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Characteristics of surface O_3 over Qinghai Lake area in Northeast Tibetan Plateau, China

Zhenxing Shen [a,b,*], Junji Cao [b], Leiming Zhang [c], Zhuzi Zhao [b], Jungang Dong [d], Linqing Wang [a], Qiyuan Wang [b], Guohui Li [b], Suixin Liu [b], Qian Zhang [a]

[a] Department of Environmental Sciences and Engineering, Xi'an Jiaotong University, Xi'an, China
[b] Key Lab of Aerosol, SKLLQG, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an, China
[c] Air Quality Research Division, Environment Canada, Toronto, Canada
[d] School of Architecture, Xi'an University of Architecture and Technology, Xi'an 710055, China

HIGHLIGHTS

- Surface O_3 was measured in Qinghai Lake area in Northeast Tibetan Plateau, China.
- The O_3 chemical formation was under a strong NOx-limited in Qinghai Lake areas.
- Stratospheric O_3 and transport might be the main sources of O_3 in this area.

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ABSTRACT

Surface O_3 was monitored continuously during Aug. 12, 2010 to Jul. 21, 2011 at a high elevation site (3200 m above sea level) in Qinghai Lake area (36°58′37″N, 99°53′56″E) in Northeast Tibetan Plateau, China. Daily average O_3 ranged from 21.8 ppbv to 65.3 ppbv with an annual average of 41.0 ppbv. Seasonal average of O_3 followed a decreasing order of summer > autumn > spring > winter. Diurnal variations of O_3 showed low concentrations during daytime and high concentrations during late night and early morning. An intensive campaign was also conducted during Aug. 13–31, 2010 to investigate correlations between meteorological or chemical conditions and O_3. It was found that O_3 was poorly correlated with solar radiation due to the insufficient NO_x in the ambient air, thus limiting O_3 formation under strong solar radiation. In contrast, high O_3 levels always coincided with strong winds, suggesting that stratospheric O_3 and long range transport might be the main sources of O_3 in this rural area. Back-trajectory analysis supported this hypothesis and further indicated the transport of air masses from northwest, northeast and southeast directions.

1. Introduction

Tropospheric O_3 has attracted extensive attention due to its important roles in influencing air quality, climate change and ecosystem health (Thompson et al., 2001; Vingarzan, 2004). O_3 is a critical photochemical oxidant in the troposphere and affects atmospheric chemistry and air quality. Excessive surface O_3 could cause adverse effects on human health, vegetation and materials (De, 2000; Coyle et al., 2003; Fischer et al., 2004). O_3 is also one of the important greenhouse gases directly contributing to climate change due to its absorption of the earth's infrared radiation at 9.6 μm. Hence, it is necessary to evaluate O_3 levels in urban and rural areas in order to improve air quality and protect humans and wildlife from adverse effects.

Many studies have been conducted around the world to understand the formation, accumulation, transportation and deposition of tropospheric O_3 (Nicolas and Dalstein, 2005; Cristofanelli et al., 2006, 2010; Zhang et al., 2006; Reddy et al., 2010; Zheng et al., 2010; Shan et al., 2010; Sicard et al., 2011). High O_3 concentrations are often associated with intense solar radiation, high temperature, stagnant air, and minimum rainfall. These conditions are favorable for the photochemical production of ozone and accumulation of pollutants in the atmospheric boundary layer. Ozone has an atmospheric lifetime from hours to several days and thus can be transported over long distances. High O_3 concentrations were observed within a metropolis and downwind locations due to high O_3-precursor emissions in urban areas (Hastie et al., 1999; Brankov et al., 2003). Ozone precursors can also be transported over long distances, resulting in O_3 formation far from the sources.

The Tibetan Plateau is one of the most environmental and climate sensitive areas in the world. Studies concerning this area were mostly

* Corresponding author at: Department of Environmental Sciences and Engineering, Xi'an Jiaotong University, Xi'an, China.
E-mail address: zxshen@mail.xjtu.edu.cn (Z. Shen).

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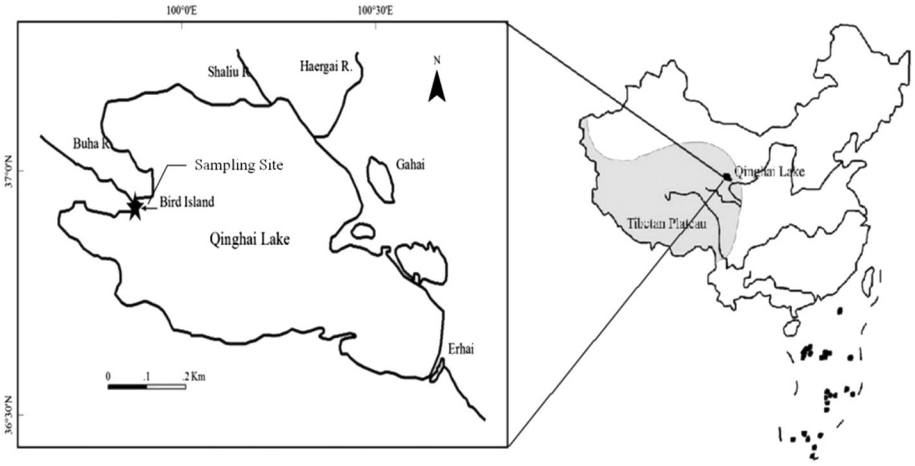

Fig. 1. Location of the sampling site.

focused on chemical compositions of ice cores and snow (Xu et al., 2009; Liu et al., 2008). Very limited effort has been allocated to understand aerosol and trace gas levels in this region. For example, at a background site located in the northeast of the Tibetan Plateau, increasing anthropogenic emissions have led to high levels of air pollution in the past few years (Zhang, 2009; Xue et al., 2011; Zhao et al., 2013). Qinghai Lake is also located in the northeast of the Tibetan Plateau. It is situated in the sensitive semi-arid zone where the Asia summer monsoon and the Westerlies influence the area. As a popular resort in summer, it faces rapid tourism expansion and development. There is a need to better understand the pollution sources and formation mechanisms in this area. This study aims to understand the current O_3 levels and its major sources in this area through analysis of nearly one year continuous O_3 data and one month intensive campaign data.

2. Methodology

2.1. O_3 measurements

Measurement instruments were installed on the roof of a 13-m high building in the Qinghai Lake rural site monitoring station (36°58′37″N, 99°53′56″E, and 3200 m above sea level) (Fig. 1). The UV photometric O_3 analyzer (Model ML/EC9810, Ecotech Pty Ltd, Australia) was used to continuously monitor surface O_3 every 5 min from Aug 12, 2010 to July 21, 2011. During the intensive campaign in August 2010, NO_x (EC9841B Analyzer, Ecotech Pty Ltd, Australia, with a detection limit of 0.02 ppb), total VOCs (non-methane hydrocarbon (NMHC), TVOCs, RAE Systems Inc., CA, USA), and meteorological variables (temperature, relative humidity, wind speed, and wind direction, Series Model 232, Weatherhawk, United States) were also recorded every 5 min to gain some insight of the controlling factors of surface O_3.

Zero checks were performed every day by automatically injecting charcoal-scrubbed air. The nearly one-year monitored O_3 concentration data (except for the periods of network maintenance and upgrading) was analyzed to characterize the ground-level O_3 at the Qinghai Lake rural site. Data was not collected for 21 days over the entire sampling period due to the upgrade and maintenance of the O_3 analyzer.

2.2. Back trajectory analysis

Air mass back-trajectory analysis is a useful tool to identify possible source regions and transport pathways of air pollutants. In this study, 96-h air mass back-trajectory was calculated using the NOAA HYSPLIT 4 trajectory model to trace the source and transport pathways of O_3 or its precursors.

3. Results and discussion

3.1. Overview of surface O_3 levels

Fig. 2 shows the daily O_3 distributions at the Qinghai Lake site. Daily values ranged from 21.8 to 65.3 ppbv with an annual average of 41.0 ± 8.4 ppbv. O_3 concentrations did not exceed the Chinese National Air Quality Standard Grade 2 (hourly average concentration 102 ppbv) on any day during the observation period. 13.8% of the observation days were above the WHO 8-hour mean concentration guideline in urban environments, which is 51.0 ppbv (WHO, 2005). The daily maximum value occurred in summer (Aug. 26, 2010), and the minimum in winter (Feb. 8, 2011). The mean O_3 level in Qinghai Lake area was comparable

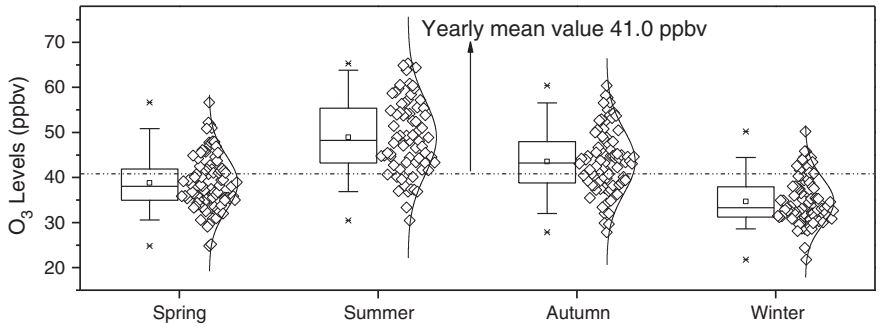

Fig. 2. Daily average O_3 mixing ratios in the four seasons with whisker–box plot on the left and diamond symbols on the right. The whisker–box plot represents 5th, 25th, median, 75th, and 95th percentile values. The small pane inside the box represents the average and the points outside the whisker represent maximum and minimum values. Each diamond symbol represents one data point.

Table 1
Ozone levels at different sites.

Location	Site description	Observation period	O_3 levels	Reference
			Mean (ppbv)	
Qinghai Lake, China	Rural site	Aug. 2010–July. 2011	41.0	This study
Waliguan, China	Remote mountain site	Aug. 1994–Dec. 2001	48.0 ppb	Nie et al. (2004)
Waliguan, China	Remote mountain site	Apr–May 2003	58	Wang et al. (2006)
		Jul–Aug 2003	54	
Waliguan, China	Remote mountain site	Summer in 2006	59	Xue et al. (2011)
Shangdianzi, China	Rural site	2004–2006	31.5 ppb	Lin et al. (2008)
Huizhou, China	Rural site	Jan. 2006–Dec. 2007	36	Zheng et al. (2010)
Anantapur, India	Rural site	Dec. 2008–July 2009	45.9	Reddy et al. (2010)
Southern France	Rural site	Summers in 2001 and 2002	39.6 ppb	Dalstein and Nicolas (2005)
Mt. Cimone, Italia	Rural site	1998–2003	54 ppbv	Cristofanelli et al. (2006)
Pyramid, Nepal	Remote mountain site	2006–2008	49 ppbv	Cristofanelli et al. (2010)

to the values observed in previous studies at various locations in China and around the world, including mountain sites (Table 1). For example, the mean O_3 at this site was slightly lower than those measured at a remote site of Waliguan, China (Nie et al., 2004; Wang et al., 2006; Xue et al., 2011), rural or remote mountain sites outside China, such as Anantapur in India (Reddy et al., 2010), the Pyramid International Observatory in Nepal (Cristofanelli et al., 2010), and Mt. Cimone in Italy (Cristofanelli et al., 2006). In contrast, the mean O_3 at this site was higher than those measured at a background site of Shangdianzi in Beijing, China (Lin et al., 2008), a rural site of Huizhou in Pearl River Delta (PRD), China (Zheng et al., 2010), and a rural site in Southern France (Dalstein et al., 2005).

Seasonal variations of O_3 depend on many factors and are certainly site-dependent. O_3 levels in this study followed a decreasing order of summer (48.9 ± 8.2 ppbv) > autumn (43.6 ± 7.0 ppbv) > spring (38.8 ± 6.0 ppbv) > winter (34.7 ± 5.2 ppbv). This was similar to what was observed at Waliguan, also a remote mountain site in northeastern Qinghai–Tibetan Plateau (Tang et al., 1995), but was different from the rural sites of Shangdianzi, a background site in North China plain (Lin et al., 2008), and Linan, a rural site in eastern China (Wang et al., 2001). The latter showed higher O_3 levels in spring than in summer. Shangdianzi and Linan sites are relatively close to large cities (Beijing, Shanghai, and Hangzhou) having high emissions of O_3 precursors, while Qinghai Lake area and Waliguan are far from urban areas. Photochemical productions of O_3 at Shangdianzi and Linan should differ significantly from those in Qinghai Lake area and at Waliguan. Other seasonal variations included a trend of autumn > spring > winter > summer at a rural site in Pearl River Delta region (Zheng et al., 2010), spring (pre-monsoon) maximum and summer (monsoon) minimum at the Pyramid Observatory in Nepal (Cristofanelli et al., 2010), and summer maximum and spring minimum at Mt. Cimone in Italy (Cristofanelli et al., 2006). O_3 at the abovementioned two mountain sites were demonstrated to be influenced heavily by stratospheric intrusions.

3.2. Diurnal variations of surface O_3

A unimodal distribution was found for O_3 concentrations in all seasons at the Qinghai Lake monitoring site (Fig. 3). In summer months except in August, a daily minimum of O_3 appeared around 10:00 (Beijing time, BJT) in the morning; O_3 concentration then increased until 22:00 before starting to decrease over night and in the early morning. In contrast, daily minimum and maximum of O_3 appeared at around 12:00 BJT and 19:00 BJT, respectively, in winter, and at around 11:00 BJT and 21:00 BJT, respectively, in spring. The different times when daily minimum and maximum of O_3 appeared should be partly

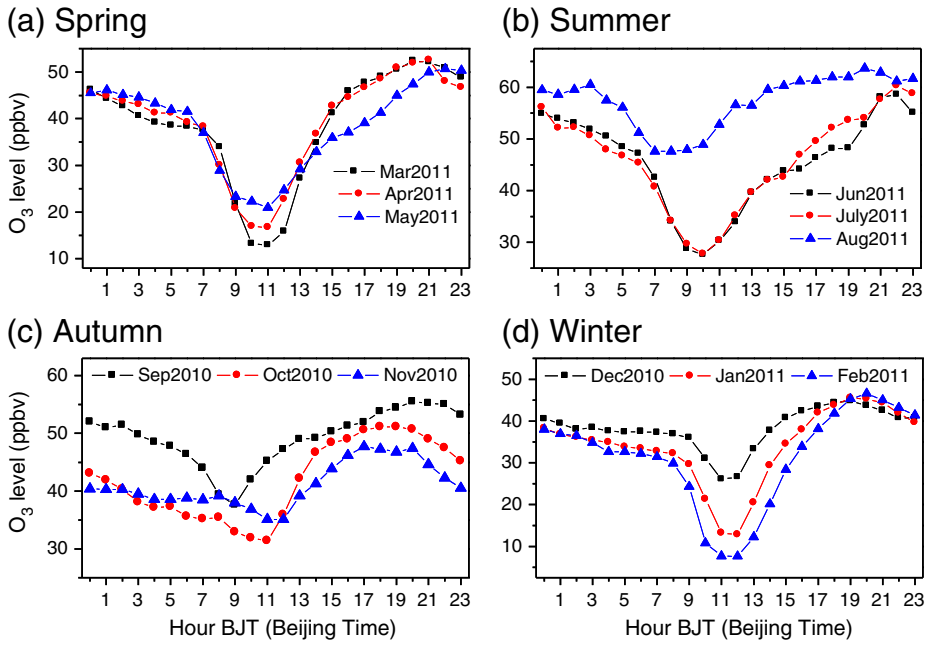

Fig. 3. Diurnal cycle of O_3 concentration in Qinghai Lake area.

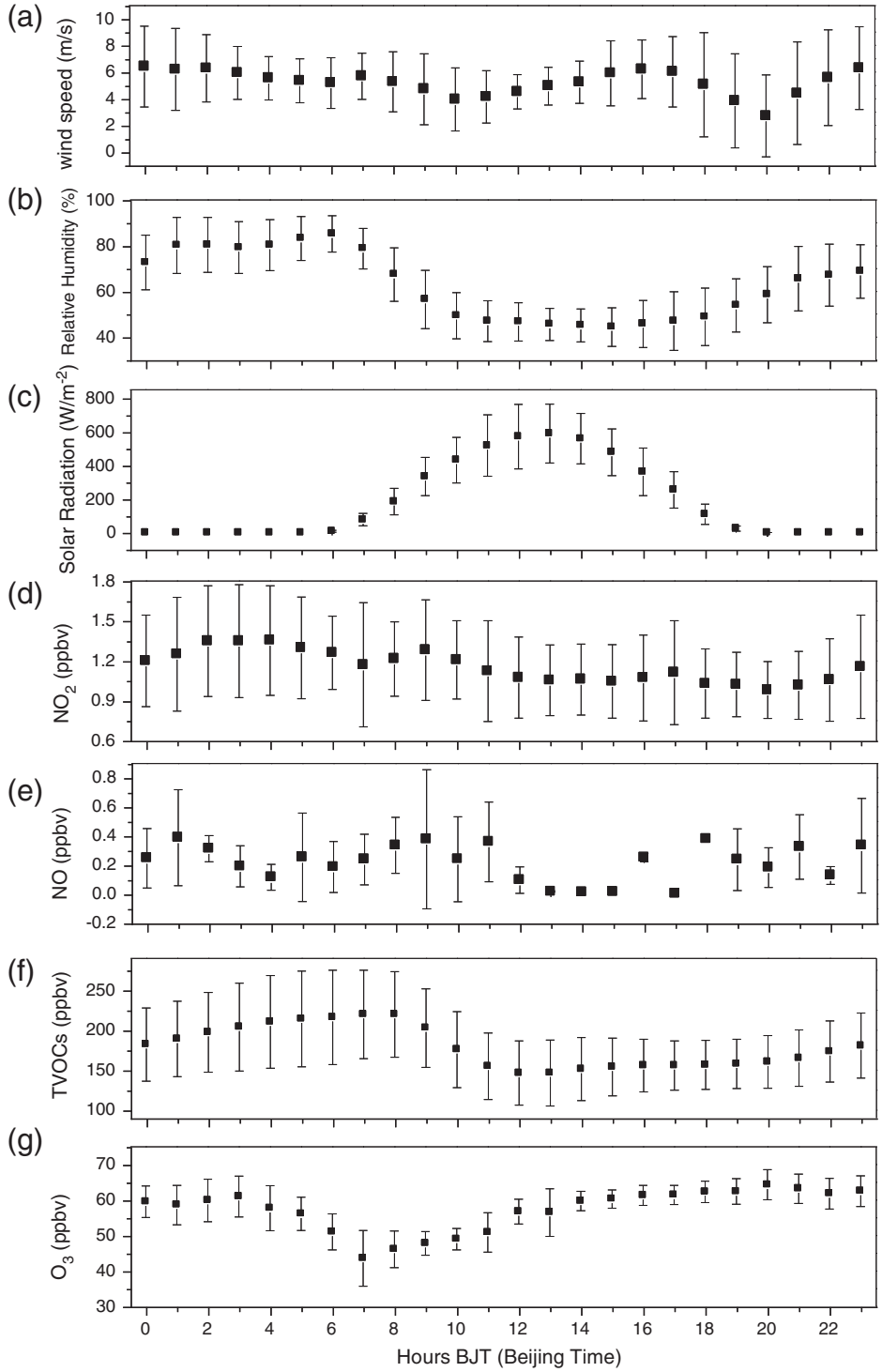

Fig. 4. Diurnal cycle of wind speed (a), RH (b), solar radiation (c), NO_2 (d), NO (e), TVOCs (f), and O_3 levels (g) in Qinghai Lake area during the intensive campaign period.

caused by the different seasonal sunrise and sunset times. Note that Beijing time is used all over China and there is a ~16° difference in longitude between Beijing and the Qinghai Lake area. Sunrise and sunset times at the Qinghai Lake area are at 6:25 and 20:29, respectively, on Jul. 31, and are at 8:33 and 18:14, respectively on Dec. 31. It is noticed that O_3 levels were not rising with the increase of solar radiation in the early morning (between sunrise and 10:00), and the O_3 peak did not coincide with the strongest solar intensity (at about 13:00 BJT), suggesting that solar radiation was not the controlling factor for O_3 formation in this area. The lower morning values might be due to the depositional losses in stagnant air during the nighttime and early morning hours, and its enhancement in the late afternoon and night could be due to downward transport of the free troposphere air (Fischer et al., 1998). The diurnal variations of O_3 in spring and autumn were similar to that in the summer except the 1 to 3 h shift when minimum and maximum appeared, which should be caused by the different hours of daylight in different seasons.

Similar to the seasonal patterns discussed above, diurnal patterns are also affected by many factors and are site-dependent. Similarities and differences in the diurnal patterns between this and the other

Table 2
Concentration of gases and meteorological factors during the intensive campaign period.

	O_3	VOC	NO	NO_2	Solar radiation	Wind speed	RH
		ppbv			wm^{-2}	ms^{-1}	%
Ave	57.4	178.7	0.2	1.2	188.5	5.2	61.8
Max	65.3	344.6	1.5	2.4	726.1	14.0	100
Min	44.5	63.8	0.001	0.03	0	0	23.0

sites both exist. The diurnal patterns of O_3 in Qinghai Lake area were similar to those observed at Waliguan in spring and summer, which showed a unimodal diurnal variation with a minimum during 8:00–10:00 BJT and a peak at around 18:00 BJT (Wang et al., 2006). At Mt. Cimone in Italy, summer O_3 had a morning minimum and an early evening maximum (Cristofanelli et al., 2007), similar to what was observed in Qinghai Lake area. Another study at a site 5000 m above sea level on Mt. Everest, which is the highest mountain in the world located at the southern edge of the Tibetan Plateau, also revealed high O_3 concentrations from late afternoon to midnight (Zhu et al., 2006). However, at two rural sites, Shangdianzi and Huizhou, O_3 concentrations increased from its minimum in early morning (at 6:00 BJT) to its maximum in mid-afternoon (around 16:00 BJT), and then decreased until next morning (Lin et al., 2008; Zheng et al., 2010). This pattern was different from that observed at the Qinghai Lake site, but similar to that observed at an urban site (Wang et al., 2012). Many other studies observed diurnal patterns that were remarkably different from those observed in this study, e.g., high concentrations during daytime (mid-afternoon) and low concentrations during early morning hours at Anantapur, India (Reddy et al., 2010), and a mid-afternoon minimum and a night-time maximum in winter, no apparent diurnal variation in pre-monsoon season, a late morning O_3 maximum and a minimum in the early evening in summer monsoon, and a morning maximum and a late afternoon minimum in the post-monsoon season at the Pyramid Observatory in Nepal (Cristofanelli et al., 2010). Various sources and formation mechanisms caused the different diurnal patterns at different locations.

3.3. Meteorological conditions and chemical conditions affecting O_3 formation

O_3 forms photochemically in the presence of NO_x and VOCs. Solar radiation is thus frequently the dominant meteorological factor in O_3 production. Water vapor is also needed to produce O – H bonds, which oxidize hydrocarbons to produce peroxy radicals, RO_2 (where R is a hydrogen atom or carbon-containing fragment). In the presence of NO_x, the RO_2 radicals convert NO to NO_2; subsequent photolysis of NO_2 yields O_3 (Wennberg and Dabdub, 2008). O_3 production rates can be characterized as either NO_x-sensitive or VOC-sensitive regimes. The sensitivity of O_3 formation to NO_x and VOC concentrations is frequently measured using the ratio of $TVOC_s/NO_x$. The chemical reaction rate constant between NO_2 and OH is $1.7 * 10^{10}$ in minutes and between VOCs and OH, $3.1 * 10^9$ min^{-1}. Thus, the threshold value of $TVOC_s/NO_x$ ratio is estimated to be 5.5. Below this threshold O_3 production is VOC sensitive or limited, and vice versa (Tang et al., 2010). It should be noted that the estimated threshold value could range from 5.0 to 10.0 (Tie et al., 2013). The VOC_s is referred as non-methane hydrocarbon (NMHC), including oxidation of volatile organic compounds (OVOC) and biogenic VOC_s. Considering that the sampling site is located in a remote area of Northeast Tibetan Plateau, the $TVOC_s$ in Qinghai Lake should be mainly from biogenic VOC_s. To better understand the effects

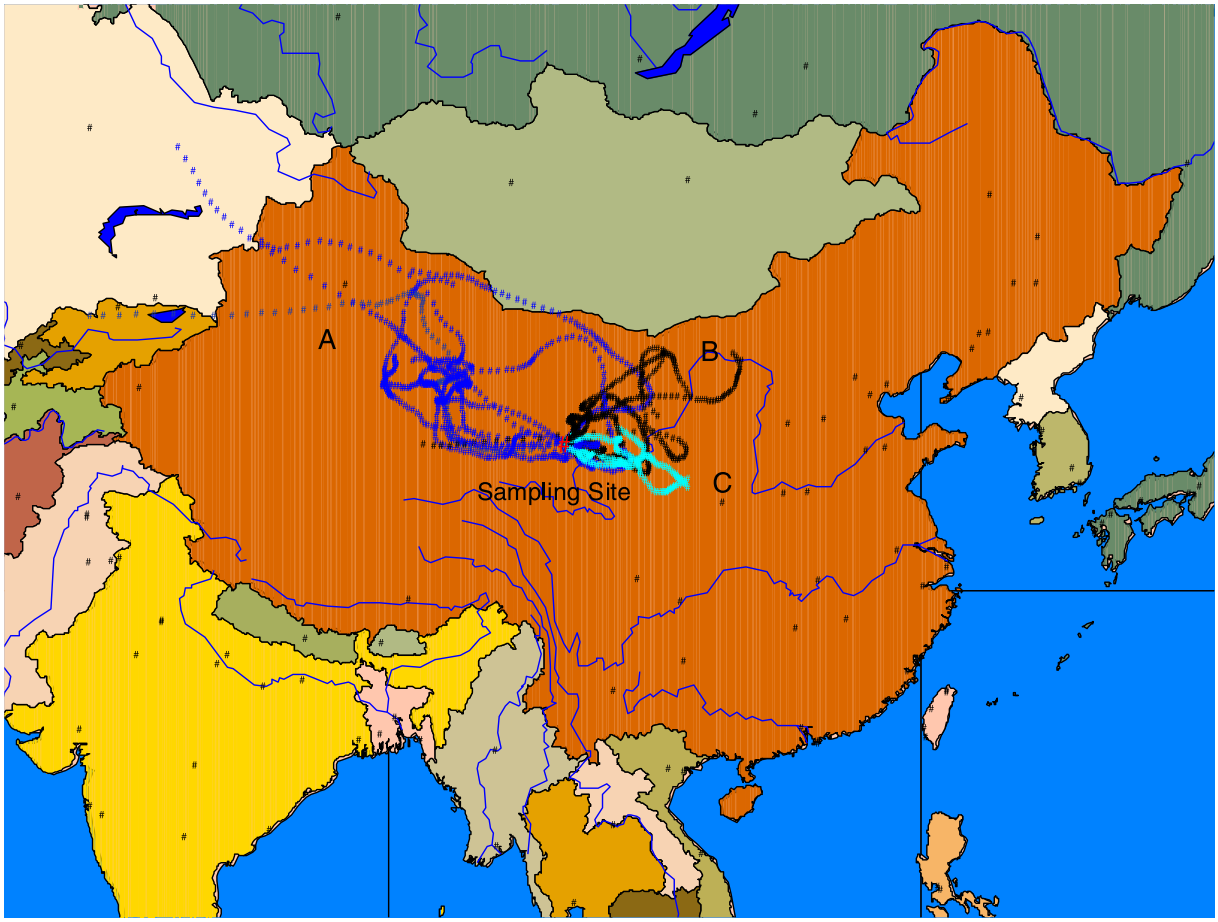

Fig. 5. Backward trajectories of air masses for two typical transport pathways during the intensive campaign period.

of local meteorological conditions and chemical precursors mentioned above on surface O_3 levels, an intensive campaign was conducted during Aug. 12–31, 2011. Fig. 4 shows the diurnal cycles of O_3, solar radiation, RH, $TVOC_s$, NO, and NO_2. The maximum and minimum hourly mean values of these variables are summarized in Table 2.

Prior studies suggested that solar radiation was one of several important factors influencing O_3 production, and the highest O_3 levels frequently coincided with the highest solar radiation (e.g., Wang et al., 2012). In this study, however, a different pattern was observed. For example, during this particular period (Aug. 12–31, 2011) O_3 concentrations increased from 7:00 BJT along with increasing solar radiation; the solar radiation peaked at 13:00 BJT and then decreased, while the O_3 levels continued increasing and peaked at 20:00. O_3 was not correlated with solar radiation during the observation period ($R = -0.27$, $P = 0.2$). The mean RH was 61.3% during the intensive campaign period with hourly values and ranged from 23.0% to nearly 100%. This indicated that water vapor should not be a limiting factor in the O_3 production process. No correlation was found between RH and O_3 ($R = -0.13$, $P = 0.53$).

The campaign-average concentrations of O_3, NO, NO_2, and $TVOC_s$ were 29.5 ppbv, 0.2 ppbv, 1.2 ppbv, and 178.7 ppbv, respectively. The $TVOC_s/NO_x$ ratio was 127.8 on average, with hourly values ranging from 101.9 to 151.9. The very high ratio of $TVOC_s/NO_x$ suggests that the O_3 chemical formation was under a strong NO_x-limited regime in the Qinghai Lake areas. When sufficient amount of NO is available, the chemistry is catalytic. That is, for each OH produced from water, many hydrocarbons can be oxidized and large amounts of O_3 produced. However, insufficient NO_x was observed in atmosphere in the Qinghai Lake area due to limited human activities. This suggests that the episodic O_3 concentration was likely not produced from local photochemical reaction processes. Thus, the intrusion of stratospheric O_3 and long range transport of O_3 could be the main factors, as discussed further below.

3.4. Transport pathways of surface O_3 or its precursors

High wind speed was frequently observed during the intensive campaign with a mean value of 5.2 ms^{-1}. The net role of wind speed on surface O_3 should be location and time dependent. For example, high wind speed can increase dilution of primary pollutants and thus decrease O_3 production in urban regions (Xu et al., 1996; Dueñas et al., 2002). Similarly, strong winds can transport O_3 and its precursors from their sources to remote areas and thus increase O_3 at remote locations. At elevated sites, strong winds might also increase the exchange between the surface and the above levels (e.g., upper troposphere and stratosphere) and led to increase of surface O_3. Data collected in this study suggested that high levels of O_3 were accompanied with strong winds during both daytime and nighttime. Thus, it is very likely that the long range transport and/or exchange between troposphere and stratosphere played important roles in the high O_3 concentrations at the surface of this high-elevation location.

96-h air mass back-trajectories arriving at 500 m above ground level at 12:00 UTC (20:00 BJT, O_3 maximum) were calculated for the Qinghai Lake station using the NOAA HYSPLIT 4 trajectory model to investigate the transport pathways and origins of surface O_3 and its precursors. Three different types of air mass trajectories (A, B and C) were seen for the 19-day campaign in August 2010 (Fig. 5). Type A trajectories were typical air masses from northwest passing through the northern part of Tibetan Plateau. 53% of the campaign days had the type A trajectories. The route represented by group B came from northeast; 31% (or six days) of the campaign period belonged to this pathway. Type C pathway showed that the air mass came from southeast regions of Qinghai Lake area; 16% (three days) of the campaign period belonged to this category.

Previous studies showed that O_3-rich air masses could be transported from the stratosphere to the troposphere (Zhu et al., 2006; Xu et al., 2008; Pont and Fontan, 2001). During the intensive campaign, six of the 19 days of the 96-h air mass back-trajectories were up to 2500 m, suggesting the potential of vertical transport of O_3 from upper levels down to the surface. However, the air masses in the rest of the days were lower than 500 m and the transport routes were also short in inland China. Thus, low-level transport of O_3 from nearby areas was dominant in this area. In contrast, O_3 at Waliguan site was mostly transported from long distance, such as Europe (mostly from middle troposphere), central Asia, Siberia/Mongolia, and Indian subcontinent, but only 12% transport pathways were from southern China (Wang et al., 2006). This comparison also highlighted that the formation of O_3 was evidently different for the Waliguan and the Qinghai Lake site.

4. Conclusions

O_3 data from September 2010 to August 2011 at a rural site in Qinghai Lake area in Northeast Tibetan Plateau, China was analyzed. The O_3 levels were lower than most measurements reported in literature including those collected at rural background or high elevation sites. Seasonal variations of surface O_3 showed the highest concentrations occurred in summer and the lowest in winter. The correlations between O_3 and meteorological factors or chemical conditions highlighted that local production of O_3 through photochemical reactions was not a dominant source in Qinghai Lake area. Air mass back-trajectory analysis revealed that air mass rich O_3 from northwest and northeast was the major contributor to the surface O_3 at this site and the intrusion of stratospheric air also played an important role. Local photochemical production of O_3 was severely limited by the low concentration of NO_x. Thus, a small increase in anthropogenic emissions of NO_x could elevate O_3 levels significantly due to the very high concentrations of TVOCs in this region. The results have important implications in the effective control of anthropogenic emissions of NO_x during economic development in this location and nearby regions.

Acknowledgments

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