Characteristics of Air Pollutants and Greenhouse Gases at a Regional Background Station in Southwestern China

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ABSTRACT

The characteristics of air pollutants and greenhouse gases at regional background sites are critical to assessing the impact of anthropogenic emissions on the atmospheric environment, ecosystems and climate change. However, observational studies are still scarce at such background sites. In this study, continuous hourly observations of air pollutants (O_3 , CO , SO_2 , NO_x , $\text{PM}_{2.5}$ and PM_{10}) and greenhouse gases (CO_2 , CH_4 and N_2O) were performed for one year (from January 1 to December 31, 2017) at the Gongga Mountain background station (GGs; $101^\circ 97'\text{E}$, $29^\circ 55'\text{N}$; elevation: 3541 m) in southwestern China. The concentrations and variations of these air pollutants and greenhouse gases were determined, and the effect of transboundary atmospheric transport on the air pollution at the study site was investigated. The results showed that the average annual concentrations (mixing ratios) of the O_3 , CO , SO_2 , NO_2 , CO_2 , CH_4 , N_2O , $\text{PM}_{2.5}$ and PM_{10} were $74.7 \pm 22.0 \mu\text{g m}^{-3}$, $0.3 \pm 0.2 \text{ mg m}^{-3}$, $0.5 \pm 0.6 \mu\text{g m}^{-3}$, $1.7 \pm 1.3 \mu\text{g m}^{-3}$, $406.1 \pm 9.5 \text{ ppm}$, $1.941 \pm 0.071 \text{ ppm}$, $324.5 \pm 14.8 \text{ ppb}$, $6.5 \pm 6.2 \mu\text{g m}^{-3}$ and $10.6 \pm 11.2 \mu\text{g m}^{-3}$, respectively. The concentrations (mixing ratios) of the abovementioned substances at the GGS are comparable to those at other background sites in China and around the world. The slight differences among concentrations at different sites may be mainly attributable to the impacts of anthropogenic emissions near the background sites and meteorological conditions. High values of O_3 were observed in spring and summer, while SO_2 and $\text{PM}_{2.5}$ showed higher concentrations in summer than in autumn. Relatively high CO , NO_2 and PM_{10} values were mostly observed in spring and winter. Relatively low CO_2 concentrations were observed in summer due to the vigorous summertime photosynthesis of vegetation. The lowest concentrations for CH_4 were recorded in summer, whereas the levels in the other three seasons were similar to each other; by contrast, the highest N_2O concentrations were observed in summer due to enhanced microbial activity resulting from high ambient summer temperatures. A diurnal variation in O_3 was observed, with early morning minima and afternoon maxima. CO and NO_2 displayed higher concentrations in the daytime than in the nighttime. A slight increase in both $\text{PM}_{2.5}$ and PM_{10} concentrations was also recorded in the daytime. These patterns were closely related to scattered anthropogenic emissions and regional atmospheric transport. Nevertheless, CO_2 exhibited lower concentrations in the daytime than in the nighttime, although CH_4 showed no obvious diurnal variation. The N_2O concentration peaked between 10:00 and 12:00 (local time), which can be ascribed to the enhancement of microbial activity due to the increased soil temperature. The results based on the relationship between the wind and the concentrations of air pollutants and greenhouse gases were almost consistent with those based on the potential contribution source function. It appears that O_3 and its precursors in parts of Inner

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Mongolia and Gansu, Ningxia, Sichuan, Chongqing and Hubei Provinces as well as adjacent areas of Hunan, Guizhou and Guangxi Provinces contributed to the increase in O_3 at the study site. The potential source areas for CO and SO_2 were similar and mainly distributed in India and Pakistan and areas of Inner Mongolia and Gansu and Guizhou Provinces in China. Potential source areas for NO_2 , $PM_{2.5}$ and PM_{10} were found in neighboring countries of South Asia in addition to domestic regions, including Inner Mongolia, Gansu Province and the Cheng-Yu economic region. Furthermore, parts of Yunnan Province (China) as well as India and Pakistan were potential source areas for CO_2 , CH_4 and N_2O .

Keywords: Air pollutants; Greenhouse gases; Background station; Southwestern China.

INTRODUCTION

Increased occurrence levels of air pollutants (ozone (O_3), carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO_x), particulate matter with a diameter of 2.5 micrometers or less ($PM_{2.5}$) and particulate matter with a diameter of 10 micrometers or less (PM_{10})) and greenhouse gases (carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O)) caused by intensified anthropogenic emissions have adversely affected the atmospheric environment, ecosystems, climate change and human health (Orri *et al.*, 2017; Wuebbles *et al.*, 2017). Hence, there have been increasing concerns about air pollutants and greenhouse gases at local, regional and even global scales (Meng *et al.*, 2009; Lin *et al.*, 2011; Thunis *et al.*, 2016). Ground-level O_3 is a powerful oxidant that can harm lung function and irritate the respiratory system; O_3 is also linked to premature deaths, heart attacks and other cardiopulmonary problems (www.epa.gov/ozone-pollution-and-your-patients-health; Weinhold, 2008). In addition, O_3 acts as a greenhouse gas (IPCC, 2001). As a precursor of O_3 , CO influences the oxidation of the atmosphere via interactions with hydroxyl radical (OH) (Gligorovski *et al.*, 2015). Frequent occurrences of acid rain and smog are regional-scale environmental problems in China, and SO_2 and NO_x play important roles in the formation of both problems (Ji *et al.*, 2014; Seinfeld and Pandis, 2016). Additionally, NO_x is a photochemical precursor resulting in the substantial enhancement of global background O_3 concentrations (Lin *et al.*, 2014; Sun *et al.*, 2016). CO_2 , CH_4 and N_2O are the most important greenhouse gases (Watson *et al.*, 1992) that can absorb infrared radiation emitted from the earth and partially reradiate this radiation back to the earth's surface (Seinfeld and Pandis, 2016). Therefore, to assess the impact of anthropogenic activities on the atmospheric environment, ecosystems, climate change and human health, it is necessary to conduct long-term continuous measurements of air pollutants (O_3 , CO, SO_2 , NO_x , $PM_{2.5}$ and PM_{10}) and greenhouse gases (CO_2 , CH_4 and N_2O). Nonetheless, colocated and simultaneous measurements of air pollutants and greenhouse gases at regional background sites are scarce, although a series of studies have been performed in regions with high anthropogenic emissions.

In contrast to studies in urban areas, studies on air pollutants and greenhouse gases at regional background sites not only provide valuable information on the influence of human activities on the atmospheric environment and global change but also are helpful for understanding the

transboundary transport of air pollution at a regional scale. Regional background sites are affected by very limited local anthropogenic emissions; consequently, the medium- or long-range transport of air pollutants could be the main contributor to local air pollution. Therefore, given their critical importance, a number of studies on air pollutants and greenhouse gases have been carried out at several regional background sites of the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) regions in China (Chao *et al.*, 2014; Pu *et al.*, 2015; Wang *et al.*, 2016). The results of these studies reflected the distinctive air pollution characteristics in the abovementioned regions in China and showed significant impacts of human activities on regional air quality. However, such studies at regional background sites in southwestern China are still scarce. The Gongga Mountain background station (GGS) is representative of the regional background in southwestern China (Fu *et al.*, 2008; Zhang *et al.*, 2012; Zhang *et al.*, 2014; Li *et al.*, 2017). Several field observations have been conducted at this station on volatile organic compounds, polar organic tracers in $PM_{2.5}$ and total particulate, reactive gaseous mercury and major chemical species of PM_{10} in specific periods or months (Fu *et al.*, 2008; Zhang *et al.*, 2012; Zhang *et al.*, 2014; Li *et al.*, 2017). However, to the best of our knowledge, no long-term continuous measurement of air pollutants and greenhouse gases has been performed at any regional background sites in southwestern China.

In this study, we present observations of major air pollutants, including O_3 , CO, SO_2 , NO_x , $PM_{2.5}$ and PM_{10} , as well as greenhouse gases, such as CO_2 , CH_4 and N_2O , at the GGS (101°97'E, 29°55'N; elevation: 3541 m) in southwestern China for the first time. The occurrence levels and temporal variations in these pollutants are discussed in detail, and potential contribution areas of the above substances are identified using the potential source contribution function (PSCF) method.

METHODS AND INSTRUMENTS

Sampling Site

As shown in Fig. 1, the GGS (101.97°E, 29.55°N; elevation: 3541 m) was established in the Gongga Mountain Observation and Experimental Station of Alpine Ecosystem, which is located in the Hailuoguo scenic area of the southeastern edge of the Qinghai-Tibetan Plateau. The Hailuoguo scenic area is famous for its large unique glacier and forest park areas. Neither private vehicular transport nor industrial activities operate near the sampling

Fig. 1. Location of the GGS atmospheric background station.

site. This station is approximately 250 km from Chengdu, the capital of Sichuan Province. There are two major roads in the north and east, 500 m and 400 m away from the station, respectively. The study area is dominated by the Southeast Asian Monsoon, with an annual average temperature, relative humidity, wind speed, atmospheric pressure and visibility range of $2.1 \pm 6.6^\circ\text{C}$, $82.5 \pm 20.9\%$, $1.7 \pm 0.9 \text{ m s}^{-1}$, $656.5 \pm 25.0 \text{ hPa}$ and $21.7 \pm 24.8 \text{ km}$, respectively. In addition, higher precipitation mainly occurs in both summer and autumn.

Instruments and Measurement Data

The sampling campaign was conducted from January 1, 2017, to December 31, 2017. All instruments were deployed, operated and maintained by following the regulations and standard operating procedures defined by the Ministry of Ecology and Environment of the People's Republic of China (http://bz.mep.gov.cn/bzwb/dqhjbh/jcgfffbz/index_2.shtml). The precision, detection limits, and calibration methods of all analyzers/monitors for major air pollutants of interest, including O_3 , CO , SO_2 , NO_x , CO_2 , CH_4 , N_2O , $\text{PM}_{2.5}$ and PM_{10} , have been described elsewhere in detail (Ji *et al.*, 2014; Christiansen *et al.*, 2015; Cortus *et al.*, 2015). Briefly, O_3 , CO , SO_2 , NO – NO_2 – NO_x , CO_2 , CH_4 and N_2O were observed using an ultraviolet photometric analyzer (Model 49i; Thermo Fisher Scientific (Thermo), USA), a gas filter correlation nondispersive infrared method analyzer (Model 48i TLE; Thermo, USA), a pulsed-fluorescence analyzer (Model 43i-TLE; Thermo, USA), a chemiluminescence analyzer (Model 42i-TL; Thermo, USA), a cavity ring-down spectroscopy analyzer for CO_2 and CH_4 (Model G2301; Picarro, Inc., USA) and a gas filter correlation N_2O analyzer (Model 320EU; Teledyne Technologies, USA), respectively. $\text{PM}_{2.5}$ and PM_{10} were simultaneously monitored using a Tapered Element Oscillating Microbalance with a Filter Dynamics Measurement System (TEOM-FDMS; 1405-DF TEOM; Thermo, USA). The gases were calibrated

daily by injecting a mixture of calibration gases (Scott-Marrin, Inc., CA, USA) and scrubbed ambient air. In addition, an internal catalytic converter was used to calibrate the CO blank. The TEOM-FDMS was calibrated with free-particle and standard filters (Thermo, USA). Meteorological parameters such as relative humidity (RH), wind direction (WD), wind speed (WS) and atmospheric temperature (T) were recorded via a colocated automatic meteorological station (Model AWS310; Vaisala, Finland). All data were processed using an Igor-based software (Wu *et al.*, 2018).

Source Area Identification

The PSCF method has been extensively used in identifying source locations of atmospheric species (Lupu and Maenhaut, 2002). PSCF is defined as the probability that an air parcel with a concentration more than a specified threshold reaches the study site after having resided in a certain grid cell of the spatial domain of interest (Lupu and Maenhaut, 2002). In this study, the potential source areas are identified based on the PSCF method using a GIS-based tool, named Trajstat, which can perform a comprehensive investigation of the geographical distribution of atmospheric species origins (Wang *et al.*, 2009).

RESULTS AND DISCUSSION

Occurrence Levels

The time series of O_3 , CO , SO_2 , NO , NO_2 , $\text{PM}_{2.5}$ and PM_{10} concentrations as well as CO_2 , CH_4 and N_2O mixing ratios are presented from January 1 to December 31, 2017, in Fig. 2. In addition, the means and standard deviations (STDEVs) of air pollutants and greenhouse gases are listed in Table 1. For clarification, due to regular maintenance activities (e.g., periodic zero-span checks, data dump/collection, calibrations, etc.) and disruptions by severe weather conditions, a number of datasets were not recorded during the above sampling period. The annual average

Fig. 2. Time series of O₃, CO, SO₂, NO_x, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ concentrations and mixing ratios in 2017.

Table 1. Average concentrations and standard deviations (STDEVs) of O₃, CO, SO₂, NO_x, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ in 2017.

Sampling period	O ₃	STDEV	CO	STDEV	SO ₂	STDEV	NO	STDEV	NO ₂	STDEV	CO ₂	STDEV	CH ₄	STDEV	N ₂ O	STDEV	PM _{2.5}	STDEV	PM ₁₀	STDEV
Jan-2017	81.0	10.0	0.4	0.2	0.5	0.3	NA	NA	2.3	1.4	412.5	7.5	1.971	0.094	321.9	9.9	7.3	5.7	14.9	9.5
Feb-2017	81.4	11.8	0.3	0.2	0.4	0.3	NA	NA	2.6	1.3	412.1	6.3	1.955	0.079	321.3	10	6.6	6.9	16.3	16
Mar-2017	89.0	14.1	0.4	0.2	0.2	0.1	NA	NA	2.6	1.3	414.0	5.1	1.986	0.071	328.2	9.9	4.9	5.2	10.5	11.6
Apr-2017	95.1	15.1	0.2	0.1	0.2	0.3	NA	NA	1.6	0.9	412.6	4.0	1.946	0.057	NA	NA	6.2	7.3	10.8	11.6
May-2017	88.2	16.7	0.2	0.1	0.5	0.3	NA	NA	1.6	0.7	405.9	6.7	1.932	0.046	322.1	10.8	6.2	7.2	10.1	10.4
Jun-2017	82	22.8	0.2	0.2	1.1	1.3	0.8	3.2	1.7	1.7	397.3	7.1	1.918	0.041	329.8	13.9	6.6	6.8	10.1	18.2
Jul-2017	82.6	25.0	0.2	0.1	0.6	0.5	1	1.4	0.9	0.6	396.9	9.6	1.878	0.043	328.8	14.3	8.9	7.2	12.7	9.8
Aug-2017	63.2	20.6	0.3	0.2	0.8	1.3	1.3	2.6	1.2	1.7	397.5	9.3	1.896	0.023	322.4	17	5.7	3.8	8.2	6.1
Sep-2017	55.6	18.9	0.2	0.1	0.4	0.3	0.9	1.8	0.9	0.4	396.8	9.1	1.947	0.071	324.3	11.1	4.3	2.6	5.3	3.5
Oct-2017	52.0	13.2	0.3	0.1	0.4	0.3	0.9	2.9	1.3	0.7	405.9	6.2	1.958	0.071	325.1	13.1	4.4	3.2	5.1	4.4
Nov-2017	60.8	11.2	0.2	0.1	0.5	0.2	0.6	1.9	2	1.2	408.3	6.4	1.948	0.073	322.4	11.5	7.0	4.3	8.7	6.7
Dec-2017	73.8	16.6	0.3	0.1	0.7	0.8	0.6	2.4	2.6	1.7	409.8	5.6	1.943	0.072	322.8	10.9	9.3	7.4	15.5	12.2
The whole year	74.7	22.0	0.3	0.2	0.5	0.6	0.8	2.3	1.7	1.3	406.1	9.5	1.941	0.071	324.5	14.8	6.5	6.2	10.7	11.3

Units for O₃, CO, SO₂, NO, NO₂, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ are $\mu\text{g m}^{-3}$, mg m^{-3} , $\mu\text{g m}^{-3}$, $\mu\text{g m}^{-3}$, ppm, ppm, ppb, $\mu\text{g m}^{-3}$ and $\mu\text{g m}^{-3}$, respectively (in this manuscript, all units for the observed O₃, CO, SO₂, NO, NO₂, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ concentrations are the same).

concentrations (mixing ratios) of O₃, CO, SO₂, NO, NO₂, PM_{2.5}, and PM₁₀ as well as CO₂, CH₄ and N₂O were $74.7 \pm 22.0 \mu\text{g m}^{-3}$, $0.3 \pm 0.2 \text{ mg m}^{-3}$, $0.5 \pm 0.6 \mu\text{g m}^{-3}$, $0.8 \pm 2.3 \mu\text{g m}^{-3}$, $1.7 \pm 1.3 \mu\text{g m}^{-3}$, $6.5 \pm 6.2 \mu\text{g m}^{-3}$, $10.6 \pm 11.2 \mu\text{g m}^{-3}$, $406.1 \pm 9.5 \text{ ppm}$, $1.941 \pm 0.071 \text{ ppm}$ and $324.5 \pm 14.8 \text{ ppb}$, respectively. According to the Chinese Ambient Air Quality Tier II Standards (CAAQS), there were no sampling days when the thresholds of O₃ (maximum 8 h mean: $160 \mu\text{g m}^{-3}$), NO₂ (24 h mean: $80 \mu\text{g m}^{-3}$), SO₂ (24 h mean: $150 \mu\text{g m}^{-3}$), CO (24 h mean: 4 mg m^{-3}), PM_{2.5} (24 h mean: $75 \mu\text{g m}^{-3}$) and PM₁₀ (24 h mean: $150 \mu\text{g m}^{-3}$) were exceeded. However, there were 82, 1 and 1 days violating the limits of O₃ (maximum 8 h mean: $100 \mu\text{g m}^{-3}$), PM_{2.5} (24 h mean: $25 \mu\text{g m}^{-3}$) and PM₁₀ (24 h mean: $50 \mu\text{g m}^{-3}$), respectively, according to the World Health Organization (WHO) air quality standards. Notably, 54.9% of all days exceeded the WHO ozone standard in the spring, which is consistent with the spring O₃ maximum observed in the Northern Hemisphere (Monks, 2000; Lin *et al.*, 2015). In addition, the sampling days exceeding the WHO PM standards were closely related to the long-range transport of PM from regions of intense anthropogenic emissions and/or local emissions.

Frequency distributions are a commonly used visualization tool used to display the number of observations within a given interval. In this study, histograms showing the normalized, lognormal and cumulative frequency distributions of O₃, CO, SO₂, NO, NO₂, PM_{2.5} and PM₁₀ concentrations and CO₂, CH₄ and N₂O mixing ratios were plotted and are presented in Fig. 3. The results show that all of the air pollutants of interest in this study possessed a unimodal bell-shaped lognormal distribution pattern during the entire sampling period. For example, O₃ concentrations ranging from 30 to $130 \mu\text{g m}^{-3}$ were dominant, and the distribution shape for O₃ was generally symmetrical, with the highest frequency at $90 \mu\text{g m}^{-3}$, occupying more than 95% of the total sample data. It was reported that the frequency distribution of O₃ concentrations can be a good indicator for the studied type of sampling site because this distribution captures the impact of NO_x emission sources mainly associated with combustion activities (Escudero *et al.*, 2014). Based on the diurnal variations in both O₃ and NO (Section Seasonal and Diurnal Variations), in such a remote site, NO titration most likely seldom affects the O₃ frequency distribution, although other processes, such as deposition or scavenging, might play more important roles in removing O₃ from the troposphere (Monks *et al.*, 2015). The frequency distributions of CO, SO₂, NO₂, PM_{2.5} and PM₁₀ were more skewed to the right compared with the other distributions and featured a pronounced peak at approximately 0.25 mg m^{-3} , $0.22 \mu\text{g m}^{-3}$, $1.0 \mu\text{g m}^{-3}$, $4 \mu\text{g m}^{-3}$ and $6 \mu\text{g m}^{-3}$, respectively, indicating the impact of relatively clean background continental air masses, while the long tails on the right side of the graph toward high concentration levels with a low cumulative distribution could indicate the minor influence of local rural pollution and/or urban pollution plumes. NO concentrations ranging from 0.1 to $1.0 \mu\text{g m}^{-3}$ were dominant, accounting for approximately 98% of the total sample data. Although the

Fig. 3. Frequency distributions of O₃, CO, SO₂, NO, NO₂, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ concentrations or mixing ratios.

NO dataset was only slightly more than half of that for the remaining pollutants, the right-skewed frequency distribution of NO was similar to the above trend, with a long tail on the right side. In addition, Fig. 3 clearly shows that the frequency distributions of the CO₂, CH₄ and N₂O mixing ratios were very narrow and were in the ranges of 380.1–435.0 ppm, 1.802–2.232 ppm and 300.0–379.9 ppb, respectively; such patterns might indicate that high emissions of these gases had almost no impacts during the study period.

As shown in Table 2, a comparative analysis was conducted between the observations in the present study

and those reported earlier at background stations in China and worldwide. The annual mean O₃ concentration ($74.7 \pm 22.0 \mu\text{g m}^{-3}$) at the GGS was higher than the values observed at the Dinghushan (DHS) background site in southern China (24.6 ppb ; Chao *et al.*, 2014), the Jinsha (JS) background station in central China (24.6 ppb ; Lin *et al.*, 2011) and the Lin'an (LA) background station in the YRD of China (34.7 ppb ; Xu *et al.*, 2008); and it was similar to the values observed at the Shangdianzi (SDZ) background station in the BTH region of China (36.9 ppb ; Lin *et al.*, 2008) and the remote highland site of Dangxiong

Table 2. Comparison of the present study with previous measurements from background stations in China and around the world.

Site	Altitude	Period	Substance	Concentrations	References
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	O ₃	74.7 µg m ⁻³	This study
Dinghushan, a regional background site in southern China	100 m	January–December 2013	O ₃	24.6 ppb	Chao et al., 2014
Jinsha, a regional background site in central China	750 m	June 2006–July 2007	O ₃	24.6 ppb	Lin et al., 2011
Lin'an, a regional background station in the Yangtze River Delta	138.6 m	November 2003–November 2004	O ₃	32.4 ppb	Yang et al., 2008
Shangdianzi, a global background station in northern China	293.3 m	January–December 2006	O ₃	30.9 ppb	Meng et al., 2009
Xinglong, a regional background station in northern China	940 m	June 2005–September 2006	O ₃	49.2 ppb	Ma et al., 2011
Dangxiong, a remote highland site, China	4187 m	September 2009–August 2011	O ₃	38.5 ppb	Lin et al., 2015
Nam Co, a remote highland site, China	4745 m	January 2011–October 2015	O ₃	47.6 ppb	Yin et al., 2017
Shangri-La, China	3580 m	December 2007–November 2009	O ₃	24.2–58.3 ppb	Ma et al., 2014
Climate Observatory–Pyramid, a background site, Nepal	5079 m	March 2006–February 2008	O ₃	49 ± 12 ppb	Cristofanelli et al., 2010
Oki, a regional background site in Japan	90 m	January 1995–December 1996	O ₃	47.9 ppb	Lam et al., 2004
Cheju, a regional background site in South Korea	70 m	January 1995–December 1996	O ₃	48.1 ppb	Lam et al., 2004
D'Aguilar, a regional background site in Hong Kong, China	60 m	January 1995–December 1996	O ₃	31.6 ppb	Lam et al., 2004
Longfenshan, a regional background site in northeastern China	325 m	January 1995–December 1996	O ₃	34.8 ppb	Lam et al., 2004
Waliguan, a global background station, China	3810 m	January 1995–December 1997	O ₃	48.2 ppb	Lam et al., 2004
Zhongshan Station, East Antarctica	18.5 m	2008–2013	O ₃	24.3–27.1 ppb	Bian et al., 2016
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	NO ₂	1.7 µg m ⁻³	This study
Shangdianzi, a global background station in northern China	293.3 m	January–December 2006	NO ₂	11.5 ppb	Meng et al., 2009
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	CO	0.3 mg m ⁻³	This study
Shangdianzi, a global background station in northern China	293.3 m	January–December 2006	CO	742 ppb	Meng et al., 2009
Dinghushan, a regional background site in southern China	100 m	January–December 2013	CO	348 ppb	Chao et al., 2014
Dinghushan, a regional background site in southern China	100 m	January–December 2013	SO ₂	4.0 ppb	Chao et al., 2014
Shangdianzi, a global background station in northern China	293.3 m	January–December 2006	SO ₂	7.6 ppb	Meng et al., 2009
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	CO ₂	406.1 ppm	This study
Mt. Dodaia	876 m	2013	CO ₂	400 ppm	Jia et al., 2017
King's Park site, Hong Kong, China	66 m	2013	CO ₂	407.6 ppm	Jia et al., 2017
Korean site, South Korea	-	2013	CO ₂	404.9 ppm	Jia et al., 2017
Mauna Loa, USA	3400 m	2017	CO ₂	406.53 ppm	a*
Li'an, China	138.6 m	September 2006–August 2007	CO ₂	387.8 ppm	Liu et al., 2009
Waliguan, China	3816.0 m	September 2006–August 2007	CO ₂	383.5 ppm	Liu et al., 2009
Waliguan, China	3816.0 m	2013	CO ₂	397.2 ppm	Jia et al., 2017
Longfengshan, China	330.5 m	September 2006–August 2007	CO ₂	384.3 ppm	Liu et al., 2009
Shangdianzi, a global background station in northern China	293.3 m	September 2006–August 2007	CO ₂	385.9 ppm	Liu et al., 2009
Shangri-La, China	3580 m	September 2009–August 2010	CO ₂	386.82–394.78 ppm	Li et al., 2012
Xinglong, China	940 m	1995–2000	CO ₂	376.7 ppm	Cheng et al., 2003
Waliguan, China	3816.0 m	1995–2000	CO ₂	373.5 ppm	Cheng et al., 2003
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	CH ₄	1.941 ppm	This study
Xinglong, China	940 m	1995–2000	CH ₄	1.886 ppm	Cheng et al., 2003
Waliguan, China	3816.0 m	1995–2000	CH ₄	1.831 ppm	Cheng et al., 2003
Li'an, China	138.6 m	January 2009–December 2013	CH ₄	1.965 ppm	Yang et al., 2018

Table 2. (continued).

Site	Altitude	Period	Substance	Concentrations	References
Longfengshan, China	330.5 m	January 2009–December 2013	CH ₄	1.939 ppm	Yang et al., 2018
Shangdianzi, a global background station in northern China	293.3 m	January 2009–December 2013	CH ₄	1.914 ppm	Yang et al., 2018
Waliguan, China	3816.0 m	January 2009–December 2013	CH ₄	1.864 ppm	Yang et al., 2018
Shangri-La, China	3580 m	July 2010–December 2013	CH ₄	1.861 ppm	Yang et al., 2018
Zhongshan Station, East Antarctica	18.5 m	2008–2013	CH ₄	1.740–1.766 ppm	Bian et al., 2016
Global average	-	2008–2013	CH ₄	1.798–1.824 ppm	Bian et al., 2016
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	N ₂ O	324.5 ppb	This study
Zhongshan Station, East Antarctica	18.5 m	2008–2012	N ₂ O	321.9 ppb	Ye et al., 2016
Xinglong, China	940 m	1995–2000	N ₂ O	316.7 ppb	Cheng et al., 2003
Waliguan, China	3816.0 m	1995–2000	N ₂ O	314.9 ppb	Cheng et al., 2003
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	PM _{2.5}	6.5 µg m ⁻³	This study
Lasmas de Olo, a Portuguese background site, Portugal	1086 m	2004–2010	PM _{2.5}	9 µg m ⁻³	Cusack et al., 2012
Schauinsland, a German background site, Germany	1205 m	2002–2009	PM _{2.5}	7.2 µg m ⁻³	Cusack et al., 2012
Campisabalos, a background site, Spain	1360 m	2002–2010	PM _{2.5}	6.9 µg m ⁻³	Cusack et al., 2012
Payerne, a Swiss background site, Switzerland	489 m	2002–2010	PM _{2.5}	14.5 µg m ⁻³	Cusack et al., 2012
Illmitz, an Austrian background site, Austria	117 m	2002–2010	PM _{2.5}	19.7 µg m ⁻³	Cusack et al., 2012
Aspvreten, a regional background site in Stockholm, Sweden	30 m	1998–2010	PM _{2.5}	8 µg m ⁻³	Jönsson et al., 2013
Gonggashan, a background site in southwestern China	3541 m	January–December 2017	PM ₁₀	10.6 µg m ⁻³	This study
Zhuzhang, in the mountainous rural area of southwestern China	3583 m	from August 2004–February 2005	PM ₁₀	~10 µg m ⁻³	Qu et al., 2009

Conversion factors between ppb and µg m⁻³ and ppm and mg m⁻³ can be found in the following link:

https://ukair.defra.gov.uk/assets/documents/reports/cat06/0502160851_Conversion_Factors_Between_ppb_and_pdf

a* <https://www.esrl.noaa.gov/gmd/ccgg/trends/data.html>

(38.5 ppb; Lin *et al.*, 2015); while it was lower than those observed at Nam Co, a remote highland site in the inland Tibetan Plateau of China (47.6 ± 11.6 ppb, 4730 m a.s.l.), the Xinglong background station in northern China (49.2 ppb, 960 m a.s.l.), and the Nepal Climate Observatory-Pyramid (49 ± 12 ppbv, 4745 m a.s.l.; Cristofanelli *et al.*, 2010). Given that the GGS site is at a much higher latitude than the other sites, the contribution of stratospheric ozone intrusion at the GGS may be more significant than the stratosphere-to-troposphere transport at most of the other stations discussed here (Mauzerall *et al.*, 1996). The concentrations of NO_2 at the GGS were lower than those at the DHS, SDZ, and LA sites, which was consistent with the fact that the DHS, SDZ, and LA sites are in rapidly developing areas with much more intensive industrial emissions and a high increase in the number of motor vehicles. In contrast, the concentrations of SO_2 and CO observed at the GGS were lower than those observed at the DHS, SDZ, JS and LA stations, which also indicates the limited influence of anthropogenic activities at the GGS.

The annual mean CO_2 concentration (406.1 ± 9.5 ppm) at the GGS was similar to that at the background sites of Mt. Dodaira, Japan (> 400 ppm in 2013); King's Park, Hong Kong, China (407.6 ppm in 2013); Korea (404.9 ppm in 2013); Mauna Loa (global background site; 406.53 ppm in 2017) and LA (404.7 ± 8.2 ppm, 405.6 ± 5.3 ppm and 407.0 ± 5.3 ppm for 2009, 2010 and 2011, respectively). However, the annual mean CO_2 concentration at the GGS was higher than that observed from September 2006 to August 2007 at the Waliguan (WLG; 383.5 ppm), SDZ (385.9 ppm), LA (387.8 ppm) and Longfengshan (LFS; 384.3 ppm) background sites. This trend suggests that CO_2 concentrations at the abovementioned background sites in China have increased significantly over the past decade, possibly due to the rapid economic development and extensive enhancement of energy consumption (<http://www.stats.gov.cn/tjsj/ndsj/2017/indexch.htm>). The average CH_4 concentrations at the GGS were higher than those observed at the Antarctica site (1.740–1.766 ppm), the WLG background site (1.864 ppm) and the Shangri-La background site (1.861 ppm) as well as the global background concentration (1.798–1.824 ppm) (Bian *et al.*, 2016) but were similar to those at the SDZ (1.914 ppm), LA (1.965 ppm) and LFS (1.939 ppm) background sites. In addition, the monthly average N_2O concentrations, which ranged from 321.3 to 329.8 ppb in the GGS, were similar to those at Zhongshan Station, East Antarctica, which varied from 320.5 to 324.8 ppb (Ye *et al.*, 2016), but higher than those at the Xinglong (316.7×10^{-9} , from 1995 to 2000) and WLG (314.9×10^{-9} , from 1995 to 2000) background sites.

Compared to the observations at other background sites in the world, the $\text{PM}_{2.5}$ concentrations measured at the GGS site ($6.5 \pm 6.2 \mu\text{g m}^{-3}$) were slightly lower than or similar to those at the Portugal ($9.4 \mu\text{g m}^{-3}$), Germany ($10 \mu\text{g m}^{-3}$) and Scandinavia ($6.6 \mu\text{g m}^{-3}$) background sites and were considerably lower than those recorded at the Switzerland ($14.5 \mu\text{g m}^{-3}$) and Austria ($19.7 \mu\text{g m}^{-3}$) background sites. The annual PM_{10} concentration was almost the same as a previous result ($10.8 \mu\text{g m}^{-3}$) from the

Stockholm regional background site in Europe (Jonsson *et al.*, 2013). Although most background monitoring sites are distant from urban areas, they might be affected by anthropogenic emissions to different extents. In addition, synoptic conditions play an important role in PM levels (Ji *et al.*, 2012); for instance, differences in the precipitation intensity and atmospheric stagnation of various sites could affect the removal, transport and accumulation of particulate matter. Furthermore, the transboundary transport of air masses from regions of intense anthropogenic activities can lead to spatial variation in the levels of air pollutants at various background sites.

Overall, the air quality at the GGS site was better than that at the background sites located in the most populated and developed city clusters, such as the BTH, YRD and PRD regions of China. In addition to meteorological conditions and various sinks, the spatial variation in the levels of air pollutants in different regions could be highly affected by regional emission intensities of major air pollutants of interest (National Bureau of Statistics of the People's Republic of China, 2017).

Seasonal and Diurnal Variations

The average seasonal variations in O_3 , CO, SO_2 , N_2O , $\text{PM}_{2.5}$, PM_{10} , NO_2 , CO_2 and CH_4 and the RH, T and WS values at the GGS are shown in Fig. 4. The highest values of O_3 were observed in spring, while the lowest values occurred in autumn. The maximum O_3 value in spring is consistent with the spring O_3 maximum frequently observed in the Northern Hemisphere (Monks, 2000; Vingarzan, 2004; Ran *et al.*, 2014; Lin *et al.*, 2015). Stronger ultraviolet (UV) radiation has been observed in spring at the GGS (Liu *et al.*, 2017), which was favorable for O_3 production and further contributed to the highest O_3 concentrations in spring. The lower levels of CO from April to July were almost opposite to the seasonal variation in O_3 , which could be caused by the yield of O_3 from CO oxidation under stronger UV radiation (Seinfeld and Pandis, 2016). The higher CO_2 concentrations observed in April and May might originate from CO oxidation, which supports the above deduction to a degree. However, vigorous summertime photosynthesis resulted in a decline in CO_2 concentrations in the summer. The O_3 levels were relatively lower in late summer and autumn compared with those in spring. The high occurrence frequency of precipitation could scavenge more O_3 in late summer and autumn than the other two seasons. SO_2 and $\text{PM}_{2.5}$ showed the highest concentrations in summer and the lowest concentrations in autumn. As shown in Fig. 2, high concentration spikes of both SO_2 and $\text{PM}_{2.5}$ could be observed. Given that the lifetime of SO_2 is normally short under conditions of high RH and O_3 concentrations (Seinfeld and Pandis, 2016), it might be reasonable to infer that local emissions outweighed the regional transport of both pollutants, resulting in the increased concentrations of RH and O_3 at this background site. Note that pulse spikes of SO_2 and $\text{PM}_{2.5}$ accompanied the increase in CO and NO_2 concentrations. In addition, PM_{10} did not increase as significantly as $\text{PM}_{2.5}$ during the sampling period. Hence, all these observations could lead

Fig. 4. The average seasonal variations in O_3 , CO, SO_2 , NO_2 , CO_2 , CH_4 , N_2O , $\text{PM}_{2.5}$ and PM_{10} (a) as well as RH, T and WS; (b) at the GGS.

to the hypothesis that the local air quality at such a remote site might be affected more by traffic emissions during summer than during other seasons. Because the Hailuoguo scenic area is most attractive to tourists during summer, this difference could be ascribed to vehicular emissions from tourists. The seasonal pattern of CO was similar to that of NO_2 and PM_{10} . The highest CO, NO_2 and PM_{10} values were mostly observed in winter. The lowest CO_2 concentrations were observed in summer, which could be

because vigorous summertime photosynthesis resulted in a decline in CO_2 mixing ratios. The lowest CH_4 concentrations were recorded in summer, while the CH_4 levels in the other three seasons were very similar to each other. Such a seasonality of the CH_4 mixing ratio in a clean-background environment is possibly related to OH radicals that are dependent on the seasonally varying intensity of ultraviolet radiation (Necki *et al.*, 2003). In contrast, higher N_2O concentrations were observed in summer than during the

other seasons due to increases in the emission of N_2O caused by enhanced microbial activity at higher ambient temperatures (Wang *et al.*, 2018).

The average diurnal variations in O_3 , CO, SO_2 , NO_x , CO_2 , CH_4 , N_2O , $\text{PM}_{2.5}$ and PM_{10} as well as meteorological

conditions, namely, RH, T and WS, at the GGS are shown in Fig. 5. The diurnal cycles of O_3 showed minimum and maximum values in the early morning and afternoon, respectively. During the nighttime, the ozone concentration decreased slowly, mainly due to the titration of NO and

Fig. 5. Average diurnal variations in O_3 , CO, SO_2 , NO_x , CO_2 , CH_4 , N_2O , $\text{PM}_{2.5}$ and PM_{10} concentrations or mixing ratios (a) as well as RH, T and WS; (b) at the GGS station.

deposition processes (Seinfeld and Pandis, 2016); during the daytime, with the increase in solar radiation after sunrise, O_3 started increasing as more ozone was generated by photochemical reactions (Seinfeld and Pandis, 2016). Note that the daily amplitude of O_3 at the background site was lower than that at urban sites (Reddy *et al.*, 2011). The diurnal variations in CO and NO_2 resulted in higher concentrations in the daytime and lower concentrations in the nighttime. The diurnal variations in NO and N_2O resulted in obvious peaks at approximately 09:00 and 10:00, respectively. No obvious diurnal variation in SO_2 was observed. The diurnal variations in CO and NO_2 might be ascribed to two reasons. One reason could be that increasing anthropogenic activities in the daytime around the study site enhanced the primary emissions of these gases. The other reason may possibly be related to the evolution of the planetary boundary layer (PBL) and the transboundary transport of air pollutants. It is known that CO and NO_2 concentrations increase with the decay of the nocturnal boundary layer, consequently making the high concentrations of these pollutants over source areas migrate to downwind receptor sites and leading to enhancements in CO and NO_2 concentrations at the receptor sites during the daytime. The $PM_{2.5}$ and PM_{10} concentrations did not show the obvious bimodal distribution that they show at urban sites (Wang *et al.*, 2015). Similar to CO and NO_2 , a slight increase in the $PM_{2.5}$ and PM_{10} concentrations was recorded in the daytime. Although the GGS is a regional background site, scattered anthropogenic emissions in the daytime could partly lead to the increase in the $PM_{2.5}$ and PM_{10} concentrations; in addition, secondary aerosol formation via the atmospheric transformation of precursors from local sources or regional transport might contribute to higher concentrations of $PM_{2.5}$ and PM_{10} during the daytime than during the nighttime.

The average diurnal variation in CO_2 is shown in Fig. 5. The diurnal cycle of CO_2 reflects a diurnal periodicity of atmospheric vertical mixing, the release and uptake of CO_2 by respiration, photosynthesis in the biosphere and human activities. In the daytime, photosynthesis is dominant in the net exchange between the biosphere and atmosphere, which could lead to the decline in CO_2 concentrations. During the nighttime, photosynthesis stops and vertical mixing is weak, while plant respiration continues. The CO_2 emitted by plant respiration accumulates near the ground. In addition, anthropogenic emissions also contributed slightly to the observed CO_2 , although the GGS site was distant from densely populated areas. The CH_4 mixing ratios showed a very weak diurnal variation, with minima during midday, which might be caused by the elevated PBL, as anthropogenic emissions of CH_4 are scarce in such a remote mountain area. N_2O peaked at 10:00–12:00 and remained almost stable over the other time period. The N_2O peak can probably be attributed to N_2O emissions caused by enhanced microbial activity with the increase in soil temperatures. With the increase in solar radiation in the daytime, the soil temperature did not immediately increase with the atmospheric temperature due to the different heat capacities of the soil and atmosphere. With

the increase in the soil temperature (generally 2 h later than that in the atmospheric temperature), the enhancement of microbial activity led to N_2O peaks. In addition, N_2O peaks did not occur with NO peaks, suggesting that they did not come from the same sources, i.e., vehicular emissions (Vojtišek-Lom *et al.*, 2018).

Impacts of Transboundary Air Pollution and Local Emissions

The impact of the regional transport and local emissions of air pollutants and greenhouse gases can be visualized based on both polar plots of wind and occurrence levels of the abovementioned pollutants as well as PSCF results of the hourly resolved input data. Fig. 6 shows the dependent distributions between WS/WD and the concentrations of all pollutants of interest. The concentrations of air pollutants and greenhouse gases showed evident wind sector gradients. For example, higher O_3 , SO_2 and $PM_{2.5}$ concentrations were correlated with winds from the northeast (NE), and lower concentrations were correlated with northwest (NW) winds. This wind sector dependence of O_3 , SO_2 and $PM_{2.5}$ concentrations is highly consistent with the spatial distribution of their concentrations in the surrounding areas. It was reported that relatively high and low O_3 , SO_2 and $PM_{2.5}$ concentrations were observed in the Cheng-Yu region and Tibetan Plateau, located to the northeast and northwest of the study site, respectively (China Environmental Statement, 2016). Clearly, zones with high concentrations of NO_2 , CO, CO_2 and CH_4 were in the center of these dependent distribution areas where low WS was recorded, indicating that local emissions might play an important role in the accumulation of these air pollutants. In contrast to other air pollutants, higher PM_{10} concentrations were more correlated with high WS, although $PM_{2.5}$ concentrations did not vary as much as the PM_{10} . This result suggests that wind-blown sand or dust contributed significantly to PM_{10} . In addition, winds from the southeast resulted in relatively high PM_{10} concentrations, possibly implying that mid- and/or long-range transport of aerosols from both the south and southeast could contribute to PM_{10} .

Fig. 7 shows potential source areas for O_3 , CO, SO_2 , NO_2 , $PM_{2.5}$ and PM_{10} concentrations as well as the CO_2 , CH_4 and N_2O mixing ratios during the study period. As shown in Fig. 7, the potential source areas of air pollutants and greenhouse gases showed obvious spatial distributions. It appeared that parts of Inner Mongolia and Gansu, Ningxia, Sichuan, Chongqing and Hubei Provinces as well as adjacent areas of Hunan, Guizhou and Guangxi Provinces were potential source areas for O_3 , which is consistent with the link between WD and relatively high O_3 concentrations. With the rapid economic development in inland areas of China, more intensive emissions of O_3 precursors could lead to O_3 formation during transport processes (China Environmental Statement, 2016). The potential source areas for CO were mainly focused on neighboring countries in South Asia, such as India and Pakistan, which were associated with high pollution emissions (<https://nar.ucar.edu/sites/default/files/labs/nesl/ACD-EmissionsParticulates02.jpg>), in addition to areas of Inner Mongolia and Gansu and

Fig. 6. The dependent distributions between wind speed and direction and the concentrations of O₃, CO, SO₂, NO_x, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ at the GGS station.

Guizhou Provinces. Considering that SO₂ has similar emission sources as CO, the potential source areas for SO₂ overlapped with those of CO, but an intense SO₂ band caused by the emissions of power plants appeared in parts of Inner Mongolia and Gansu Province (Liu *et al.*, 2015). The potential source areas for NO₂, PM_{2.5} and PM₁₀ were also found in neighboring countries in South Asia in addition to Inner Mongolia, Gansu Province and the Cheng-Yu economic region. High emissions of air pollutants in the abovementioned areas may result in the increase in NO₂, PM_{2.5} and PM₁₀ concentrations at the GGS site via transboundary transport. This result is almost consistent with that obtained by Qu *et al.* (2008), who found that anthropogenic sources in the Cheng-Yu economic region (Sichuan Basin), southeastern Yunnan Province and South Asian countries evidently influence Zhuzhang (in a mountainous rural area of southwestern China, 3583 m a.s.l.). For CO₂, the potential source areas were focused on South Asian countries neighboring China and the adjacent areas of the Tibet Autonomous Region, Yunnan Province and Myanmar, which could be caused by high fuel consumption resulting in high CO₂ emissions (<http://edgar.jrc.ec.europa.eu/overview.php?v=CO2andGHG1970-2016>). The potential source areas of CH₄ were

mainly distributed in Myanmar, India and Pakistan and parts of Yunnan Province, China. The potential source areas of N₂O were recorded in the Cheng-Yu economic region, Yunnan Province, borders of the Tibet Autonomous Region of China and countries of South Asia; it is understandable that these areas with intense agricultural activities are the main sources of N₂O and CH₄ (Davidson and Kanter, 2014; Saunio *et al.*, 2016).

CONCLUSION

This report presents the first year-long (from January 1 to December 31, 2017) and real-time measurement study of O₃, NO_x, SO₂, CO, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ performed at the GGS background site in southwestern China. The frequency of occurrence and temporal variations of this pollution were discussed in detail, and potential contribution areas of the above substances were identified using PSCF. The conclusions of this study are as follows.

High O₃ and PM_{2.5} concentrations that exceeded WHO thresholds were observed and were closely associated with the yield from CO oxidation and long-range transport and/or local emissions of PM. The O₃, CO, SO₂, NO_x,

Fig. 7. Potential source areas for O_3 , CO , SO_2 , NO_2 , $\text{PM}_{2.5}$ and PM_{10} concentrations as well as CO_2 , CH_4 and N_2O mixing ratios during the study period. The color code denotes the PSCF probability. The location of the site is indicated by ●.

CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀ concentrations presented typical lognormal patterns during the entire study period. The concentrations of these air pollutants at the GGS site were comparable to those at most background sites around the world. The slight differences in the levels of major air pollutants and greenhouse gases between background sites may be affected, to different extents, by anthropogenic emissions.

Obvious seasonal and diurnal variations were observed in the concentrations of O₃, CO, NO_x, SO₂, CO₂, CH₄, N₂O, PM_{2.5} and PM₁₀. High values were observed for O₃ in spring and summer, and the diurnal variations were characterized by higher daytime than nighttime values. SO₂ and PM_{2.5} reached their highest concentrations in summer and their lowest concentrations in autumn. Relatively high CO, NO₂ and PM₁₀ concentrations were mostly observed in spring and winter. CO and NO₂ exhibited higher concentrations in the daytime than in the nighttime. The vigorous summertime photosynthesis of vegetation resulted in lower CO₂ concentrations during summer than during the other seasons. The lowest concentrations for CH₄ were recorded in summer, whereas its levels in the other three seasons were similar to each other. High temperatures enhanced microbial activity, resulting in higher N₂O concentrations during summer (compared to the other seasons) and in the daytime (compared to the nighttime).

Meteorological conditions significantly affect the background concentrations of the studied air pollutants. These pollutants can accumulate because of stagnant meteorological conditions and/or the contribution of mid- and long-range transport. The potential source areas for the air pollutants and greenhouse gases showed an obvious spatial distribution. The high potential source areas were distributed in parts of Inner Mongolia and Gansu, Ningxia, Sichuan, Chongqing and Hubei Provinces as well as adjacent areas of Hunan, Guizhou and Guangxi Provinces. High emissions in India and Pakistan also played an important role in increasing the CO, SO₂, NO₂, PM_{2.5} and PM₁₀ concentrations. In addition, anthropogenic emissions from Inner Mongolia and Gansu Province contributed greatly to the enhancement of CO, SO₂, PM_{2.5} and PM₁₀. Increases in the CO₂ concentration due to high fuel consumption were traceable primarily to South Asian countries neighboring China, and the adjacent areas of the Tibet Autonomous Region, Yunnan Province and Myanmar, whereas the enhancement of CH₄ and N₂O concentrations in the Cheng-Yu economic region, Yunnan Province, the Tibet Autonomous Region of China, Myanmar, India and Pakistan were attributable to intense agricultural activities.

The results of this study contribute to a better understanding of how anthropogenic activities impact the occurrence of air pollution against regional backgrounds, which may improve the modeling of regional air quality in the future.

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REFERENCES

- Bian, L.G., Gao, Z.Q., Sun, Y.L., Ding, M.H., Tang, J. and Schnell, R. (2016). CH₄ monitoring and background concentration at Zhongshan Station. *Atmos. Clim. Sci.* 6: 135–144.
- Chao, N., Ji, D.S., Chen, J.S., Xin, J.Y., Hu, B., Wang, Y.S. and Wang, H. (2014). Characteristics of gaseous pollutants at a regional background station in Southern China. *Atmos. Ocean Sci. Lett.* 7: 340–345.
- China Environmental Statement (2016). <http://www.zhb.gov.cn/hjzl/zghjzkgb/lnzghjzkgb/201706/P020170605833655914077.pdf>.
- Christiansen, J.R., Outhwaite, J. and Smukler, S.M. (2015). Comparison of CO₂, CH₄ and N₂O soil-atmosphere exchange measured in static chambers with cavity ring-down spectroscopy and gas chromatography. *Agric. For. Meteorol.* 211: 48–57.
- Cortus, E.L., Jacobson, L.D., Hetchler, B.P., Heber, A.J. and Bogan, B.W. (2015). Methane and nitrous oxide analyzer comparison and emissions from dairy freestall barns with manure flushing and scraping. *Atmos. Environ.* 100: 57–65.
- Cristofanelli, P., Bracci, A., Sprenger, M., Marinoni, A., Bonafè, U., Calzolari, F., Duchi, R., Laj, P., Pichon, J.M., Roccato, F., Venzac, H., Vuillermoz, E. and Bonasoni, P. (2010). Tropospheric ozone variations at the Nepal climate observatory-pyramid (Himalayas, 5079 m a.s.l.) and influence of deep stratospheric intrusion events. *Atmos. Chem. Phys.* 10: 1483–1516.
- Davidson, E.A. and Kanter, D. (2014). Inventories and scenarios of nitrous oxide emissions. *Environ. Res. Lett.* 9: 105012.
- Escudero, M., Lozano, A., Hierro, J., Valle, J.D. and Mantilla, E. (2014). Urban influence on increasing ozone concentrations in a characteristic Mediterranean agglomeration. *Atmos. Environ.* 99: 322–332.
- Fu, X., Feng, X., Zhu, W., Zheng, W., Wang, S. and Lu, J. Y. (2008). Total particulate and reactive gaseous mercury in ambient air on the eastern slope of the Mt. Gongga area, China. *Appl. Geochem.* 23: 408–418.
- Gligorovski, S., Strekowski, R., Barbati, S. and Vione, D. (2015). Environmental implications of hydroxyl radicals (•OH). *Chem. Rev.* 115: 13051–13092.
- IPCC (2001). *Climate Change 2001: Synthesis report*. <http://www.grida.no/publications/267>
- Ji, D.S., Wang, Y.S. and Sun, Y. (2008). A comparative study on gaseous air pollutants in Beijing and London. *Chinese J. Environ. Eng.* 2: 1199–1206 (in Chinese with English Abstract).
- Ji, D.S., Wang, Y.S., Wang, L.L., Chen, L., Hu, B. and Tang, G.Q. (2012). Analysis of heavy pollution episodes in selected cities of northern China. *Atmos. Environ.* 50:

- 338–348.
- Ji, D.S., Wang, L.L., Wang, Y.S., Zhang, J.K., Cheng, M.T. and Sun, Y. (2014). The heaviest particulate air-pollution episodes occurred in northern China in January, 2013: Insights gained from observation. *Atmos. Environ.* 92: 546–556.
- Jia, L. and Xu, Y.F. (2014). Effects of relative humidity on ozone and secondary organic aerosol formation from the photooxidation of benzene and ethylbenzene. *Aerosol Sci. Technol.* 48: 1–12.
- Jonsson, O., Andersson, C., Forsberg, B. and Johansson, C. (2013). Air pollution episodes in Stockholm regional background air due to sources in Europe and their effects on human population. *Boreal Environ. Res.* 18: 280–302.
- Lam, K.S., Wang, T.J., Wang, T., Tang, J., Kajii, Y., Liu, C.M. and Shim, S.G. (2004). Overview of surface ozone variability in East Asia-North Pacific region during IGAC/APARE (1994–1996). *J. Environ. Sci.* 16: 599–609.
- Li, L., Lai, W., Pu, J., Mo, H., Dai, D., Wu, G. and Deng, S. (2017). Polar organic tracers in PM_{2.5} aerosols from an inland background area in Southwest China: Correlations between secondary organic aerosol tracers and source apportionment. *J. Environ. Sci.-China* 69: 281–293.
- Lin, M., Horowitz, L.W., Oltmans, S.J., Fiore, A.M. and Fan, S. (2014). Tropospheric ozone trends at Mauna Loa Observatory tied to decadal climate variability. *Nat. Geosci.* 7: 136–143.
- Lin, W., Xu, X. and Zhang, X.H. (2008). Contributions of pollutants from North China Plain to surface ozone at the Shangdianzi GAW Station. *Atmos. Chem. Phys.* 8: 5889–5898.
- Lin, W., Xu, X. and Sun, J. (2011). Characteristics of gaseous pollutants at Jinsha, a remote mountain site in Central China. *Sci. China Ser. B* 41: 136–144.
- Lin, W., Xu, X., Zheng, X., Dawa, J., Baima, C. and Ma, J. (2015). Two year measurements of surface ozone at Dangxiong, a remote highland site in the Tibetan Plateau. *J. Environ. Sci.* 31: 133–145.
- Liu, F., Zhang, Q., Tong, D., Zheng, B., Li, M., Huo, H. and He, K.B. (2015). High-resolution inventory of technologies, activities, and emissions of coal-fired power plants in China from 1990 to 2010. *Atmos. Chem. Phys.* 15: 13299–1331.
- Liu, H., Hu, B., Wang, Y.S., Liu, G.R., Tang, L.Q., Ji, D.S., Bai, Y.F., Bao, W.K., Chen, X., Chen, Y. M., Ding, W.X., Han, X.Z., He, F., Huang, H., Huang, Z.Y., Li, X.R., Li, Y., Liu, W.Z., Lin, L.X., Ouyang, Z., Qin, B.Q., Shen, W.J., Shen, Y.J., Su, H.X., Song, C.C., Sun, B., Sun, S., Wang, A.Z., Wang, G.X., Wang, H.M., Wang, S.L., Wang, Y.S., Wei, W.X., Xie, P., Xie, Z.Q., Yan, X.Y., Zeng, F.J., Zhang, F.W., Zhang, Y.J., Zhang, Y.P., Zhao, C.Y., Zhao, W.Z., Zhao, X.Y., Zhou, G.Y. and Zhu, B. (2017). Two ultraviolet radiation datasets that cover China. *Adv. Atmos. Sci.* 34: 805–815.
- Lupu, A. and Maenhaut, W. (2002). Application and comparison of two statistical trajectory techniques for identification of source regions of atmospheric aerosol species. *Atmos. Environ.* 36: 5607–5618.
- Ma, J., Lin, W.L., Zheng, X.D., Xu, X.B., Li, Z. and Yang, L.L. (2014). Influence of air mass downward transport on the variability of surface ozone at Xianggelila Regional Atmosphere Background Station, southwest China. *Atmos. Chem. Phys.* 14: 5311–5325.
- Mauzerall, D.L., Jacob, D.J., Fan, S.M., Bradshaw, J.D., Gregory, G.L. and Sachse, G.W. (1996). Origin of tropospheric ozone at remote high northern latitudes in summer. *J. Geophys. Res.* 101: 4175–4188.
- Meng, Z., Zhang, R., Lin, W., Jia, X., Yu, X., Yu, X. and Wang, G. (2014). Seasonal variation of ammonia and ammonium aerosol at a background station in the Yangtze River Delta region, China. *Aerosol Air Qual. Res.* 14: 756–766.
- Monks, P.S. (2000). A review of the observations and origins of the spring ozone maximum. *Atmos. Environ.* 34: 3545–3561.
- Monks, P.S., Archibald, A.T., Colette, A., Cooper, O., Coyle, M., Derwent, R., Fowler, D., Granier, C., Law, K.S., Mills, G.E., Stevenson, D.S., Tarasova, O., Thouret, V., Von Schneidmesser, E., Sommariva, R., Wild, O. and Williams, M.L. (2015). Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. *Atmos. Chem. Phys.* 15: 8889–8973.
- National Bureau of Statistics of the People's Republic of China (2017). <http://www.stats.gov.cn/english/>
- Necki, J., Schmidt, M., Rozanski, K., Zimnoch, M., Korus, A., Lasa, J., Graul, R. and Levin, I. (2003). Six-year record of atmospheric carbon dioxide and methane at a high-altitude mountain site in Poland. *Tellus B* 55: 94–104.
- Orru, H., Ebi, K.L. and Forsberg, B. (2017). The interplay of climate change and air pollution on health. *Curr. Environ. Health Rep.* 4: 504–513.
- Pu, W.W., Shi, X.F., Ma, Z.Q., Zhao, X.J., Zhang, X.L. and Xu, X.F. (2015). Impact of atmospheric pollutants transport pathways on aerosol optical properties at Shangdianzi background station. *Environ. Sci.* 36: 379–387 (in Chinese with English Abstract).
- Qu, W.J., Zhang, X.Y., Arimoto, R., Wang, D., Wang, Y. Q., Yan, L.W. and Li, Y. (2008). Chemical composition of the background aerosol at two sites in southwestern and northwestern China: Potential influences of regional transport. *Tellus B* 60: 657–673.
- Qu, W.J., Zhang, X.Y., Arimoto, R., Wang, Y.Q., Wang, D., Sheng, L.F. and Fu, G. (2009). Aerosol background at two remote CAWNET sites in western China. *Sci. Total Environ.* 407: 3518–3529.
- Ran, L., Lin, W.L., Deji, Y.Z., La, B., Tsering, P.M., Xu, X.B. and Wang, W. (2014). Surface gas pollutants in Lhasa, a highland city of Tibet – current levels and pollution implications. *Atmos. Chem. Phys.* 14: 10721–10730.
- Reddy, B.S.K., Reddy, L.S.S., Cao, J.J., Kumar, K.R., Balakrishnaiah, G., Gopal, K.R., Reddy, R.R., Narasimhulu, K., Lal, S. and Ahammed, Y.N. (2011).

- Simultaneous measurements of surface ozone at two sites over the Southern Asia: A comparative study. *Aerosol Air Qual. Res.* 11: 895–902.
- Saunio, M., Bousquet, P., Poulter, B., Peregon, A., Ciais, P., Canadell, J.G., Dlugokencky, E.J., Etiope, G., Bastviken, D., Houweling, S., Janssens-Maenhout, G., Tubiello, F.N., Castaldi, S., Jackson, R.B., Alexe, M., Arora, V.K., Beerling, D.J., Bergamaschi, P., Blake, D.R., Brailsford, G., Brovkin, V., Bruhwiler, L., Crevoisier, C., Crill, P., Covey, K., Curry, C., Frankenberg, C., Gedney, N., Höglund-Isaksson, L., Ishizawa, M., Ito, A., Joos, F., Kim, H.S., Kleinen, T., Krummel, P., Lamarque, J.F., Langenfelds, R., Locatelli, R., Machida, T., Maksyutov, S., McDonald, K.C., Marshall, J., Melton, J.R., Morino, I., Naik, V., O'Doherty, S., Parmentier, F.J.W., Patra, P.K., Peng, C., Peng, S., Peters, G.P., Pison, I., Prigent, C., Prinn, R., Ramonet, M., Riley, W.J., Saito, M., Santini, M., Schroeder, R., Simpson, I.J., Spahni, R., Steele, P., Takizawa, A., Thornton, B.F., Tian, H., Tohjima, Y., Viovy, N., Voulgarakis, A., van Weele, M., van der Werf, G.R., Weiss, R., Wiedinmyer, C., Wilton, D.J., Wiltshire, A., Worthy, D., Wunch, D., Xu, X., Yoshida, Y., Zhang, B., Zhang, Z. and Zhu, Q. (2016). The global methane budget 2000–2012. *Earth Syst. Sci. Data* 8: 697–751.
- Seinfeld, J.H. and Pandis, S.N. (2016). *Atmospheric chemistry and physics: From air pollution to climate change*. John Wiley & Sons, New York.
- Sun, L., Xue, L., Wang, T., Gao, J., Ding, A., Cooper, O.R., Lin, M.Y., Xu, P.J., Wang, Z., Wang, X.F., Wen, L., Zhu, Y.H., Chen, T.S., Yang, L.X., Wang Y., Chen, J.M. and Wang, W.X. (2016). Significant increase of summertime ozone at Mt. Tai in central eastern China: 2003–2015. *Atmos. Chem. Phys.* 16: 10637–10650.
- Thunis, P., Miranda, A., Baldasano, J.M., Blond, N., Douros, J., Graff, A., Janssen, S., Juda-Rezler, K., Karvosenoja, N., Maffei, G., Martilli, A., Rasoloharimahefa, M., Real, E., Viaene, P., Volta, M. and White, L. (2016). Overview of current regional and local scale air quality modelling practices: Assessment and planning tools in the EU. *Environ. Sci. Policy* 65: 13–21.
- Vingarzan, R. (2004). A review of surface ozone background levels and trends. *Atmos. Environ.* 38: 3431–3442.
- Vojtišek-Lom, M., Beránek, V., Klír, V., Jindra, P., Pechout, M. and Voříšek, T. (2018). On-road and laboratory emissions of NO, NO₂, NH₃, N₂O and CH₄ from late-model EU light utility vehicles: Comparison of diesel and CNG. *Sci. Total Environ.* 616: 774–784.
- Wang, B., Li, J., Wan, Y., Li, Y.E., Qin, X., Gao, Q., Waqas, M.A., Wilkes, A., Cai, W., You, S. and Zhou, S. (2018). Responses of yield, CH₄ and N₂O emissions to elevated atmospheric temperature and CO₂ concentration in a double rice cropping system. *Eur. J. Agron.* 96: 60–69.
- Wang, H., Zhu, B., Shen, L., Xu, H., An, J., Pan, C. and Liu, D. (2016). Regional characteristics of air pollutants during heavy haze events in the Yangtze River Delta, China. *Aerosol Air Qual. Res.* 16: 2159–2171.
- Wang, Y.Q., Zhang, X.Y. and Draxler, R.R. (2009). TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data. *Environ. Modell. Software* 24: 938–939.
- Wang, Y.Q., Zhang, X.Y., Sun, J.Y., Zhang, X.C., Che, H.Z. and Li, Y. (2015). Spatial and temporal variations of the concentrations of PM₁₀, PM_{2.5} and PM₁ in China. *Atmos. Chem. Phys.* 15: 13585–13598.
- Watson, R.T., Meira Filho, L.G., Sanhueza, E. and Janetos, A. (1992). Greenhouse gases: Sources and sinks. *Clim. Change* 92: 25–46.
- Weinhold, B. (2008). Ozone nation: EPA standard panned by the people. *Environ. Health Perspect.* 116: A302–A305.
- Wu, C., Wu, D. and Yu, J.Z. (2018). Quantifying black carbon light absorption enhancement with a novel statistical approach. *Atmos. Chem. Phys.* 18: 289–309.
- Wuebbles, D.J., Fahey, D.W., Hibbard, K.A., DeAngelo, B., Doherty, S., Hayhoe, K., Horton, R., Kossin, J.P., Taylor, P.C., Waple, A.M. and Weaver, C.P. (2017). Executive summary. In *Climate science special report: Fourth national climate assessment*, Volume I, Wuebbles, D.J., Fahey, D.W., Hibbard, K.A., Dokken, D.J., Stewart, B.C. and Maycock, T.K. (Eds.), U.S. Global Change Research Program, Washington, DC, USA, pp. 12–34, <http://science2017.globalchange.gov/>
- Xu, W.Y., Lin, W.L., Xu, X.B., Tang, J., Huang, J.Q., Wu, H. and Zhang, X.C. (2016). Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China-Part 1: Overall trends and characteristics. *Atmos. Chem. Phys.* 16: 6191–6205.
- Xu, W.Y., Xu, X.B., Lin, M.Y., Lin, W.L., Tarasick, D., Tang, J., Ma, J.Z. and Zheng, X.D. (2018). Long-term trends of surface ozone and its influencing factors at the Mt. Waliguan GAW station, China-Part 2: The roles of anthropogenic emissions and climate variability. *Atmos. Chem. Phys.* 18: 773–798.
- Xu, X., Lin, W., Wang, T., Yan, P., Tang, J., Meng, Z. and Wang, Y. (2008). Long-term trend of surface ozone at a regional background station in eastern China 1991–2006: Enhanced variability. *Atmos. Chem. Phys.* 8: 2595–2607.
- Ye, W.J., Bian, L.J., Wang, C., Zhu, R.B., Zheng, X.D. and Ding, M.H. (2016). Monitoring atmospheric nitrous oxide background concentrations at Zhongshan Station, east Antarctica. *J Environ. Sci.* 47: 193–200.
- Zhang, J., Sun, Y., Wu, F., Sun, J. and Wang, Y. (2014). The characteristics, seasonal variation and source apportionment of VOCs at Gongga Mountain, China. *Atmos. Environ.* 88: 297–305.
- Zhang, N., Cao, J., Ho, K. and He, Y. (2012). Chemical characterization of aerosol collected at Mt. Yulong in wintertime on the southeastern Tibetan Plateau. *Atmos. Res.* 107: 76–85.

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