and Zhou, 2012). Soil nutrients are important for nitrification which releases NO as a by-product. According to state-of-the-art studies on the relationship between nutrient application and

**Table 1** Agriculture development in Ruoqiang County from2005 to 2011

Year	Agriculture GDP (10 <sup>4</sup> USD)	Sown area $(10^3 \text{ ha})$	Irrigated area (10 <sup>3</sup> ha)	N-fertilizer application (t)
2005	2339.51	3.71	3.00	886.00
2006	3243.48	4.59	3.02	824.00
2007	4825.03	6.46	3.67	901.00
2008	5731.73	11.34	3.67	1748.00
2009	9083.19	11.41	5.94	2266.00
2010	12630.08	10.58	7.25	2864.00
2011	27843.72	12.21	7.27	3344.00

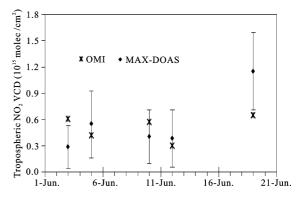
soil NO emission, at most 0.2% of the total available N in soil is emitted into the atmosphere by nitrification as  $N_2O$  and NO.

#### 3.4 Validation of satellite data

As mentioned previously, satellite data need to be validated. Daily average NO<sub>2</sub> VCDs observed by ground-based MAX-DOAS in Milan oasis in June 2011 were compared with OMI retrievals. For satellite-retrieved NO<sub>2</sub> VCDs, we selected the results with a pixel center within the area located between 39°00'-39°30'N and 88°36'-89°12'E. The OMI instrument passes by the study area approximately at 13:00 daily. We retain the satellite retrievals from cloud-free days as in section 3.1. Therefore, some of the observations had to be skipped. By comparing the two datasets, we obtained five days with both satellite and ground-based measurements, and the comparison results are shown in Fig. 6. We compared the averaged MAX-DOAS measured NO<sub>2</sub> VCDs with the average of satellite retrievals. The comparisons show that the results of ground-based and satellite-based observations were of the same magnitude and mostly in the same range. Thus the results for ground-based and satellite measured tropospheric NO<sub>2</sub> were generally in agreement.

### 4 Conclusions

The following conclusions are arrived based on the OMI satellite-observed tropospheric  $NO_2$  VCDs over the Ruoqiang oases in the Taklimakan Desert: 1) the seasonal variations in tropospheric  $NO_2$  (highest during summer and lowest during winter) are consistent with soil biogenic NO emission. Compared with the seasonal



**Fig. 6** Comparison between satellite (OMI) and groun-based (MAX-DOAS) measurements of NO<sub>2</sub> vertical column density (VCD)

pattern of polluted urban area, soil biogenic NO emission is the most important contributor for tropospheric  $NO_2$  in the oases of Taklimakan Desert; 2) as the center of human activities and a relatively wetter area in the desert, oases are hotspots of tropospheric  $NO_x$ ; 3) pulses of soil biogenic NO emissions are observed during spring, induced by the increase in irrigation resulting from precipitation and snowmelt runoff from the Altun Mountains; and 4) the increase in tropospheric NO<sub>2</sub> is greater in the oases (farmland) than in other areas. We applied ground-based MAX-DOAS observation of tropospheric NO2 in Milan oasis and compared the daily average results with the satellite data. The consistency between the satellite and the ground-based measurements at least during summer strengthened the remote sensing results.

The findings of this study are expected to facilitate long-term monitoring of atmospheric substances for further investigations. Atmospheric NO<sub>x</sub> has an important function in chemical reactions and can harm humans. The OMI NO<sub>2</sub> columns over Ruogiang oases indicate that soil biogenic NO emission is the dominant source, which is affected by soil management and agricultural activities. According to state-of-the-art studies on the relationship between nutrient application and soil NO emission, the enhancement of tropospheric NO<sub>2</sub> originating from soil biogenic NO emission can be predicted. A significant enlargement in sown and irrigated areas, as well as increase in nitrogen fertilizer application, was reported for the period of 2005-2011. Reclamation, irrigation, and fertilization influence soil biogenic NO emissions. Although the oases (farmland) occupy a small proportion of land relative to the vase desert, almost all human necessities are produced in the area. Given the ongoing agricultural development in the oasis chain around the Taklimakan Desert, soil biogenic NO emission may increase significantly further affecting local/regional atmospheric chemistry and human health. Therefore, biogenic NO emission form arid soil and the increase in tropospheric NO<sub>2</sub> over the oases area need to be further investigated for further knowledge and human health concerns.

### Acknowledgments

The author would like to thank Dr. Stefen. Beirle, Prof. Tagner. Wagner, Prof. Franz X. Meixner, and Dr. Buhalqem. Mamtimin of the Max Planck Institute for Chemistry for the advising and help. We acknowledge the free use of tropospheric  $NO_2$  column data from the *YYY* sensor from www.temis.nl.

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