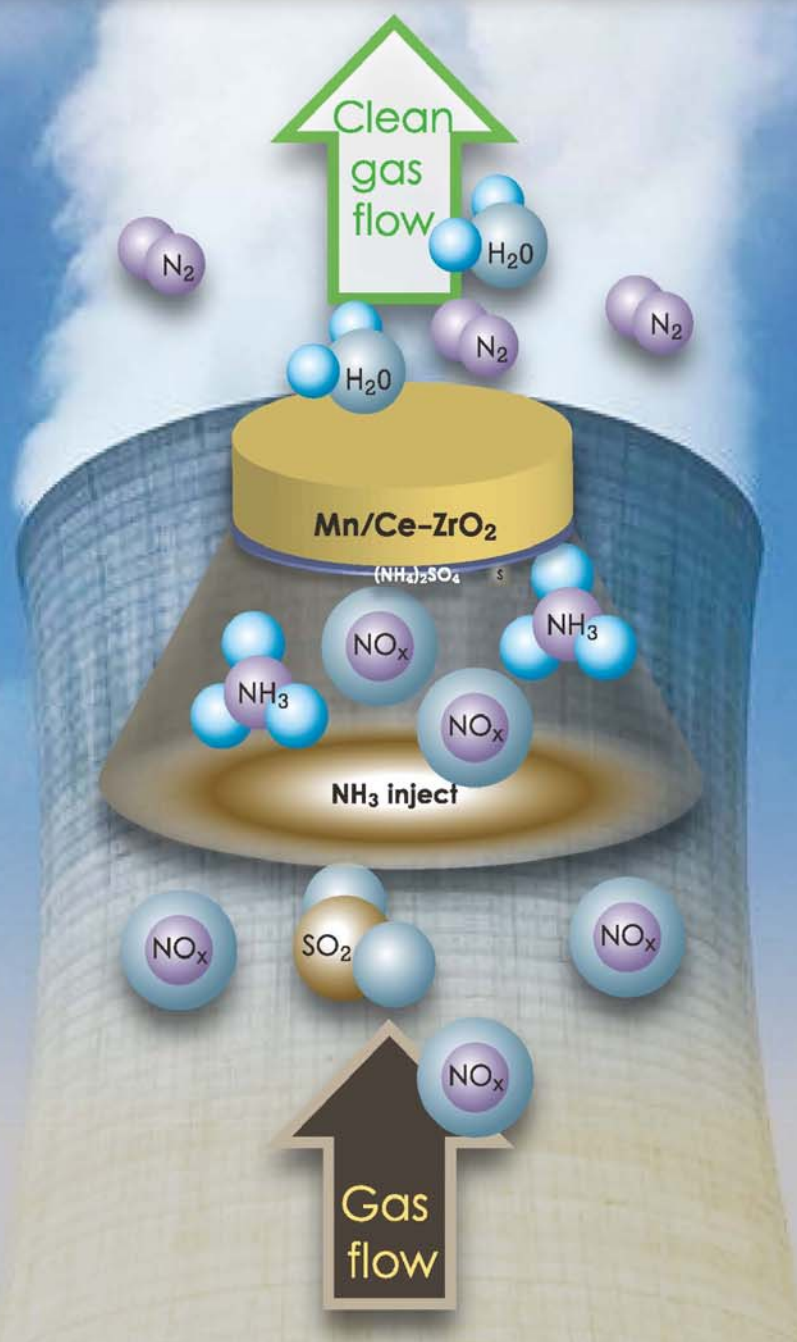


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Observed levels and trends of gaseous SO₂ and HNO₃ at Mt. Waliguan, China: Results from 1997 to 2009

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Abstract

Long-term measurements of SO₂ and HNO₃, particularly those from the background sites, are rarely reported. We present for the first time the long-term measurements of SO₂ and HNO₃ at Waliguan (WLG), the only global baseline station in the back-land of the Eurasian Continent. The concentrations of SO₂ and HNO₃ were observed at WLG from 1997 to 2009. The observed annual mean concentrations of SO₂ and HNO₃ at WLG were 1.28 ± 0.41 and 0.22 ± 0.19 $\mu\text{g}/\text{m}^3$, respectively. The HNO₃ concentrations were much higher in warmer seasons than in colder seasons, while the SO₂ concentrations showed a nearly reversed seasonal pattern. In most months, the concentration of HNO₃ was significantly correlated with that of SO₂, suggesting that some common factors influence the variations of both gases and the precursors of HNO₃ may partially be from the SO₂-emitting sources. The SO₂ concentration had a very significant ($P < 0.0001$) decreasing trend (-0.2 $\mu\text{g}/(\text{m}^3 \cdot \text{yr})$) in 1997–2002, but a significant ($P < 0.05$) increasing trend ($+0.06$ $\mu\text{g}/(\text{m}^3 \cdot \text{yr})$) in 2003–2009. The HNO₃ concentration showed no statistically significant trend during 1997–2009. While the decrease of SO₂ in 1997–2002 agrees with the trend of global SO₂ emissions, the increase in 2003–2009 is not consistent with the decreasing trends in many other regions over the world. Trajectory analysis suggests that the airmasses from the northern Qinghai-Tibetan Plateau and the Takla Makan Desert regions contributed significantly to the increasing trends of SO₂ and HNO₃ at WLG in 2003–2009, with a rate of $+0.13$ $\mu\text{g}/(\text{m}^3 \cdot \text{yr})$ and $+0.007$ $\mu\text{g}/(\text{m}^3 \cdot \text{yr})$, respectively.

Key words: SO₂; HNO₃; long-term trends; Waliguan; global baseline

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Introduction

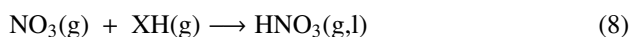
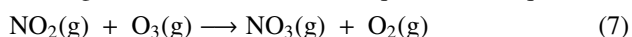
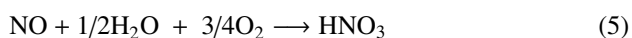
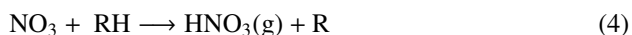
Sulphur dioxide (SO₂) is one of the key atmospheric species relevant to climate change, air quality, and ecosystems. The level of SO₂ in the natural atmosphere is usually low and sustained by natural sources, such as volcanic emissions, conversion of algae-producing dimethyl sulfide, etc. However, anthropogenic emissions have greatly enhanced the SO₂ level. Since 1962, human burning of fossil fuels has been adding SO₂ to the atmosphere at a rate equivalent to one “large” volcanic eruption each 1.7 years (Ward, 2009) and the anthropogenic component was substantially greater than natural emissions on a global basis (Smith et al., 2001). Atmospheric SO₂ reacts photochemically and on airborne particles to produce sulphate aerosol, which exerts a large influence on world climate (Charlson et al., 1987, 1991; Ward, 2009). Atmospheric

sulphate particles are active cloud condensation nuclei. An increase in the number of cloud condensation nuclei on a global scale may increase cloud albedo. Atmospheric SO₂ also plays a significant role in producing acid deposition by forming sulphate particles which return to the Earth's surface via dry or wet deposition processes (Galloway, 1989). Both processes transfer excess acidity to their deposition sites and the increasing acidity may damage sensitive ecosystems (Seinfeld and Pandis, 2006). Many measurements have been made in association with its role as a regional pollutant, particularly its role as a precursor of fine particles and acid rain. In view of the importance of SO₂, it was proposed to monitor atmospheric SO₂ within the Global Atmosphere Watch (GAW) programme of World Meteorological Organization (WMO, 2001). Since about two decades, SO₂ has been observed at a few global sites and many regional sites of the GAW network using various techniques. However, most of the GAW sites

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monitoring SO₂ are distributed in the European countries (<http://gaw.empa.ch/gawsis/default.asp>), where the levels and trends of SO₂ are well documented, and there have been very few measurements of SO₂ in the background atmosphere over China (Xu et al., 2009; Lin et al., 2008a; Meng et al., 2009).

The formation of nitric acid (HNO₃) in the troposphere can take place in a number of gas- and heterogeneous-phase reactions (Finlayson-Pitts and Pitts, 2000), including:



The characteristic time scale for HNO₃ production from NO_x is of the order of days. Once formed, HNO₃ is fairly stable because of its lower photolysis rate (Atkinson et al., 1997) in the actinic region and relatively slow hydroxyl radical (OH) attack rate (DeMore et al., 1997) at typical atmospheric temperatures and pressures. Therefore, HNO₃ production plays a key role in limiting the atmospheric cycling of OH and in controlling the oxidation rate of NO and the production of secondary oxidants O_x including O₃. Thus, characterizing the levels, production, and loss of HNO₃ will lead to better knowledge of the rates and yields of NO_x transformation and O_x production in the atmosphere.

Long-term measurements of SO₂ and HNO₃, particularly those from the background sites, are highly needed but have been scarcely reported. It is important to create databases suitable for the proper validation of models used to predict global or regional distributions of sulphate, nitrate, and photochemical products, and their present and future influences on climate, human health, and ecosystems. Long-term data of such gases may also be useful in the validation of satellite measurements. In this study, we present the long-term measurement results of ambient SO₂ and HNO₃ at the Mt. Waliguan Observatory, a WMO/GAW global baseline station in China, analyze and discuss the seasonal variations and trends of these gases, and explain the reasons for the observed trends by literature review and combined trajectory analysis.

1 Site and measurements

The Mt. Waliguan Observatory (WLG; 36.28°N, 100.90°E, 3810 m a.s.l.) is one of the global baseline stations of the WMO/GAW programme. It is situated on the northeastern edge of the sparsely populated

Qinghai-Tibetan Plateau and at an isolated mountain peak with an elevation of about 500 m relative to the surrounding landmass. WLG is about 90 km southwest of Xining (population: 2.13 million) and 260 km west of Lanzhou (population: 3.62 million). The surrounding areas are naturally preserved arid/semiarid lands and scattered grasslands. It is relatively dry, windy and short of precipitation with a typical continental plateau climate.

Ambient aerosol, HNO₃ and SO₂ samples were collected simultaneously by a train of filters in series, placed in a filter holder (Bio-Tech, Canada). The sample inlet is installed about 10 m above the ground and faces the ground to prevent dust fall. The sampling flow rate is recorded using a MKS flowmeter and a maximum flow rate of 50 LPM can be available using a high-volume Gast dual-diaphragm pump. The filter holder and all separators are made of Teflon material, which has been proved to be inert toward HNO₃ and SO₂ (Ayers and Gillert, 1990; Anlauf et al., 1985; Neuman et al., 1999).

The preparation and installation of filters was done in the Key Laboratory for Atmospheric Chemistry, China Meteorological Administration. The first layer is the Teflon filter (Gelman Sciences ZeffluorTM, 47 mm I.D., 2 μm) for aerosol collection. The second layer is the Nylon filter (Gelman Sciences Nylasorb^R, 47 mm I.D., 1 μm) for capturing gaseous HNO₃ and about 4% of gaseous SO₂. The third layer is the Whatman-41 filter (W41, soaked with 10% K₂CO₃/5% glycerin solution and then dried in vacuum) for collection of gaseous SO₂. After installation the filter assemblies were sealed and transported to WLG for field sampling.

From 1997 to 2002, weekly daytime (1:00–13:00 UTC) samples, nighttime (13:00–1:00 UTC) samples, and blank samples were collected for 7 days each. It was intended to see if there was a significant concentration difference between the daytime (more upslope winds) and the nighttime (more downslope winds). However, it was found that the daytime-nighttime concentration differences were insignificant. Therefore, 3-day continuous samples have been collected since 2003 to increase the time resolution of sampling, instead of the daytime and nighttime partition.

After sampling, the filter assemblies were sealed again and transported to the laboratory. The Nylon and W41 filters were extracted under sonication for two hours using a 0.5 mol/L NaCO₃/0.5 mol/L NaHCO₃ and 0.3% H₂O₂ solution and a 0.3% H₂O₂ solution, respectively. The extracted samplers were then analysed for NO₃[−] and SO₄^{2−} using ion chromatography (DX-500, Dionex). The ion chromatographic analysis followed the lab's QA/QC procedures for precipitation chemistry. The laboratory has been participating in the WMO-GAW Acid Rain Performance Survey since the late 1980s. The survey results show that for the 1997–2009 period, the lab and the “true” values agree within 2% for NO₃[−] and 8% for SO₄^{2−}. The NO₃[−] and SO₄^{2−} data were converted to the HNO₃ and

SO₂ masses that were captured by the filters. The concentrations of ambient HNO₃ and SO₂ were derived from the HNO₃ and SO₂ masses by considering the recorded actual flowrate and sampling duration.

2 Results and discussion

2.1 Seasonal variations and comparisons with data from other sites

From 1997 to 2009, a total of 1050 sets of samples were effectively collected and analyzed. **Figure 1** shows the average seasonal variations of the HNO₃ and SO₂ concentrations with (+1σ), and the ratio of HNO₃ to SO₂ (HNO₃/SO₂) at WLG. **Figure 1a** indicates that the concentrations of HNO₃ in warmer seasons were much higher than those in colder seasons. The monthly average HNO₃ concentration reached its maximum ($0.62 \pm 0.58 \mu\text{g}/\text{m}^3$) in August and minimum ($0.05 \pm 0.02 \mu\text{g}/\text{m}^3$) in February. The overall mean HNO₃ concentration during 1997–2009 was $0.22 \pm 0.19 \mu\text{g}/\text{m}^3$. For the purpose of comparison, we list here some measurement results of HNO₃ from a few sites over the world. For example, Dibb et al. (1998) reported a quite low mean HNO₃ concentration, $0.5 \text{ nmol}/\text{m}^3$, for the summers of 1994 and 1995 at Summit, Greenland. Gupta et al. (2003) reported an annual mean HNO₃ concentration of $0.7 \pm 0.6 \mu\text{g}/\text{m}^3$ and a summer to winter HNO₃ ratio of 3.8 at Rampur, a rural site in a semi-arid region of India. Lin et al. (2006) reported HNO₃ concentrations in the range of $0.4\text{--}5.3 \mu\text{g}/\text{m}^3$ with a mean value of $1.9 \pm 1.1 \mu\text{g}/\text{m}^3$ in Taichung City, an urban city of Central Taiwan, for the year 2002. They also showed that the average concentration of HNO₃ was $2.6 \pm 1.3 \mu\text{g}/\text{m}^3$ in the summer, which is 2.4 times

higher than that in the winter of 2002. Chiwa et al. (2008) summarized average concentrations of HNO₃ with a range of $0.3\text{--}3.9 \mu\text{g}/\text{m}^3$ at some forested and other locations in Japan and other places, with high HNO₃ concentrations in summer. Ferm et al. (2005) reported that the monthly HNO₃ concentration varied from 0.05 to $4.3 \mu\text{g}/\text{m}^3$ and annual concentrations from 0.16 to $2.0 \mu\text{g}/\text{m}^3$ at 11 rural and 23 urban sites in Europe and one rural site in Canada during 2002–2003. Adon et al. (2010) reported annual mean HNO₃ concentrations ranging from 0.6 to $1.4 \mu\text{g}/\text{m}^3$ for a few African sites. The level of HNO₃ at WLG is close to the lower values reported in the literature, consistent with the remoteness of this baseline site.

In the atmosphere, HNO₃ is primarily photochemically formed from the conversion of NO_x (Finlayson-Pitts and Pitts, 2000). In the remote and clean areas, like WLG, NO_x emission sources are mainly natural sources like soil and lightning. Wang et al. (2006) suggest that the reactive nitrogen at WLG is mainly from soil emission though long-range transport cannot be fully excluded. NO_x production and emissions from soils are controlled by a suite of environmental variables, especially soil temperature (Yienger and Levy, 1995). The high temperature in summer facilitates the emission of NO_x from soil. In the western region of China, the frequency and intensity of lightning in summer are 5–6 times higher than in winter (Christian et al., 2003). Therefore, it is reasonable that the resulting NO_x concentration in the western region is high in summer and low in winter. The results of satellite remote sensing on tropospheric NO₂ over WLG and its surrounding areas also show a similar seasonal variation (Zhang et al., 2007). In addition to the high level of NO_x in summer, the high summer level of HNO₃ also can result from the more rapid reaction between NO₂ and OH radical in summer, higher summer level of OH (Lin et al., 2008b), and the lower stability of particulate NH₄NO₃ at high temperatures in summer.

Figure 1b shows the seasonal cycle of SO₂ at WLG. The seasonal pattern of SO₂ looks quite different from that of HNO₃ (**Fig. 1a**), showing higher concentrations in colder months. The highest SO₂ concentration at WLG was $2.14 \pm 1.31 \mu\text{g}/\text{m}^3$ in January, followed by $1.95 \pm 1.15 \mu\text{g}/\text{m}^3$ in February, and the lowest value was $0.75 \pm 0.35 \mu\text{g}/\text{m}^3$ in September. The SO₂ concentration in June was $0.86 \pm 0.34 \mu\text{g}/\text{m}^3$, which is close to the lowest value in September. The overall average SO₂ concentration during 1997–2009 was $1.28 \pm 0.41 \mu\text{g}/\text{m}^3$. The seasonal amplitude of SO₂ looks much smaller than that of HNO₃. As an inland site, WLG should be less subject to natural sulfur emissions. Instead, anthropogenic emissions and physical/chemical process are the key factors determining the level and variation of SO₂ there. The relatively smaller seasonal amplitude of SO₂ at WLG leads to a seasonal pattern of the HNO₃/SO₂ ratio similar to that of HNO₃, as shown in **Fig. 1c**.

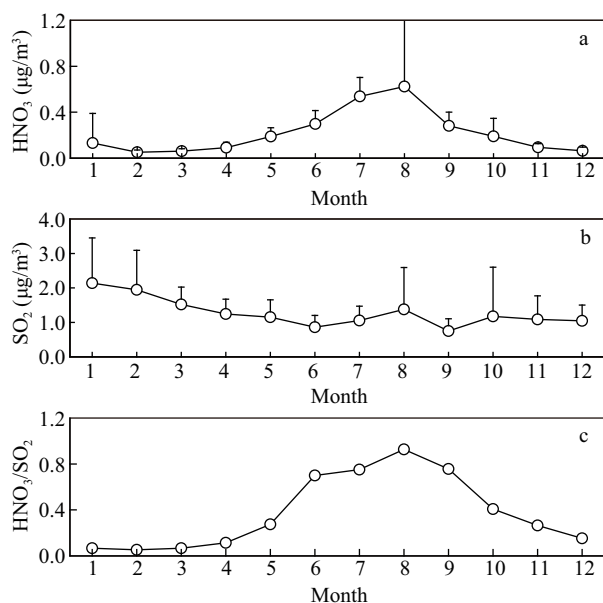


Fig. 1 Seasonal variations of HNO₃ (a) and SO₂ (b) concentrations (+1σ), and HNO₃/SO₂ ratio (c) at Waliguan.

Although the seasonal patterns of SO₂ and HNO₃ at WLJ look quite different, there was a statistically significant correlation between the concentrations of HNO₃ and SO₂. For the whole dataset, an empirical function, $[HNO_3] = 0.25 \times [SO_2] - 0.11$ ($R = 0.32$, $N = 1050$, $P < 0.0001$), can be obtained using the Reduced Major Axis Regression technique (Smith, 2009). The Reduced Major Axis Regression is a 2-sided regression technique, which is more suitable for regressions when both dependent and independent variables are measured with errors (in our case, SO₂ and HNO₃ are both measured with errors), while the standard ordinary least squares regression is more suitable for regressions when an independent variable has no error. The result suggests that some common factors influence the variations of both gases, creating the above-mentioned significant correlation ($P < 0.0001$). The common factors may include similar sources of SO₂ and NO_x (precursor of HNO₃) and removal processes for SO₂ and HNO₃ (e.g., deposition). However, these common factors seem not to be effective all the time. **Table 1** shows the regression results for different months. Although the correlation between HNO₃ and SO₂ is highly significant in most months, it is insignificant in October and December, and weak in June.

In **Table 2**, we list monthly average levels of SO₂ at

Table 1 Correlations between HNO₃ and SO₂ in different months at WLJ

Month	Intercept	slope	R ²	Number	P
1	-0.32	0.22	0.488	92	< 0.0001
2	0.02	0.02	0.181	86	< 0.0001
3	0.01	0.04	0.188	90	< 0.0001
4	-0.01	0.09	0.305	92	< 0.0001
5	-0.01	0.18	0.267	87	< 0.0001
6	0.07	0.25	0.053	76	0.03
7	0.05	0.42	0.251	78	< 0.0001
8	-0.09	0.49	0.496	90	< 0.0001
9	0.03	0.34	0.186	91	< 0.0001
10	-0.23	0.48	0.003	94	0.59
11	0.06	0.03	0.075	85	0.008
12	0.01	0.05	0.004	89	0.54
Overall	-0.11	0.25	0.105	1050	< 0.0001

four regional background stations in China, i.e., Shangdianzi (40.39°N, 117.07°E, 293.9 m a.s.l.), Lin'an (30.3°N, 119.73°E, 138 m a.s.l.), Longfengshan (44.73°N, 127.6°E, 310 m a.s.l.), and Jinsha (29°38'N, 114°12'E, 750 m a.s.l.), and at three sites in the relatively polluted North China Plain, i.e., Beijing urban (39.95°N, 116.32°E, 96 m a.s.l.), Gucheng (39.13°N, 115.12°E, 15.1 m a.s.l.) in Hebei Province, and Wuqing (39.39°N, 117.02°E, 5 m a.s.l.) in Tianjin City. The periods of the SO₂ datasets are 2005–2009 for Shangdianzi and Lin'an, 2005–2008 for Longfengshan, 2005–2007 for Jinsha, 2007–2009 for Beijing urban, 2006–2009 for Gucheng, and 2009 for Wuqing. Detailed analysis and interpretation of the SO₂ data from these stations can be found in the literature (Lin et al., 2009, 2011a, 2011b, 2012; Meng et al., 2009; Qi et al., 2012). For direct comparison, the unit of the SO₂ data from WLJ is converted to the mixing ratio, ppb.

As can be seen in **Table 2**, the SO₂ concentrations at different sites show similar seasonal patterns, with higher levels in colder months and lower levels in warmer months. In colder seasons, SO₂ is more strongly accumulated due to more emissions from heating, weaker photochemical conversion, and less strong atmospheric mixing. SO₂ is easily removed in warm and rainy seasons by rapid photochemical conversion, more rainfall, and better atmospheric diffusion. On average, the ratio of the winter (DJF) to summer (JJA) concentrations are 2.5, 4.8, 7.8, 18.2, and 1.5 for Lin'an, Shangdianzi, Longfengshan, Jinsha, and Waliguan, respectively. Among the background stations, Lin'an in China's most economically developed Yangtze River Delta region has the highest average SO₂ concentration (11.3 ± 4.4 ppb), followed by Shangdianzi in North China (7.5 ± 4.0 ppb), Jinsha in Central China (3.2 ± 3.2 ppb), and Longfengshan in Northeast China (2.5 ± 2.1 ppb). Waliguan, as a baseline GAW station, has the lowest SO₂ concentration (0.41 ± 0.14 ppb) among all the sites. Very high annual mean concentrations of SO₂ were observed at the Beijing urban site (16.8 ± 13.1 ppb) and at Gucheng (14.8 ± 9.4 ppb) a few years ago. To some extent, the levels of SO₂ at different sites can reflect the different

Table 2 Seasonal variations of SO₂ at several stations in China (unit: ppb)

Month	Beijing urban	Gucheng	Wuqing	Shangdianzi	Longfengshan	Lin'an	Jinsha	Waliguan
Jan	40.9	32.2	31.2	12.2	6.7	16.3	3.3	0.64
Feb	35.3	24.2		13.0	4.9	9.5	5.2	0.70
Mar	20.5	15.8		10.2	2.4	13.7	4.0	0.49
Apr	9.2	10.8		7.7	1.5	11.2	3.7	0.46
May	7.6	9.4		5.7	0.8	10.3	2.8	0.42
Jun	6.9	8.8		4.0	1.0	6.5	0.5	0.34
Jul	6.0	6.0	6.7	1.6	0.6	5.4	0.2	0.33
Aug	5.4	5.8	4.9	2.2	0.6	6.2	0.4	0.42
Sep	6.8	7.1	11.2	4.5	0.8	8.7	0.6	0.23
Oct	7.7	9.7	12.7	6.5	2.0	13.1	1.6	0.31
Nov	22.1	17.7	23.1	9.3	3.1	14.4	4.8	0.29
Dec	33.0	30.6	26.7	12.6	5.6	19.7	11.5	0.33
Overall	16.8	14.8		7.5	2.5	11.3	3.2	0.41
STD	13.1	9.4		4.0	2.1	4.4	3.2	0.14

energy demands and consumptions in certain regions with their different levels of economic development. In China, coal burning has been the largest anthropogenic source of SO_2 and main energy source. For a long time, the huge energy demands, high-sulfur content in coal, and lagging desulfurization technology have made China the highest SO_2 emitting country in the world (Stern, 2005). Coal-fired power production and industrial coal consumption are the two most important sources of SO_2 (Lu et al., 2010). Sparse population and less industrial activity in the vast western region of China are the reasons for the low SO_2 concentration at WLG.

2.2 Long-term trends

The time series of the HNO_3 and SO_2 concentrations and HNO_3/SO_2 ratio at WLG are shown in Fig. 2. Because of the change of sampling frequency at the end of 2002, the whole dataset is divided into two subsets, covering the 1997–2002 and 2003–2009 periods, respectively. Linear fittings are done respectively for the two subsets of data. As seen in Fig. 2, the monthly concentrations of atmospheric HNO_3 and SO_2 at WLG fluctuated within the ranges of

0.02–2.25 $\mu\text{g}/\text{m}^3$ and 0.12–5.78 $\mu\text{g}/\text{m}^3$, respectively. Most of the data points are distributed around the regression lines, while some of them show large deviations from the regression lines. The mean (median) concentrations of HNO_3 in 1997–2002 and in 2003–2009 were 0.21 ± 0.46 (0.08) $\mu\text{g}/\text{m}^3$ and 0.21 ± 0.34 (0.12) $\mu\text{g}/\text{m}^3$, respectively. The mean (median) concentrations of SO_2 in 1997–2002 and in 2003–2009 were 1.25 ± 1.39 (0.91) $\mu\text{g}/\text{m}^3$ and 1.27 ± 1.52 (0.74) $\mu\text{g}/\text{m}^3$, respectively. The SO_2 concentration in 1997–2002 had a very significant decreasing trend ($P < 0.0001$) of -0.2 $\mu\text{g}/(\text{m}^3\cdot\text{yr})$, but the SO_2 concentration in 2003–2009 showed a significant increasing trend ($P < 0.05$) of $+0.06$ $\mu\text{g}/(\text{m}^3\cdot\text{yr})$. The trends of HNO_3 for both periods are not statistically significant. The HNO_3/SO_2 ratio showed no significant trend in 1997–2002 but a significant downward trend ($P < 0.05$) in 2003–2009.

On the basis of a new estimate of global and country-level anthropogenic emissions of SO_2 over the 1850–2005 period, Smith et al. (2011) pointed out that global emissions peaked in the early 1970s and decreased until 2000. The SO_2 measurements in 1997–2002 at WLG show a de-

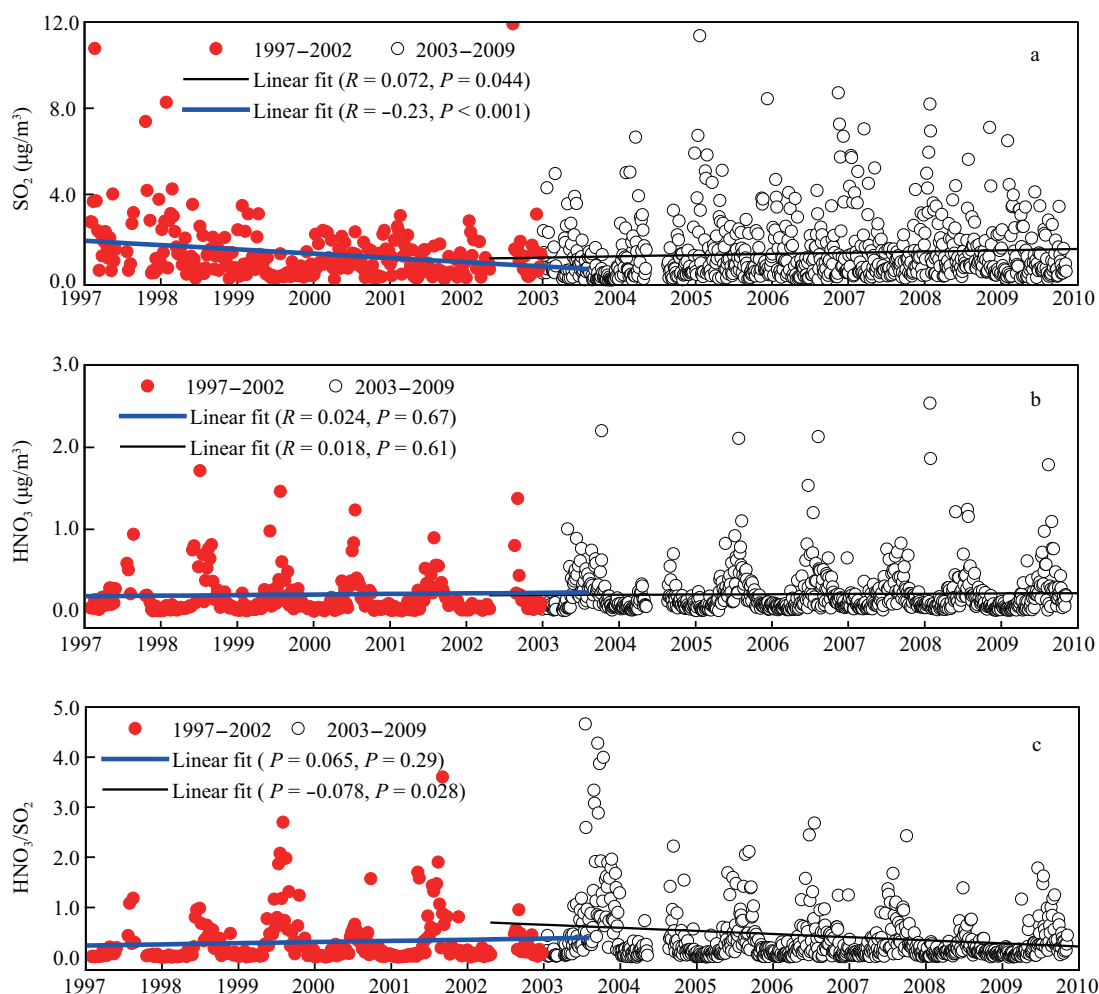


Fig. 2 Long-term trends of SO_2 (a) and HNO_3 (b) concentrations and HNO_3/SO_2 ratio (c) at Waliguan.

creasing SO₂ trend, consistent with the global decreasing emission trend. Smith et al. (2011) also pointed out that global anthropogenic emissions increased in recent years (2000–2005) because of increased emissions in China, international shipping, and developing countries in general. Lu et al. (2010) showed that the SO₂ emission in China increased dramatically starting in 2000, began to slow down around 2005, and then began to decrease after 2006. Based on the measurements using a passive sampling technique, Meng et al. (2010) reported an average SO₂ concentration of 0.7 ± 0.4 ppb in 2007–2008 for WLG. Comparing this value with the reported SO₂ concentration of 0.3 ppb in 1999–2000 for WLG (Carmichael et al., 2003), they concluded that the level of SO₂ in 2007–2008 had more than doubled within a decade. In our data, the mean (median) concentrations of SO₂ are 1.47 ± 1.75 $\mu\text{g}/\text{m}^3$ (0.91 $\mu\text{g}/\text{m}^3$) in 2007–2008 and 0.92 ± 0.77 $\mu\text{g}/\text{m}^3$ (0.727 $\mu\text{g}/\text{m}^3$) in 1999–2000. All these indicate a large increase of SO₂ at WLG from 1999–2000 to 2007–2008.

Recently, downward trends of atmospheric SO₂ have been observed in the economically developed eastern China. Qi et al. (2012) reported a significant downward trend of surface SO₂ from 2005 to 2010 with a rate of -2.4 ppb/yr ($P < 0.0001$) at Lin'an, located in the Yangtze Delta region, China. Significant downward trends of SO₂ in Beijing and in the North China region have also been reported (Lin et al., 2012; Zhang et al., 2011). SO₂ in Central Europe showed significant decreasing trends between 1991 and 2008 (Hamed et al., 2010). In USA, average SO₂ concentrations decreased substantially over the years from 1980 to 2010 (<http://epa.gov/airtrends/sulfur.html>). Against the background of the global decreasing trend of SO₂, the increasing trend of SO₂ at WLG may have resulted from enhanced emissions in the upwind areas, which has probably been caused by the Grand Western Development Program initiated around 2000 by the Chinese Central Government. The increase in energy demand caused by the economic development and the urbanization of West China (Huang et al., 2009) may have contributed more to the emission of SO₂ in the western regions. Recently, Zhang et al. (2012) presented the measurement results of tropospheric SO₂ from satellite data over China during 2004–2009 and conclude that the SO₂ load in West China is increasing continuously.

2.3 Influence of long-range air mass transport

The influence of air mass originating from different regions on the surface measurements is studied. Since WLG is a global baseline station and far from significant anthropogenic sources, the observed concentrations of SO₂ and HNO₃ (albeit short-lived) may be influenced by long-range transport of air masses if the emission sources in the upwind regions are strong enough. To see the potential impacts from the medium-range to long-range air transport, 120-hour backward trajectories were calculated and

clustered using the HYSPLIT model (version 4.9) and NCEP meteorological data (Draxler and Hess, 1998). In order to correlate the measurements with the air masses of different origins, backward trajectories were calculated for 12 UTC of the second sampling day of each sample during 2003–2009. The trajectory endpoint height was set to 100 m above ground level and the vertical motion method was set with the default model selection, which uses the vertical velocity fields of the meteorological model and is terrain following. Four trajectory clusters were obtained and their mean trajectories are shown in Fig. 3. The seasonal distributions of trajectories in each cluster and the corresponding SO₂ and HNO₃ concentrations are shown in Table 3.

The trajectories in cluster 1, which appear dominantly in winter, are from the west of WLG and almost cover the northern Qinghai-Tibetan Plateau and the Takla Makan Desert. The trajectories in cluster 2, which appear mainly in spring and autumn, are from the northwest sector of WLG, and the 120-hour trajectory can reach the Gobi area of Xinjiang, China. The number of trajectories in these two clusters amount to 55% of all the trajectories. The mean HNO₃ and SO₂ concentrations corresponding to these two clusters are close to each other and both lower than the mean values of the whole data. The trajectories in cluster 3 are from the northeast sector of WLG, with relatively shorter transporting paths, and mainly appear in summer and transition months, e.g., May and September. These

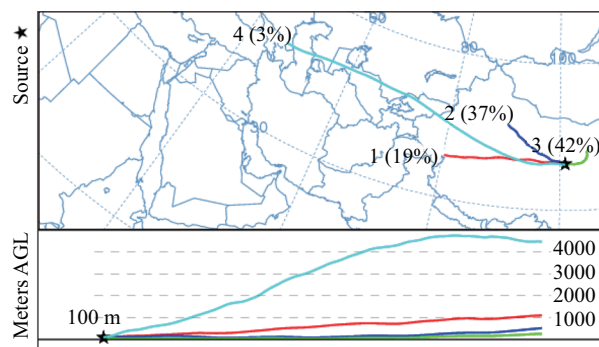


Fig. 3 Mean trajectories (120 hr) of four clusters with their ratios and geographic distributions. AGL: above ground level.

Table 3 Seasonal distributions of different clusters of trajectories and the SO₂ and HNO₃ concentrations corresponding to each cluster

Season	C1	C2	C3	C4
MAM	21	109	71	3
JJA	4	41	170	0
SON	30	89	82	4
DJF	100	63	18	16
Ratio	18.9%	36.8%	41.5%	2.8%
HNO ₃ ($\mu\text{g}/\text{m}^3$)	0.09	0.16	0.33	0.10
HNO ₃ -SD ($\mu\text{g}/\text{m}^3$)	0.09	0.23	0.46	0.08
SO ₂ ($\mu\text{g}/\text{m}^3$)	1.24	1.16	1.30	2.56
SO ₂ -SD ($\mu\text{g}/\text{m}^3$)	1.49	1.42	1.45	2.83

Ratio: the percentage ratio of the number of trajectories in certain cluster to the total trajectories.

trajectories are the most frequently appearing trajectories, accounting for 41.5% of the total number of trajectories. The mean concentration of HNO_3 corresponding to cluster 3 is notably higher than those for other clusters and also higher than the mean values of the whole data. The trajectories in cluster 4 are fast-moving ones, and appear least frequently and mainly in winter. Airmasses corresponding to this cluster of trajectories originate from the areas around the Mediterranean and Middle East, travel most of the time in the free troposphere, and descend finally to the surface of WLJ. These airmasses contain the highest concentration of SO_2 and a relatively low concentration of HNO_3 .

Based on the above clustering results, we further deduce the trends of HNO_3 and SO_2 at WLJ for airmasses of different origins during 2003–2009. As shown in Fig. 4, there are significant ($P < 0.05$) increasing trends of SO_2 and HNO_3 in cluster 1, with rate of $0.13 \mu\text{g}/(\text{m}^3 \cdot \text{yr})$ and $0.007 \mu\text{g}/(\text{m}^3 \cdot \text{yr})$, respectively. A significant decreasing trend of SO_2 with a rate of $-1.8 \mu\text{g}/(\text{m}^3 \cdot \text{yr})$ is found in cluster 4. No statistically significant trends are found in other clusters. Trajectories in cluster 1 are mainly from the northern Qinghai-Tibetan Plateau and the Takla Makan

Desert. According to National Bureau of Statistics of China (2011), the total emission of SO_2 in the whole country increased by about 20% from 2003 to 2006 and decreased by almost the same amount from 2006 to 2009. However, the total emission of SO_2 in the three western regions (Xinjiang, Qinghai, and Tibet) increased continuously from 0.392 million tons in 2003 to 0.728 million tons in 2009. Such continuous increase in SO_2 emission may be caused by the increases in economic activities, tourism, and populations in these western regions. The observed increasing trends of the SO_2 and HNO_3 concentrations in cluster 1 may be attributable to the enhanced emissions of SO_2 and NO_x in the northern Qinghai-Tibetan Plateau and the Takla Makan Desert regions. However, the emissions in the southern Asian countries (like India) have been increased in recent years, too (Smith et al., 2011), and the pollutant outflow from these countries may exert strong impacts on the air compositions over the surrounding areas (Lawrence and Lelieveld, 2010). This may have partially caused the observed trends in cluster 1. The decreasing trend of SO_2 in cluster 4 may be a result of the decreasing SO_2 trends over European areas, which were reported by Smith et al. (2011).

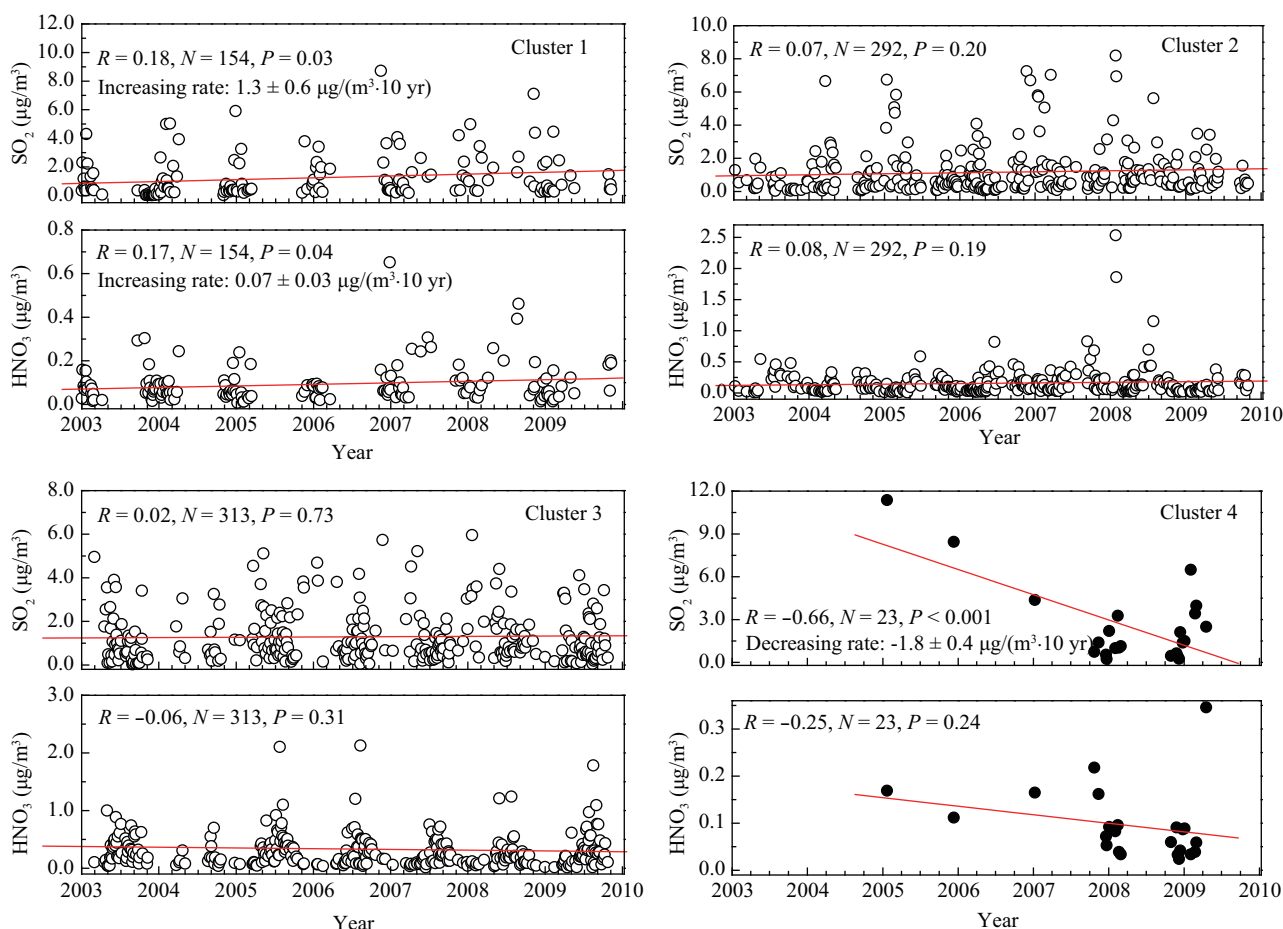


Fig. 4 Trends of the HNO_3 and SO_2 concentrations for airmasses of different origins at Waliguan during 2003–2009.

3 Conclusions

We have reported for the first time the long-term measurements of gaseous SO₂ and HNO₃ at the WLG observatory. The overall mean concentrations of SO₂ and HNO₃ during 1997–2009 were $1.28 \pm 0.41 \mu\text{g}/\text{m}^3$ and $0.22 \pm 0.19 \mu\text{g}/\text{m}^3$, respectively. The HNO₃ concentration was much higher in warmer months than in colder months, while the SO₂ concentration showed nearly reversed seasonal patterns. It is highly possible that the higher HNO₃ concentration in warmer seasons is caused mainly by enhanced soil emissions and higher oxidation rate of NO_x under warmer conditions. In most months, the concentrations of SO₂ and HNO₃ at WLG were significantly correlated, suggesting that the HNO₃ precursors may be partially from the sources emitting SO₂. The SO₂ concentration had a very significant decreasing trend ($P < 0.0001$) in 1997–2002 ($-0.2 \mu\text{g}/(\text{m}^3 \cdot \text{yr})$), but a significant increasing trend ($P < 0.05$) in 2003–2009 ($+0.06 \mu\text{g}/(\text{m}^3 \cdot \text{yr})$). No significant trend was found in the HNO₃ data. The observed negative trend of SO₂ in 1997–2002 can be well explained by the reported trend of global SO₂ emission. The positive trend of SO₂ at WLG in 2003–2009 is inconsistent with the negative trends in many regions over the world, including those found in eastern parts of China, Europe, and the USA. Trajectory analysis suggests that airmasses from the northern Qinghai-Tibetan Plateau and the Takla Makan Desert regions contributed significantly to the increasing trends of HNO₃ and SO₂ in 2003–2009. Airmasses from the areas around the Mediterranean and Middle East caused a decrease of SO₂ at WLG. These airmasses appeared least frequently; hence they could not change the general trend of SO₂ at WLG in 2003–2009. This study shows that although emissions of air pollutants in some major industrialized and populated regions have been significantly reduced, emissions can increase in some developing regions, which may exert impacts on the levels and trends of pollutants within and outside the regions and potentially cause environmental problems. More future measurements of key atmospheric species at remote sites are warranted to record their levels, variations, and long-term trends.

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