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RESEARCH ARTICLE

Long-term (2005–2015) trend analysis of $PM_{2.5}$ precursor gas NO_2 and SO_2 concentrations in Taiwan

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Abstract

Ground air monitoring stations have been installed in Taiwan since 1993 to ensure whether the criteria air pollutants meet the ambient air quality standards. In the present study, the data from the monitoring stations were used to evaluate long-term (2005–2015) trend of NO_2 and SO_2 in three metropolitan cities (northern Taipei, central Taichung, and southern Kaohsiung), two eastern coastal cities (Hualien and Taitung), and one agricultural city in west-central plain (Douliu); those cities essentially covered the entire region of Taiwan. The results indicate that SO_2 and NO_2 concentrations of all studied six cities meet the annual average standards of 30 and 50 ppb, respectively. After deseasonalizing the original data and using 7-month moving average, the trend analysis reveals a decreasing trend ranging from 0.15 to 0.57 ppb/year (R^2 from 0.33 to 0.85) for NO_2 and 0.06 to 0.45 ppb/year (R^2 from 0.32 to 0.92) for SO_2; the corresponding reductions over the 10-year span are 4 to 42% for NO_2 and 22 to 52% for SO_2. The reduction trend, despite the growth in GDP, vehicle numbers and energy consumption, industrial output, etc., is similar to those of developed countries. Clearly, there are seasonal/monthly variation patterns for these two precursor gases with minimum levels in summer (July) and maximum in winter (December). The concentration reductions, however, were lagging behind the respective emission reductions. There are significant correlations among six cites for NO_2 ($r = 0.58$–0.93) and, to some extent, SO_2 (0.32–0.66). The correlation between SO_2 and NO_2 ($r = 0.46$–0.74) indicates same or similar emission sources. Furthermore, the correlation between observed pollutant concentrations and their emission is excellent for SO_2 in two cities (0.79–0.96). The SO_2/NO_2 ratios vary with city and time and the value is site specific. For example, in 2005, the SO_2/NO_2 ratio was 0.38 in Kaohsiung and 0.18 in both Taipei and Taichung, the latter reflecting significant contribution from mobile sources. However, they all converged to 0.18–0.28 in 2015 in the six cities evaluated. All in all, the policies/measures made by the central and local government are effective in reducing ambient SO_2 and NO_2 levels.

Keywords Taiwan air quality · SO_2/NO_2 ratio · Ground air monitoring data · Long-term trend analysis · Seasonal/monthly variation pattern

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Introduction

As one of four "Asian Tigers," Taiwan has paid a huge environmental price due to rapid industrialization resulting in spectacular economic success in the 1970s. Despite the numerous efforts by Taiwan Environmental Protection Administration (TEPA, established in 1987) in combating air pollution, different air pollution problems are still causing public concerns and affecting public welfare. In general, the public are most concerned with the elevated levels of particulate matter (PM_{10} in the past and $PM_{2.5}$ nowadays) and O_3—both can cause haze conditions and related health problems. As a result, the recent air quality forecast focuses on Taiwan's main pollutants of PM_{10}, $PM_{2.5}$ and O_3. However, $PM_{2.5}$ or O_3 concentrations do not necessary reflect NO_2 and SO_2 levels. For example, between 2004 and 2012, there is an increasing trend of O_3 (0.94 ppb/year) in Taiwan (Chen et al. 2014) but with slight decreases in ambient NO_2 and SO_2 concentrations (Chen et al. 2015; Ding et al. 2016). On the other hand, there is some correlation between $PM_{2.5}$ and precursor gases, or the Pearson correlation coefficients between $PM_{2.5}$ and NO_2 and SO_2 were either high or moderate based on 286 monitoring sites in 31 Chinese provincial capital cities from 2013 to 2014 (Xie et al. 2015). As it is well known, photochemical reaction of converting SO_x and NO_x gases into aerosol sulfate and nitrate is an important process for secondary $PM_{2.5}$ formation. The contribution of sulfate and nitrate in $PM_{2.5}$ depends on locality, season, solar radiation, and particularly episode event (Wang et al. 2016)—the secondary $PM_{2.5}$ generally varies from 10 to 50%. For example, Fan et al. (2015) reported 25–30% sulfate in $PM_{2.5}$ in three pollution episodes in March 2012 in Pearl River Delta of China, while 18–24% sulfate was present in $PM_{2.5}$ (70–100 $\mu g/m^3$) in eastern Yangtze River Delta of China (Wang et al. 2016). Therefore, the precursor gases of SO_2 and NO_2 responsible for the formation of secondary $PM_{2.5}$ and O_3 should be paid more attention to.

In addition to the formation of secondary $PM_{2.5}$ and O_3, the environmental consequences caused by SO_2 and NO_2 are well known including acidification of lakes, acid rain, reduced visibility, affecting cloud formation, photochemical smog, and, more importantly, public health and environment deterioration (e.g., Zhang et al. 2016).

In Taiwan, only a few studies evaluated air quality trend in Taiwan: one particularly from 1994 to 2003 with the trend line slopes for SO_2 and NO_2 −2.05 and −0.51 ppb/year, respectively (Chang and Lee 2007). Between 1993 and 2012, NO_2 level was decreased by 16% with SO_2 reduction by 45% in Taipei (Ding et al. 2016)—these two studies only considered air quality in one city, Taipei. Unfortunately, there is no spatial and temporal study done in other areas of Taiwan regarding these two precursor gases. Thus, the present study is undertaken through long-term analysis of NO_2 and SO_2 in Taiwan to see the extent of progress made by different governmental policies/measures on the reduction of ambient SO_2 and NO_2 levels along with SO_x emission evaluation, to observe pollutant distribution among different cities in Taiwan, to evaluate monthly and seasonal variations of these two precursor gases, and to determine NO_2/SO_2 ratios so the extent of contribution from mobile sources can be evaluated. The results should provide TEPA with relevant information as to the effectiveness of different progresses made in Taiwan and to subsequently formulate future action plans to further reduce these pollutants in polluted regions to improve overall air quality.

The ground air quality monitoring station network was established in Taiwan in 1993. As of 2015, a total of 75 stations were installed including 26 in northern, 17 in central, 5 in eastern and 25 in southern region with 3 in offshore islands, covering all parts of Taiwan. The monitoring data are useful for TEPA to see whether these criteria pollutants meet the ambient air quality standards and correspondingly to ensure public health. The long-term data from these monitoring stations are also useful for performing trend analysis and evaluating the extent of effectiveness of pollutant emission reduction. Thus, we used data collected from monitoring stations in six cities in Taiwan, representing the northern, central, southern, and eastern regions of Taiwan to evaluate their long-term trend of NO_2 and SO_2 (2005 to 2015). Specifically, the trend deviation can be quantified and hotspots, if any, can be identified. Moreover, the reasons for increasing and decreasing trends are discussed. The future perspectives in reducing NO_2 and SO_2 emissions/concentrations are discussed with several recommendations to regulatory agencies to improve overall air quality.

Methodology

Monitoring stations in six cities

Six cities, representing the geographic distribution of Taiwan, were selected to analyze their trend of SO_2 and NO_2 concentrations. The six cities include two urban ones (Taipei, Taichung), one highly industrialized city (Kaohsiung), two remote east coastal cities (Hualien and Taitung), and Douliu, a small city located at west-center plain in a typical agriculture area. The relevant information for each of six cites including the location, area, number of monitoring stations used, population, vehicle numbers, and GDP is listed in Table 1 with their geographic location shown in Fig. 1; the coordinate of each monitoring station is shown in Table SM-1. In addition, nitrate and sulfate data were obtained from two supersites, located near two monitoring stations, to see any correlation between precursor gas and secondary $PM_{2.5}$.

Table 1 Relevant information for studied areas

City	Location	Area (km^2)	Number of stations used	Population[a]	Vehicles[b]	GDP[c]	TEPA NO_2				TEPA SO_2			
				(10^3)	(10^3)	(10^9 USD)	2005 (ppb)	2015 (ppb)	Annual decreasing rate (ppb/year)	Percentage decrease from 2005 to 2015 (%)	2005 (ppb)	2015 (ppb)	Annual decreasing rate (ppb/year)	Percentage decrease from 2005 to 2015 (%)
Taipei	25° 02′ N 121° 38′ E	276	5	2705	1820	81.7	26	20	0.57 ($R^2 = 0.77$)	23	4.8	3.1	0.20 ($R^2 = 0.77$)	35
Taichung	24° 08′ N 120° 41′ E	163	2	1155	1032	20.9	20	17	0.39 ($R^2 = 0.69$)	16	3.7	2.9	0.082 ($R^2 = 0.56$)	22
Kaohsiung	22° 37′ N 120° 19′ E	154	4	1516	1654	32.9	26	19	0.57 ($R^2 = 0.74$)	27	9.9	4.8	0.45 ($R^2 = 0.92$)	52
Douliu	23° 43′ N 120° 33′ E	94	1	108	107[d]	1.8[d]	15	15	0.21 ($R^2 = 0.32$)	4	4.1	3.2	0.056 ($R^2 = 0.30$)	22
Hualien	23° 58′ N 121° 36′ E	29	1	106	103[d]	2.1[d]	13	7.6	0.44 ($R^2 = 0.85$)	42	2.3	1.8	0.071 ($R^2 = 0.40$)	22
Taitung	22° 27′ N 121° 06′ E	110	1	107	106[d]	1.8[d]	6.9	5.5	0.15 ($R^2 = 0.40$)	20	2.1	1.2	0.12 ($R^2 = 0.67$)	43

[a] 2015 data; source: http://statdb.dgbas.gov.tw/pxweb/Dialog/varval.asp?ma=CS0201A1A&ti=&path=../database/CountyStatistics/&lang=9

[b] 2010 data including motorcycles; source: http://erdb.epa.gov.tw/DataRepository/ReportAndStatistics/StatSceMotors.aspx

[c] 2011 data for 2011, no update from 2012 for Taiwan Government; source: http://bbs.tianya.cn/post-333-303106-1.shtml

[d] Estimated from ratio of the population of these cities and their counties

Fig. 1 Map for selected six cities in Taiwan with star symbol indicating the monitoring stations used

QA/QC of monitoring station data

Each air quality monitoring station is required to establish a stringent quality assurance and quality control (QA/QC) program along with assurance standard operation procedures to ensure the quality of monitored data. The TEPA has contracted two independent companies to ensure QA/QC of these monitoring stations, including system and performance auditing. The detailed QA/QC procedures are described in TEPA document (www.taqm.epa.gov.tw/taqm/en/b0804.aspx). Briefly, SO_2 analyzing instrument is based on the principle of SO_2 absorbing UV at 220 to 240 nm (Table SM-2); the amount of produced fluorescence is directly proportional to the amount of SO_2. The NO_x analyzer is based on the theorem of chemiluminescence (Table SM-2); the instrument would release ozone into the reaction cell, and O_3 then reacts with nitric oxide. The strength of fluorescence is proportional to the concentration of nitric oxide.

The station evaluation is performed regularly including two elements: record checking and performance evaluation. The record checking ensures all documents to be certified and bubble meters require primary standards. Diluted standard gases of various concentrations were injected into the analysis instrument during audit with concentration ranging between zero and the maximum range on the instrument to evaluate its R^2. The gas calibration standards for station evaluation are required to be sent back to TEPA for quality assurance, and it also requires having the certificate issued by the third party to ensure the gas standards are adequate as those listed by National Institute of Standards and Technology and their standard reference materials or the their USEPA's national traceable reference materials. Table SM-3 lists quality assurance standards for monitoring facilities in performance audits. Essentially, the performance evaluation (contractor is Envimac Technology and Consultants) also includes site inspection, record keeping, instrument calibration (precision for SO_2 and NO_x should be ± 15% with linearity ≥ 0.995), manual calibration, and metrological facility, all following the detailed checklist.

Deseasonalized time series

The original monthly data from monitoring stations were first deseasonalized to remove any seasonal fluctuations and cyclical variations. The deseasonalized time series typically consists of the long-term trend and irregular components. The detailed methodology used can be found elsewhere (Zhang et al. 2017). To obtain long-term trend line, 7-month moving average was further employed to smooth data by reducing any random fluctuation. After plotting moving average data, the trend (slope) could then be easily determined using a simple linear regression technique. The magnitude of regression coefficient (R^2) can statistically determine the goodness of the trend line fit.

Results and discussion

NO_2 level in 2005

The ambient NO_2 concentrations in 2005 in Taiwan (7–26 ppb for 6 cities; Table 1) as well as those levels in 2015 (6–20 ppb; Table 2) certainly met the national annual average standard (50 ppb). For comparison, the annual mean concentration of

Table 2 Annual average of NO_2 and SO_2 in six cities in Taiwan during 2005–2015

Year	NO_2 (ppb)						SO_2 (ppb)					
	Taipei	Taichung	Kaohsiung	Douliu	Hualien	Taitung	Taipei	Taichung	Kaohsiung	Douliu	Hualien	Taitung
2005	26.4 ± 5.8	19.8 ± 4.2	25.9 ± 6.7	15.1 ± 5.6	13.1 ± 4.3	6.9 ± 2.1	4.8 ± 0.8	3.7 ± 0.7	9.9 ± 2.6	4.1 ± 0.7	2.3 ± 0.5	2.1 ± 0.7
2006	26.0 ± 3.7	20.8 ± 3.9	25.2 ± 7.2	16.3 ± 5.9	12.4 ± 3.2	6.6 ± 0.8	4.6 ± 0.4	3.5 ± 0.4	8.9 ± 1.8	3.8 ± 0.7	2.0 ± 0.3	2.3 ± 0.4
2007	25.9 ± 4.8	20.4 ± 3.6	23.9 ± 6.3	16.6 ± 5.2	11.8 ± 3.2	6.7 ± 1.2	4.3 ± 0.8	3.4 ± 0.4	8.7 ± 1.9	3.5 ± 0.5	2.2 ± 0.4	1.9 ± 0.5
2008	24.3 ± 3.7	19.8 ± 4.3	22.2 ± 7.5	16.2 ± 5.5	11.6 ± 3.3	6.5 ± 1.2	4.1 ± 0.4	3.7 ± 0.4	7.8 ± 1.6	3.3 ± 0.7	2.0 ± 0.4	2.3 ± 0.5
2009	22.2 ± 3.8	20.0 ± 3.1	21.7 ± 5.8	15.7 ± 3.9	10.8 ± 2.3	5.7 ± 1.3	3.3 ± 0.5	3.6 ± 0.4	7.5 ± 1.2	3.2 ± 0.4	2.4 ± 0.5	2.1 ± 0.4
2010	25.0 ± 4.1	19.9 ± 4.3	23.5 ± 6.1	16.5 ± 5.0	10.0 ± 2.7	6.6 ± 1.1	3.3 ± 0.5	3.5 ± 0.4	7.8 ± 1.4	3.3 ± 0.5	2.2 ± 0.3	2.0 ± 0.4
2011	23.3 ± 3.6	17.9 ± 4.1	22.6 ± 5.8	15.6 ± 4.4	10.1 ± 2.9	6.3 ± 1.0	3.0 ± 0.4	3.2 ± 0.5	7.0 ± 1.0	3.2 ± 0.4	2.0 ± 0.5	1.8 ± 0.4
2012	21.6 ± 4.1	17.7 ± 3.7	21.2 ± 4.9	14.3 ± 4.4	10.1 ± 2.7	6.5 ± 1.0	2.6 ± 0.3	2.8 ± 0.4	5.8 ± 0.6	3.1 ± 0.4	1.4 ± 0.4	1.3 ± 0.3
2013	21.5 ± 3.8	17.9 ± 3.1	21.0 ± 5.2	13.4 ± 4.1	9.6 ± 2.8	6.0 ± 1.0	3.0 ± 0.5	2.9 ± 0.4	5.8 ± 0.6	3.4 ± 0.6	1.5 ± 0.4	1.1 ± 0.3
2014	21.8 ± 3.4	17.4 ± 4.2	20.6 ± 5.5	13.9 ± 4.9	9.3 ± 2.8	5.1 ± 1.1	3.0 ± 0.4	3.1 ± 0.4	5.5 ± 0.6	3.5 ± 0.6	1.7 ± 0.2	1.3 ± 0.2
2015	20.4 ± 2.7	16.7 ± 3.7	19.0 ± 5.4	14.5 ± 4.7	7.6 ± 2.2	5.5 ± 0.7	3.1 ± 0.3	2.9 ± 0.2	4.8 ± 0.3	3.2 ± 0.5	1.8 ± 0.3	1.2 ± 0.1

NO_2 in EU in 2013 (EEA 2015) varied from approximately 5 ppb (Finland) to 15 ppb (Sweden) also meeting the EU NO_x standard (20 ppb), whereas the national daily maximum 1-h average concentration of NO_2 in 2015 in US was 56 ppb clearly meeting the standard of 100 ppb (USEPA 2016). Although monitored NO_2 levels of the six cities under study all meet the national annual mean standard as well as 1-h standard (250 ppb), there remains the further improvement of overall air quality.

It must be noted that the results from selected monitoring stations from four in Kaohsiung, two in Taichung, five in Taipei, and to one each in Douliu, Hualien, and Taitung certainly cannot represent the overall air quality of these cities; they only represent the average concentrations of the surrounding areas of those selected monitoring stations. For readers' comprehension, we nonetheless use city name to represent these areas. It is further noted that since the administrative boundary of Taichung and Kaohsiung has been changed since 2011 (so-called special municipalities), we use the old boundary before rezoning these two cities for the present study. Thus, the use of data must be made with care when results are compared with others.

The results for the east coastal cities of Hualien and Taitung (Fig. 1) exhibited much low levels of NO_2 in 2005 (13 and 7 ppb, respectively; Table 1), due to fewer industrial activities and lesser mobile sources in this region. The fact that Hualien has a higher NO_2 level than Taitung is due to Hualien being the center of Taiwan's cement industry, as was also reported that the visibility in Hualien is much less than that in Taitung (Tsai et al. 2007). In fact, the NO_x emission in Hualien county far exceeded that in Taitung county in 2005 (26.1 vs. 5.0 kt in 2005; TEPA 2017) with the one major industry in Taitung being Yuen Foong Yu Paper Manufacturing Co., located within the boundary of Taitung city.

Douliu, on the other hand, with much less population and vehicle number also exhibited the relatively high NO_2 level in 2005 (15 ppb). The main reason may be due to its geographic location and topography; it is located 10 km west of mountain terrains (1000 m above sea level and further east with famous Central Mountain Range which is 3000 m above sea level) and 6 km away from busy south-north highway with high NO_x emission. Consequently, Douliu, located on the leeside of the mountains, exhibits low wind speed and strong subsidence behavior that favor pollutant accumulation (Hsu and Cheng 2016).

As for the remaining three metropolitan cities, they all exhibited the similar NO_2 level in 2005 (20–26 ppb). The average data from the five monitoring stations were used to represent NO_2 and SO_2 concentrations in Taipei. Despite different monitoring stations used to represent city's ambient average SO_2 (4.8 ppb) and NO_2 (26 ppb) concentrations, the 2005 data were similar to those reported by Ding et al. (2016); for comparison, NO_x level in 2003 was 35 ppb (Chang and Lee 2007). Taipei, although lacking major industrial emissions, has a significant mobile pollution problem; vehicle emissions accounted for almost 84% of NO_x levels (ESC 2015).

Taichung power station, a 5500-MW coal fired plant, one of the top ten biggest power plants in the world, is the largest NO_2 and SO_2 emitter in central Taiwan, partially responsible for the high NO_2 level observed in Taichung. The other major stationary source in Taichung is Dragon Steel Corp., located about 20 km northwest of monitoring stations, producing steel plate, H-bean steel, billets, pig iron, etc., with total annual capacity of 6.16 Mt. From their environmental impact analysis, the upper limits for NO_x and SO_2 emission should be less than 5.31 and 4.1 kt (DSC 2012), respectively, with flue gas concentration of NO_2 and SO_2 less than 50 and 25 ppm, respectively, for all stacks (DSC 2012). Fortunately, with adequate treatment of flue gas emission, the observed annual NO_2 average in Taichung was able to meet the Taiwan's ambient air standard.

As noted in Table 1, the population, overall vehicle numbers, industrial activity, number of power plants, metrological condition, and GDP output are all different in these metropolitan cities. This led to different NO_x emissions in these three cities (NO_x emissions in 2005 were 44.1, 17.9, and 8.0 kt for Kaohsiung, Taipei, and Taichung, respectively; TEPA 2017). These emission differences along with other pollutant emissions clearly explain the variation in the annual $PM_{2.5}$ levels in 2005 in these three cities: the highest 55 $\mu g/m^3$ was found in Kaohsiung, followed by 42 $\mu g/m^3$ in Taichung and 26 $\mu g/m^3$ in Taipei (Ding et al. 2016). The mismatch between emission or emission intensity (different amounts) and concentration (same level) will be discussed later in the "SO_x emission" section.

It must be noted that in the ground level, NO_2 can be formed via reaction between NO and O_3. If the O_3 level is limited, the reaction may not occur efficiently. The produced NO_2 can be degraded under the condition of a sufficient flux of photons. The excited species then rapidly react with molecular oxygen to generate NO and O_3 (Ma et al. 2017).

NO_2 trend analysis

The fluctuation of the original data in Taipei is clearly shown in Fig. 2a with deseasonalized data in Fig. 2b. The NO_2 trend line with 7-month moving average (Fig. 2c) indicates decreasing trend about 0.57 ppb/year ($R^2 = 0.77$) or 23% reduction (Table 1) from 2005 to 2015 (from 26 to 20 ppb). Nonetheless, there was a slight increase NO_2 level in 2010 from 2009 data (Table 2); it is true for all cities studied as well as some cities in China (Wang et al. 2015). The reason is due to economic recovery from the global recession including Taiwan's recession from 2008 to 2009 (WWICS 2009). Overall, there are successful progresses made in reducing air pollution in Taiwan considering that the annual average NO_x level in 1994 was 50 ppb (Chang and Lee 2007). The long-term reduction rate, however, is getting lower; NO_x reduction from 1994 to 2003 was 2.05 ppb/year (Chang and Lee 2007) as compared to 0.57 ppb/year in the present study (2005–2015) (Table 1). As a result, further reduction needs more drastic changes in policies and different measures. Similar figures for other five cities including original, deseasonalized, 7-month moving average, and annual mean data are illustrated in Figs. SM-1 to SM-6.

The annual averages of NO_2 for these six cities during 2005 to 2015 are shown in Table 2 with box plots showing percentage distribution of annual concentrations illustrated in Fig. 3. There are more annual concentration variations in NO_2 as compared to SO_2 (Fig. 3). The decreasing NO_2 trend varies with cities with the highest in Taipei (0.57 ppb/year; $R^2 = 0.77$) and in Kaohsiung (0.57 ppb/year; $R^2 = 0.74$), followed by coastal area of Hualien (0.44 ppb/year; $R^2 = 0.85$). The corresponding NO_2 reductions from 2005 to 2015 ranged from 42% in Hualien to 27% in Kaohsiung and to merely 4% in Douliu. All in all, the decreasing NO_2 trend from

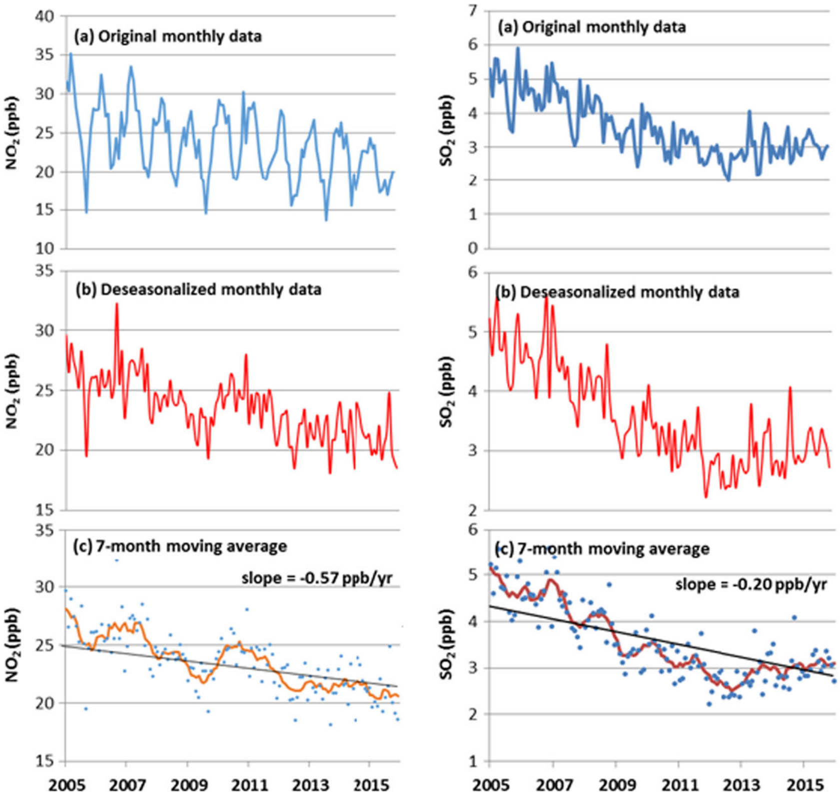


Fig. 2 Long-term original, deseasonalized, and 7-month moving average data in Taipei. **a** Original monthly data. **b** Deseasonalized monthly data. **c** Seven-month moving average

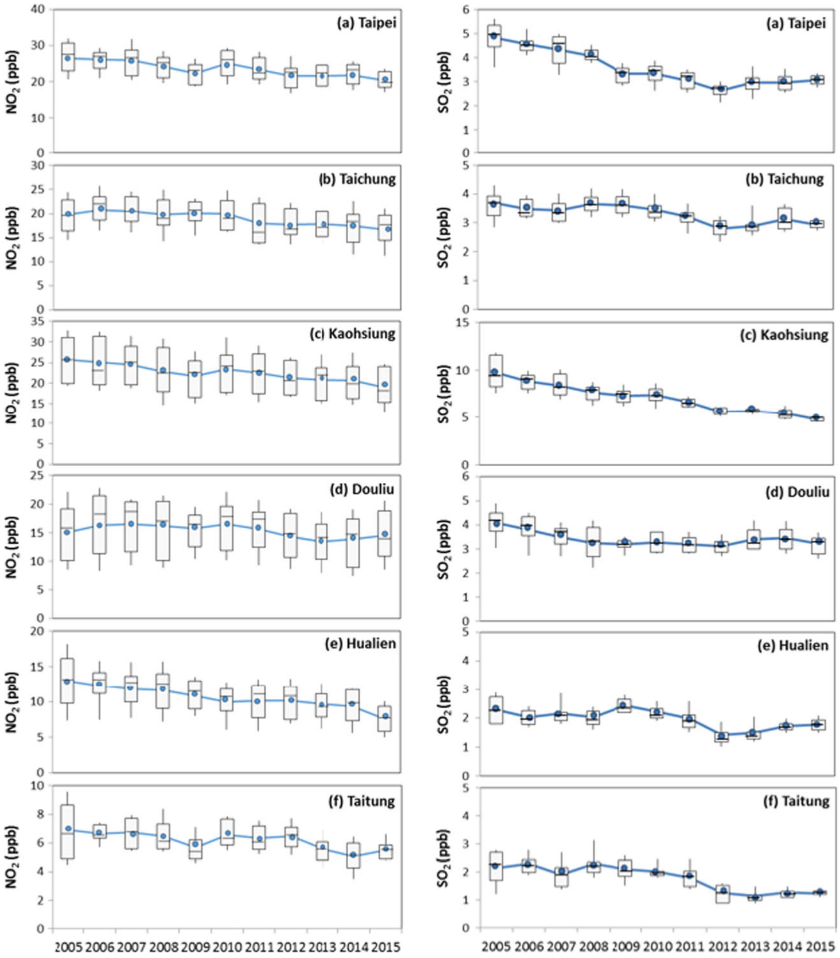


Fig. 3 Box plots showing distribution (10th, 25th, medium, 75th, 90th percentiles) and mean of annual ambient NO_2 and SO_2 concentrations. **a** Taipei. **b** Taichung. **c** Kaohsiung. **d** Douliu. **e** Hualien. **f** Taitung

2005 to 2015 clearly indicates the effectiveness of NO_x emission reduction via different governmental policies including upgraded vehicle emission standards, installation of deNO_x devices in flue gas treatment, as well as vehicle tailpipe emission control to meet the strict emission standards, effective in 2008. In particular, the utilization of denitrogenation processes in Taiwan cement industry certainly reduces NO_2 emission, resulting in significant reduction in ambient NO_2 (42%) concentration during the 10-year span in Hualien.

The long-range transport (LRT) of secondary NO_3^- and SO_4^{2-} from East Asia to neighboring countries is a well-known fact, e.g., the Chinese contribution of oxidized NO_x and SO_2 during the winter and spring to Japan and Korea due to westerly wind is significant (Qu et al. 2016), or contributing to about 38% of NO_3^- and SO_4^{2-} in Japan and 67% in Korea in winter (Duncan et al. 2016). Although the extent of precursor gas NO_2 and SO_2 transport is unclear, the following references clearly indicate some LRT contribution towards observed ambient SO_2 and NO_2 concentrations in Taiwan. A study from two background stations in Taiwan indicates LRT phenomenon doubling SO_2 concentration between 2001 and 2006 (Junker et al. 2009). In one episode event, a high SO_2 level (14 ppb) in the background Wanli station near Taipei was attributed to LRT (Lin et al. 2007). The impact of LRT on air quality in Taiwan is apparently more serious than people realize as Chen et al. (2015) suggest that the efforts of Taiwan in reducing air pollution are largely being negated by foreign contributions. On the other hand, pollutant transport out of Taiwan certainly happens under particular weather conditions. For example, images of SO_2 hot spots in near shores of western and northern parts of Taiwan from satellite-based ozone monitoring instrument have been observed (figure not shown). The concentrations were higher than some regions of inland Taiwan.

Finally, for comparison, except for some cities in China, India, Pakistan, and Middle East, the reduction trend is observed in all US cities, Europe, Mexico, Beijing, Shanghai, Japan, and Korea (Table 3). It appears that decreasing rates between 2005 and 2015 in Taiwan are comparable to those of other regions, e.g., from 2000 to 2015, 1-h daily maximum NO_2 level in the USA has been reduced by 60% over 15-year span (2000 to 2015) with 40 ppb in 2015 (USEPA 2016). In particular, the NO_2 air quality is almost comparable to those of EU countries, ranging from 5 to 15 ppb (EEA 2016). Thus, governmental regulations and policies towards reducing air pollutants in general and NO_2 level in particular are effective.

Table 3 Increasing/decreasing trend of ambient SO_2 and NO_2 concentrations in different countries/regions/cities

	NO_2	SO_2	Base years	Reference
Taiwan			2005–2015	This study
Taipei	0.57 ppb/year; 23% ↓	0.20 ppb/year; 35% ↓		
Taichung	0.39 ppb/year; 16% ↓	0.08 ppb/year; 22% ↓		
Kaohsiung	0.57 ppb/year; 27% ↓	0.45 ppb/year; 52% ↓		
Douliu	0.21 ppb/year; 4% ↓	0.06 ppb/year; 22% ↓		
Hualien	0.44 ppb/year; 42% ↓	0.07 ppb/year; 22% ↓		
Taitung	0.15 ppb/year; 20% ↓	0.12 ppb/year; 43% ↓		
Taiwan	1.4%/year ↓	6.6%/year ↓	1994–2010	Chen et al. (2015)
Taiwan	17% ↓	52% ↓	1994–2004	https://web.wpi.edu/Pubs/E-project/Available/E-project-050206-144504/unrestricted/IQP_Air_Pollution_in_Asia.pdf
USA national	22% ↓	64% ↓	Maximum 1-h level: 2005 to 2015	https://www.epa.gov/air-trends/nitrogen-dioxide-trends#nonat
227 sites (SO_2)	2015 level: 39 ppb (standard 100 ppb)	2015 level: 25 ppb (standard 75 ppb)		
178 sites (SO_2)				
Buffalo, USA	1.9%/year ↓	6.2%/year ↓	1980–2014 (annual average)	Civerolo et al. (2017)
New York City	2.4%/year ↓	5.9%/year ↓	1980–2014 (annual average)	Civerolo et al. (2017)
Hong Kong	Stay same (ca. 20–22 ppb in urban) 11% increase in roadside (38 ppb in 2015)	55% ↓	2005–2015	HKEPD (2015)
Sydney, Australia	25 ↓	20 ↓	2000–2012	OEH (2014)
EU member countries	27% ↓ in NOx emission	50% ↓ in emission	2002–2011	Guerreiro et al. (2014)
Czech				EEA (2013)
Traffic sites	13% ↓			
Urban sites	8% ↓			
Sofia, Bulgaria	15% ↓	33% ↓	2004–2012	Belis et al. (2015)
Budapest, Hungary	20% ↑	same	2004–2012	Belis et al. (2015)
Delhi	1 ppb/year ↑	0.33 ppb/year ↓	2005–2009	Mallik and Lal (2014)
Ontario, Canada	42% ↓	49% ↓	2005–2014	https://www.ontario.ca/page/air-quality-ontario-2014-report
Japan	30% ↓	50% ↓	2000–2010	Wakamatsu et al. (2013)
Korea	0.04–0.3 ppb/year ↓ dependent on land use	0.04–0.11 ppb/year ↓ dependent on land use	2003–2013	Yoo et all. (2015)
Seoul, Korea		16% ↓	1987–2013	Khan et al. (2017)
China 113 key cities		26% ↓	2005–2010	Wang and Hao (2012)
Beijing, China	11% ↓ (1998–2007)	53% ↓	1997–2007	Zhang et al. (2011)
Wuhan, China	0.36 ppb/year ↑	10% ↓	1996–2014	Song et al. (2016)
Istanbul, Turkey	33–5% ↑	21–28% ↓	2002–2010	Ozcan (2012)
Mexico City Metropolitan Area	Constant (2000–2010)	67% ↓	2000–2011	Benítez-García et al. (2014)
Athens traffic sites	13% ↓		1998–2008	Mavroidis and Chaloulakou (2011)

For the trend/reduction of long-term observations in some cities/regions/countries, data in Table 3 indicate that the progress made for NO_2 is less effective than that for SO_2 reduction. Again, the data comparsion in Table 3 must be made with care since time frame is different; reduction results from the past should be much better than those from recent studies considering deminishing return factor.

Seasonal and monthly variations of NO_2

Typically, the highest NO_2 concentrations were observed in winter or spring and lowest in summer (except slightly lower in autumn in Taipei) for six studied cities (Fig. 4). During the spring season, pollutant transport from Asian countries was most prevailing (Liu et al. 2003), partially responsible for the observed phenomena as also reported by the highest $PM_{2.5}$ level in spring in Taiwan (Chou et al. 2010). During winter time, northeasterly winds from China may bring pollutants to Taiwan. Moreover, the meteorological conditions (low mixing layer and stability) in Taiwan somehow favor the accumulation of pollutants in winter (Chen et al. 2004). On other hand, a high mixing layer in summer favors the pollutant dispersion resulting in lower pollutant concentrations. The wind during typhoon periods as well as weather conditions may bring clean oceanic fresh air to Taiwan in summer. Thus, the seasonal concentration variation is reflected by seasonal variation in sources (emissions), meteorological conditions (LRT, dispersion and stability), and subsequently their sinks (formation of secondary $PM_{2.5}$ and O_3).

For monthly variation, the lowest NO_2 levels were observed in July in cities studied except in September in Taipei, with the highest NO_2 levels noted in January, except for Taipei in March (Fig. 5). In July, in addition to high temperature resulting in a short NO_2 life time, Taiwan is always encountered with typhoons with an extremely high intensity of rainfall. This may partially explain lower NO_2 level in July due to the fact that rainfall can scavenge air pollutants (Yoo et al. 2014; Rivera-González et al. 2015).

SO_2 concentration and trend

For the 2005 data, the SO_2 levels all met the national annual standard of 30 ppb. Kaohsiung exhibited much higher level of SO_2 (9.9 ppb), almost doubling the values observed for other two metropolitan cities, Taipei (4.8 ppb) and Taichung (3.7 ppb). It is noted that the SO_2 level in Douliu (4.1 ppb) is higher than that in Taichung; reasons explained above for the higher NO_2 level are certainly applied here for SO_2 case.

The trend analysis indicates that the largest reduction between 2005 and 2015 is in Kaohsiung with 0.45 ppb/year ($R^2 = 0.92$), followed by Taipei (0.2 ppb/year; $R^2 = 0.77$) and Taitung (0.12 ppb/year; $R^2 = 0.56$) with the overall reduction of 52% in Kaohsiung (Table 1) and 35% in Taipei. As with the case of NO_2, the decreasing SO_2 trend slope with time is getting lower; from 1994 to 2003, SO_2 decreasing trend was 0.51 ppb/year in Taipei (Chang and Lee 2007), as compared to the current 0.2 ppb/year (2005 to 2015). For the same time span (2005–2015), the satellite-derived SO_2 trends (Krotkov et al. 2016) show not only significant reductions in parts of worlds (80% in Eastern USA; 65% in Eastern Europe; 60% in North China Plain) but also significant increases in India, e.g., 200% in Chhattisgarh and Odisha, India due to expanded industrial activities and more importantly no installation of FDG units in coal-fired power plants.

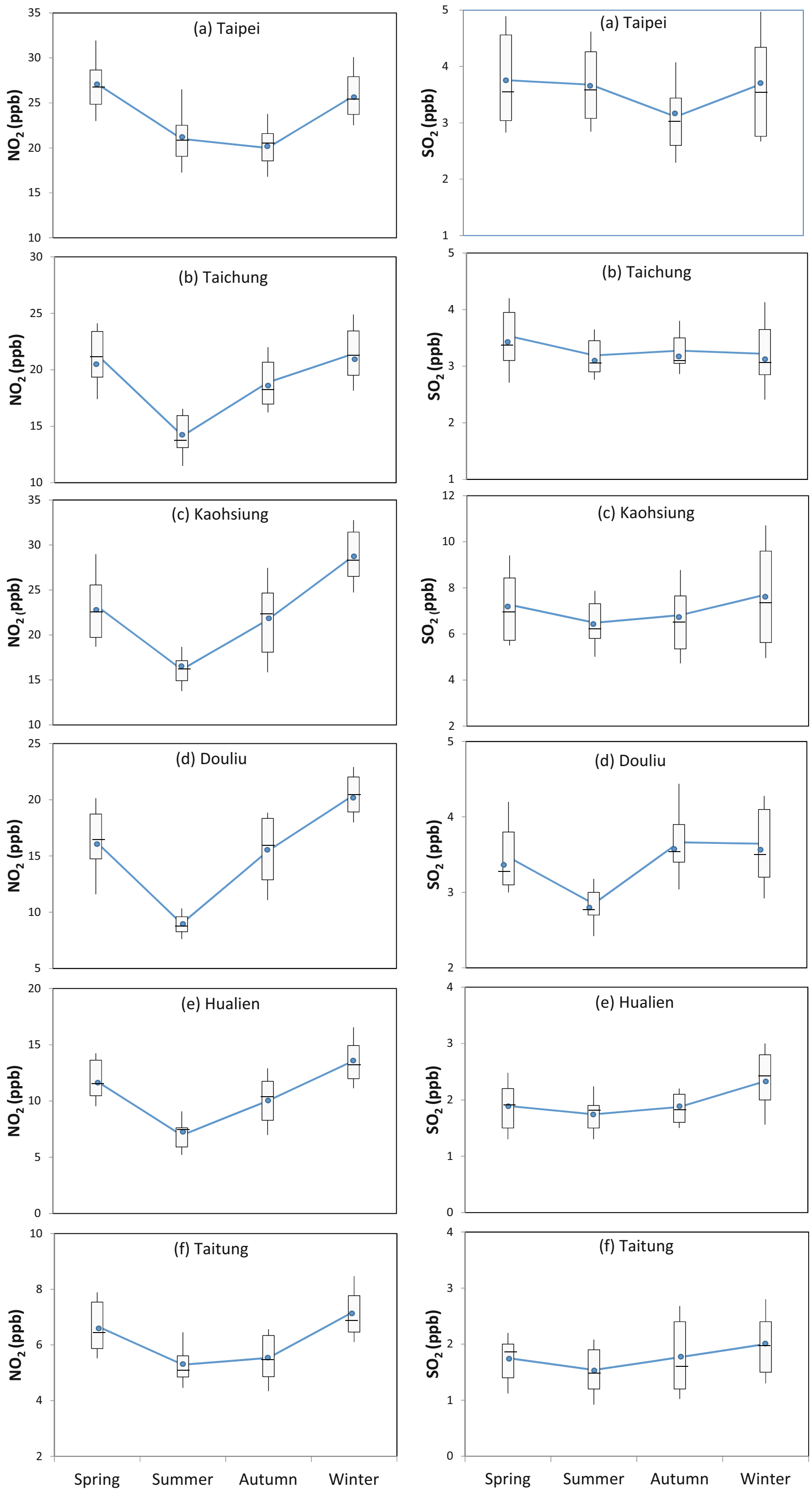
Seasonal/monthly variations of SO_2

As with the case of NO_2, the winter season exhibited the maximum SO_2 level (with almost the same level in spring in Taipei and Taichung) with summer being the lowest. The reasons (stability, rainfall, mixing layer height, among others) in explaining this phenomenon for NO_2 apply to the SO_2 case. One major difference, however, is that the SO_2 variation for seasonal average is much less than those observed for NO_2 (Table 4), e.g., SO_2 ranging from lowest 3.2 (summer) to highest 3.5 ppb (spring), or 9% difference in Taichung and from 6.5 to 7.7 ppb (18% difference) in Kaohsiung, whereas for NO_2, the corresponding ranges are from 14.1 to 21.4 ppb (50% difference) and 16.2–28.8 ppb (78% difference). This is also reflected by lower SO_4^{2-} content variations as compared to NO_3^- variations in $PM_{2.5}$ in three aerosol monitoring stations in Taiwan (Chou et al. 2017). As for monthly variations, unlike NO_2 case in which there is a clear valley in July, the minimum SO_2 levels vary with city, e.g., October in Taipei, February in Taichung, July in Kaohsiung and Taitung, and June in Douliu and Hualien (Fig. 5). The peak/valley of monthly difference between SO_2 and NO_2 is also observed in others (e.g., Pandey et al. 2015).

SO_x emission

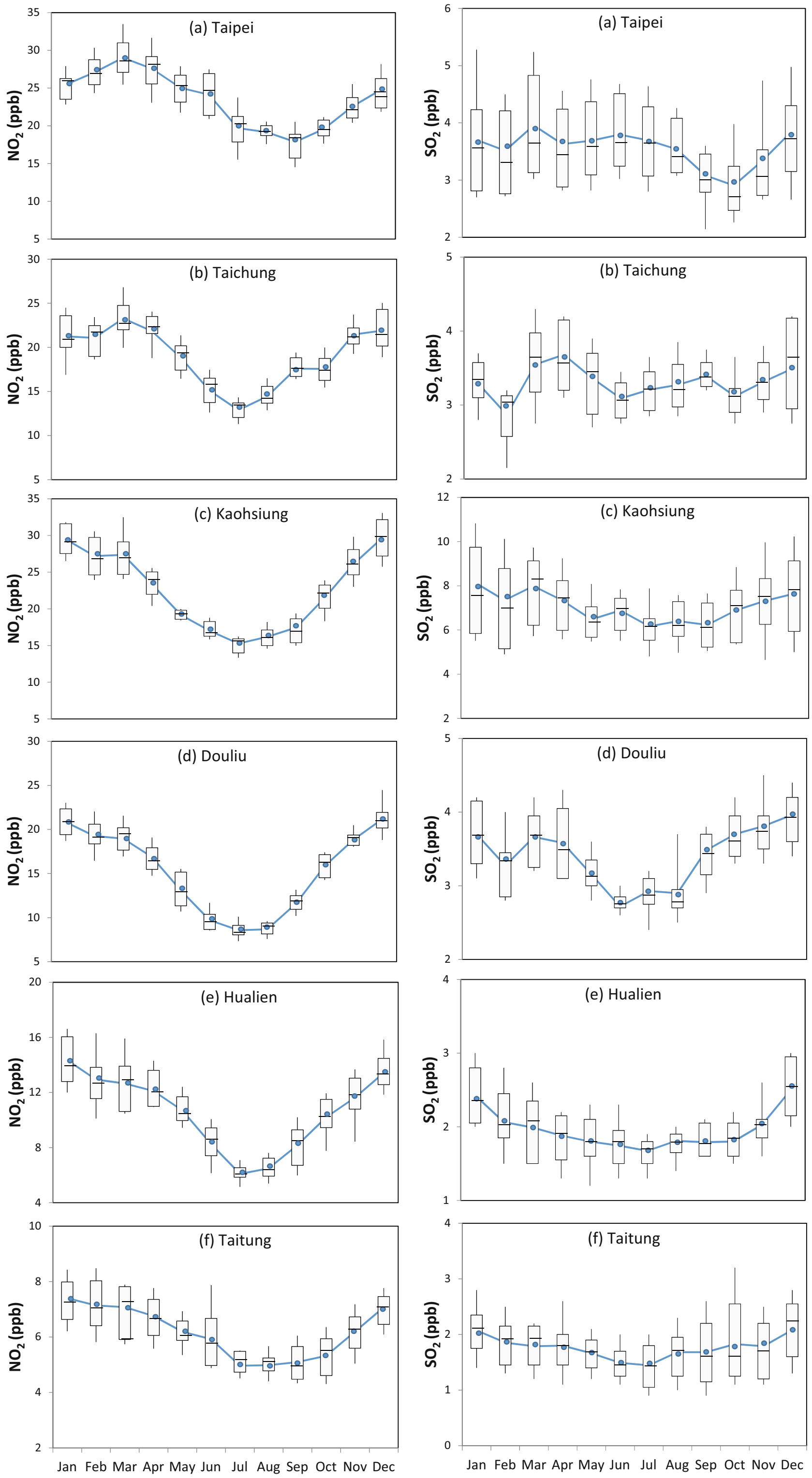
As noted in Fig. 6, the overall SO_2 emission trend shows a continued decreasing trend only in Kaohsiung while there are some fluctuations both in Taipei and in Taichung. The SO_x emission data were obtained from governmental document (TEDS 9.0; https://teds.epa.gov.tw/new_main2-0-1.htm). The total emission is the sum of point source, line (mobile) source, area (surface) source, and biogenic source. The procedures for estimating these emissions are illustrated in Figs. SM-7 to SM-9 for each of these sources. The reduction of SO_2 emission from 2005 to 2015 follows the order: 64% in Taipei, 37% in Kaohsiung, and 17% in Taichung (Table 5). For comparison, there was 50% SO_x emission reduction between 2002 and 2011 in EU-27 member countries (Guerreiro et al. 2014). The reduction in SO_x emission clearly led to reduction in ambient SO_2 concentrations as discussed above. The regression between observed SO_2 concentration and

Fig. 4 Seasonal variations (10th, 25th, medium, 75th, 90th percentiles) and mean of NO_2 and SO_2 for six cities in Taiwan during 2005–2015. **a** Taipei. **b** Taichung. **c** Kaohsiung. **d** Douliu. **e** Hualien. **f** Taitung



emission for two cites (Taipei and Kaohsiung) is excellent ($R^2 = 0.62$–0.92; Fig. 7). The reason for no correlation in Taichung is due to its emission fluctuation (Fig. 6). Nonetheless, the acid ecosystems due to reduction of SO_x emission in Taiwan surely would be improved as in the case of EU in that the area of sensitive ecosystems affected by excessive acidification from mainly SO_2 emissions has shrunk by 92% from 1990 to 2010 (EEA 2012).

Fig. 5 Monthly variations (10th, 25th, medium, 75th, 90th percentiles) and mean of NO_2 and SO_2 for six cities in Taiwan during 2005–2015. **a** Taipei. **b** Taichung. **c** Kaohsiung. **d** Douliu. **e** Hualien. **f** Taitung



One observation is puzzling; the emission of SO_x in Kaohsiung in 2015 was 20.4 kt as compared to 0.4 kt in Taipei, but the observed ambient SO_2 concentration over Kaohsiung was only slightly higher than that over Taipei (i.e., 4.8 vs. 3.1 ppb; Table 2). The uncertainty in emission estimation, SO_2 conversion to sulfate, and SO_x emissions

Table 4 Seasonal SO_2 and NO_2 concentration variations for six cities in Taiwan during 2005–2015

Season	NO_2 (ppb)				SO_2 (ppb)			
	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter
Taipei	27.2 ± 3.3	21.0 ± 3.3	20.0 ± 3.1	25.8 ± 2.9	3.8 ± 0.9	3.7 ± 0.7	3.1 ± 0.8	3.7 ± 0.9
Taichung	21.3 ± 3.1	14.1 ± 1.9	18.9 ± 2.7	21.4 ± 2.8	3.5 ± 0.6	3.2 ± 0.4	3.3 ± 0.3	3.2 ± 0.6
Kaohsiung	23.3 ± 4.2	16.2 ± 2.0	21.7 ± 4.4	28.8 ± 3.1	7.3 ± 1.7	6.5 ± 1.2	6.8 ± 1.6	7.7 ± 2.4
Douliu	16.3 ± 3.2	9.0 ± 1.2	15.4 ± 3.3	20.4 ± 2.2	3.5 ± 0.5	2.8 ± 0.4	3.7 ± 0.5	3.6 ± 0.6
Hualien	11.8 ± 2.1	6.9 ± 1.6	10.0 ± 2.3	13.6 ± 2.4	1.9 ± 0.5	1.7 ± 0.4	1.9 ± 0.3	2.3 ± 0.6
Taitung	6.7 ± 1.1	5.3 ± 1.0	5.5 ± 0.9	7.2 ± 1.1	1.8 ± 0.5	1.5 ± 0.5	1.8 ± 0.7	2.0 ± 0.6

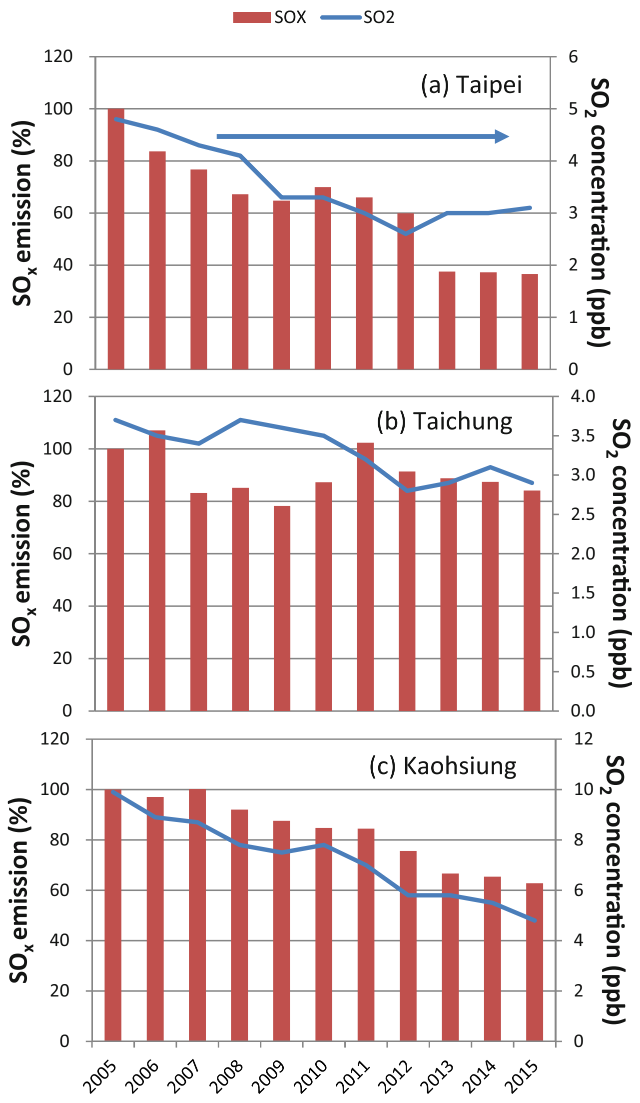


Fig. 6 SO_x emission trend (bar) for threes cites with SO_2 concentration (line) shown for comparison. **a** Taipei. **b** Taichung. **c** Kaohsiung

including shipping not reaching the local area due to high stacks in Kaohsiung cannot explain the observed fact. The inconsistency between emission and concentration has been also reported elsewhere (e.g., Xia et al. 2016).

SO_2/NO_2 ratio

Annual mean SO_2/NO_2 ratios (from 2005 to 2015) were calculated by using average of monthly SO_2/NO_2 ratios as shown in Table SM-4. The relative standard deviation ranges from 10 to 35%. The SO_2/NO_2 concentration ratio may provide a clue as to the extent of contribution from mobile sources since they release a significant amount of NO_x as well as effectiveness of FGD and $DeNO_x$ air pollution control devices in stationary point sources. Thus, the ratio is the site-specific, depending on the season, time, locality, industrial activity, and land use pattern, among others. For example, the ratio was 1.16 (vol base) in 1998 in Beijing, clearly indicating the contribution from SO_2 emission (coal burning) and it decreased to 0.51 in 2007 demonstrating the impact of NO_x emission (traffic) (Zhang et al. 2011). On the other hand, SO_2/NO_2 ratio in one particular site in Delhi increased from 0.52 to 1.1 between 2007 and 2009 (Biswas et al. 2011), demonstrating significant contribution from SO_2 emission from newly developed stationary sources including power plants.

For the three metropolitan cities exhibiting the similar NO_2 but slightly different SO_2 levels in 2005, the SO_2/NO_2 ratios for these cities are completely different with the highest ratio of 0.38 in Kaohsiung and 0.18 in both Taipei and Taichung in 2005 (Fig. 8). Clearly, low ratios represent a significant contribution from mobile sources and/or ineffective DeNOx devices for NO_x emission control. Thus, the extremely low ratios in Taipei and Taichung represent the significant contribution from mobile sources as confirmed by 84% NO_x contribution from mobile sources in Taipei and 90% in Taichung (ESC 2015), whereas Kaohsiung's contribution is also from local stationary petrochemical point sources (only 33% NO_x from vehicles) resulting in relatively high SO_2/NO_2 ratio. One interesting point is that only Taipei, Douliu, and Kaohsiung showed steady decreases of SO_2/NO_2 ratio with time; the

Table 5 Emission of SO_x in three metropolitan cities with decreasing rate from 2005 to 2015

City	SO_x emission		
	2005 (kt)	Annual decreasing rate (kt/year)	Percentage decrease from 2005 to 2015 (%)
Taipei	1.1	0.063 ($R^2 = 0.90$)	64
Taichung	0.4	0.003 ($R^2 = 0.12$)	17
Kaohsiung	32.5	1.32 ($R^2 = 0.96$)	37
Douliu	1.9 [a, b]	-[c] ($R^2 = 0.01$)	9.2
Hualien	2.0[a]	0.05 ($R^2 = 0.37$)	29
Taitung	0.2[a]	0.01 ($R^2 = 0.88$)	58

[a] Estimated from ratio of the population of these cities and their counties

[b] Data used from 2007 to 2015

[c] Insignificant

ratios for other cities varied with time demonstrating the variable contributions between point and mobile sources. They eventually converged to 0.18–0.28 in 2015 (Fig. 8).

For some cities, the SO_2/NO_2 ratio is extremely low, e.g., 0.01 (based on SO_2/NO) in Islamabad (Rasheed et al. 2014) and in New Delhi (Aneja et al. 2001); 0.02 in Rome with $PM_{2.5} = 19\ \mu g/m^3$ (Battista et al. 2016), while an extreme high SO_2/NO_y (total reactive nitrogen) ratio was reported in several rural/agricultural sites near Zhejiang, China (Wang et al. 2002), indicating a significant contribution from coal combustion, in particular high sulfur content in coal. The ratio for other cities/regions are comparable to those found in the present study or 0.25 in NYC with $PM_{2.5} = 16\ \mu g/m^3$ (Ito et al. 2007); 0.24 to 0.27 in South Korea (Yoo et al. 2014); 0.22 in Mexico City (Rivera-González et al. 2015); 0.36 in Xiamen, China (Shi et al. 2014); 0.15–0.55 (SO_2/NO_x) in different urban regions of India (Mallik and Lal 2014); and 0.27–0.31 in the urban region of Kolkata, India (Gupta et al. 2008). It must be pointed out that comparison of the ratios must be made with care since different years were used when the emission patterns of SO_2 and NO_x surely were different.

As for seasonal variation, typically winter season exhibited the highest SO_2/NO_2 ratio for all six cities evaluated (data not shown) indicating the higher contribution from point sources,

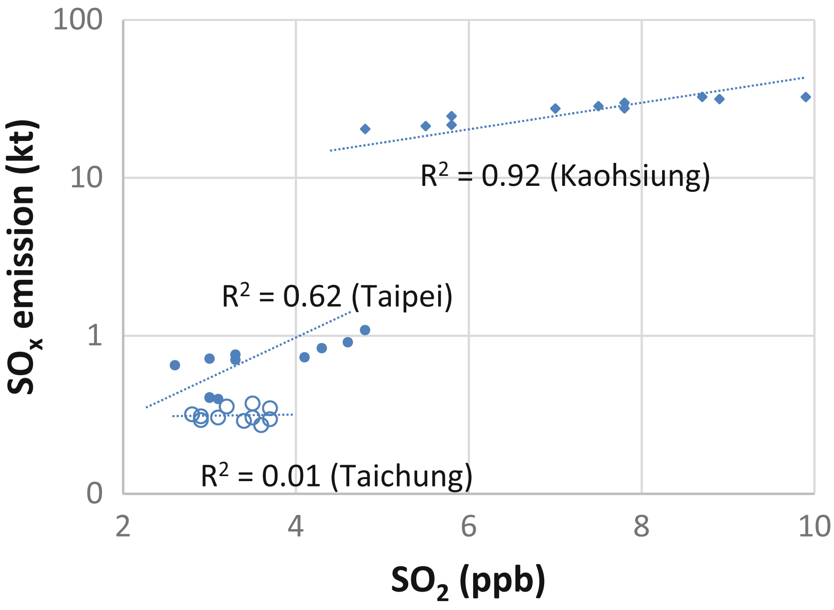
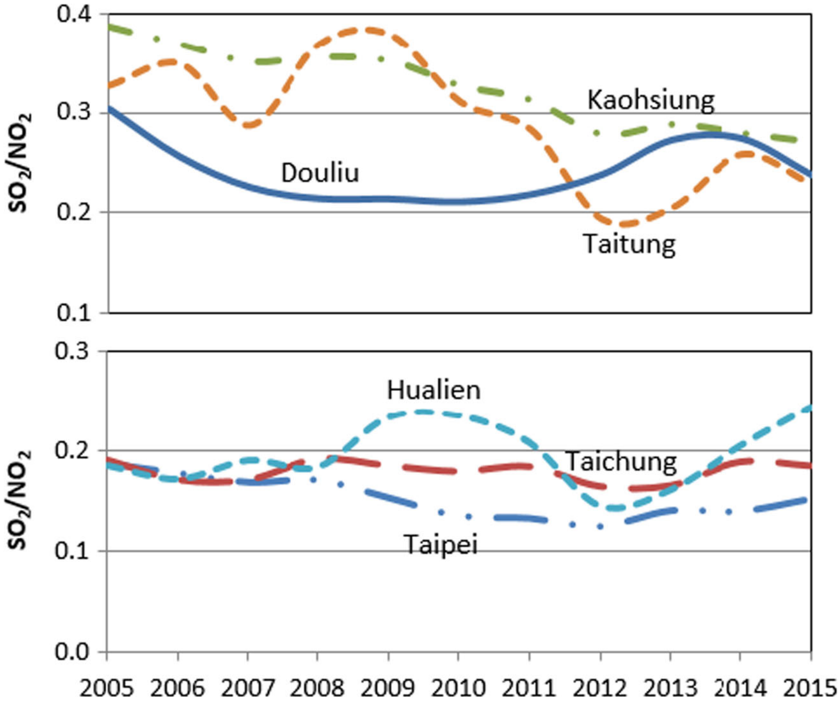


Fig. 7 Regression of annual SO_2 concentrations with the corresponding annual SO_x emissions for three cities

Fig. 8 Annual SO_2/NO_2 ratios for six cities

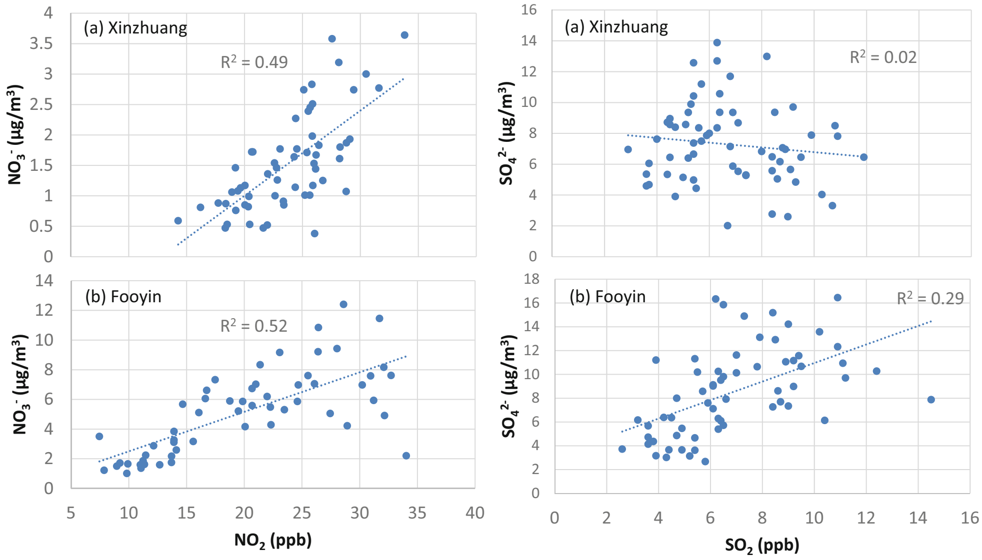


while summer or autumn the least; the results are similar to those reported by others (e.g., Beijing, Zhang et al. 2006 and urban region in India, Gupta et al. 2008).

Correlation between precursor gases and secondary $PM_{2.5}$

It is worth finding whether any correlation exists between NO_2 concentration and NO_3^- content in $PM_{2.5}$ as well as SO_2 with sulfate SO_4^{2-}; so, the extent of contribution of precursor gases to secondary $PM_{2.5}$ can be demonstrated. We chose two supersite monitoring stations to obtain the nitrate and sulfate content (2005 to 2010) and correlated their contents with the ambient NO_2 and SO_2 concentrations from the nearby monitoring stations (5 km from the Fooyin supersite station; 1.5 km from the Xinzhung supersite station). The results indicate some sort of correlation: $R^2 = 0.49$ for NO_2/NO_3^- for both Fooyin and Xinzhung Station (Fig. 9), but almost no correlation in Xinzhung ($R^2 = 0.02$) to poor correlation in Fooyin ($R^2 = 0.29$) for SO_2/SO_4^{2-}. A similar correlation ($R^2 = 0.20$) between precursor SO_2 and secondary sulfate is reported by others (Xie et al. 2015). The correlation may not

Fig. 9 Regression between NO_2 and SO_2 and their oxidized nitrate (NO_3^-) and sulfate (SO_4^{2-}). **a** Xinzhuang. **b** Fooyin



be as good as one would expected due to the distance between the supersite stations for NO_3^- and SO_4^{2-} data and ambient monitoring stations for NO_2 and SO_2 data. The effective SO_2 oxidation rate can be affected by NH_3; SO_2 is more localized in source areas while SO_4^2 spread over the region (Xing et al. 2015). Incidentally, the ratios of SO_4^{2-}/NO_3^- in these two supersites are completely different (6.0 for Xinzhung and 2.2 for Fooyin); the precursor gas ratios of SO_2/NO_2 0.25 and 0.35 in the respective nearby station are also different, demonstrating that the atmospheric NO_2 and SO_2 transformation to their respective oxidized form is different, especially considering the fact that part of NO_2 will also form O_3.

Conclusions and outlook

Both ambient SO_2 and NO_2 concentrations have met the national standards in the studied six cities. The distribution of these two pollutants follows the order of Kaohsiung > Taipei > Taichung > Douliu > Hualien > Taitung. Typically, winter exhibited the highest SO_2 and NO_2 level. The significant correlation of NO_2 pollutant among three metropolitan cites indicates similar NO_x emission sources. The different $SO_{2/}NO_2$ ratios in 2015 clearly indicate the extent of mobile contribution in two metropolitan cities (Taipei with ratio 0.15) and Taichung (018), as compared to industrial cities of Kaohsiung (0.26). The continued decrease of this ratio in Kaohsiung further indicates effective control of point sources from this cluster of petrochemical plants and power plants and to some extent increasing traffic volume.

Furthermore, ambient air concentrations of both SO_2 and NO_2 and their emissions have been declined in Taiwan, despite the increases in vehicles, fuel/energy consumption and economic growth. Nonetheless, O_3 and $PM_{2.5}$ are occasionally not meeting the standards and causing public concerns. As a result, the multi-pollutant multi-effect approach employed in 1999 Gothenburg Protocol as outlined by Elder et al. (2013) should be implemented. Essentially, to control individual, pollutant is not sufficiently cost-effective, since all pollutants and their effects are intertwined. One needs to seek simultaneous control of O_3 and $PM_{2.5}$ and thus other air pollutants such as VOCs. NH_3 and NO_x as well as SO_2 can be managed simultaneously. By the same token, the continued efforts with different measures towards energy saving and carbon reduction in Taiwan would also assist emission reduction in SO_x and NO_x since energy is closely related to air pollution.

The central government should outline the so-called action plan to combat air pollution problem. In doing so, different agencies should be put together for establishing a major task force to deal with different areas including urban planning, transport sector, renewable energy source, stringent emission standards for stationary and mobile sources, replacement of coal usage, establishment of more monitoring stations in hot spots (both area-wise and within industrial complex), and health risk assessment, among others. Case in point is that almost all past/current ambient SO_2 and NO_2 concentrations meet the 1-h and annual standards. If that is the case, why do people still complain inadequate air quality? Isn't it the time to tighten these standards? The timeline should be established to effectively meet a given immediate and long-term goal. After all, "Avoid, Innovate and Reduce" (AIR) (IEA 2016) are key elements for reducing air pollution. One important element is to set emission ceiling for problematic "Air Quality Districts" at a given year as similar ceiling has been established in the environmental impact assessment for new industry and/or expansion of industry in Taiwan.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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