Referee #2's comments

General comments:

Large reduction in emissions of anthropogenic pollutants in a mega city during the week-long holiday gives an opportunity to investigate how the urban air quality reacts to reductions in emission. This study conducted a systematical examination of the "Spring Festival effect" over three consecutive winters in Shenzhen with a population of greater than ten million, and the difference in the concentrations of various air pollutants between the Spring Festival (SF) and non-Spring Festival (NSF) periods was exploited to indicate that the origins of pollutants are primarily local or regional.

Although "holiday effect" has been discussed in a large number of studies in recent decades, the rich and comprehensive dataset provided in this study is informative and helpful for understanding of sources of a large array of species with considerable differences in properties. The authors clearly show their own contribution in the study of Spring Festival effect. In the present form, the authors focus on exploiting the percent change in the concentrations of various air pollutants to differentiate contribution from local and regional sources. The value of this paper could be further enhanced if the authors can make more in-depth discussion on the species (e.g., $PM_{0.8–2.5}$ and O_3), which had a small difference between the SF and NSF periods. It is interesting to know that these species revealed only a small difference when traffic flow dropped by ~50% and the industrial plants were almost entirely shut down. It is well known that PM has numerous and complex constituents contributed from diversified sources (primary and secondary, anthropogenic and natural), and O_3 is a secondary photochemical product with nonlinear relation with its precursors NOx and VOCs (anthropogenic and natural). I encourage the author to strengthen the link between their observation results and the possible causes (composition, property, sources, transport, physical and chemical processing, meteorology, etc.). In general, the manuscript was well written and organized. The subject of the paper is well within the scope of ACP. The paper is suitable for publication provided the general comments and following points are addressed.

REPLY:

We have looked for new useful evidence to explain the small decrease of SO_2 and $PM_{0.8–2.5}$ in Group SD, and have given new information as below in section 3.1:

"On the other hand, a piece of evidence for the regional origin of SO_2 is from the newly established 356 m meteorological and environmental monitoring iron tower in Shenzhen. The ambient SO_2 concentrations were similar at the highest platform (ave.=7.4 ppbv@325 m) and the lowest platform (ave.=7.2 ppbv@60 m) during January–February, 2017, indicating that SO_2 was already well mixed in the atmosphere and the local contributions should be minor. In contrast, the concentrations of NOx, which belongs to Group LD, had a 56% higher concentration at the lowest platform than at the highest platform (Zhuang, 2017). The small

decrease of SO_2 is thus a reasonable result of the stable emissions during the SF periods and the primarily regional origin."

"In terms of chemical composition of $PM_{0.8–2.5}$, implications can be found in our previous size distribution measurement of aerosol chemical composition, using a ten-stage micro orifice uniform deposit impactor (MOUDI), during the fall to winter in Shenzhen (Lan et al., 2011). The results clearly indicate that smaller fine particles (e.g., 0.18–0.56 μm) contains relatively more BC (BC/SO_4^{2-}=0.83), while larger fine particles (e.g., 1.0–1.8 μm) contained a higher proportion of SO_4^{2-} (BC/SO_4^{2-}=0.17). The SO_4^{2-} in $PM_{2.5}$ in Shenzhen has been well proved to be mostly a regional pollutant, with similar concentrations at various sites including both urban and rural sites (Huang et al., 2014). Therefore, the very small decrease of $PM_{0.8–2.5}$ during SF should be closely related to its enrichment of secondary regional species like SO_4^{2-}."

As to the unique variation of O3, it is clearly related to the nonlinear relation with its precursors NOx and VOCs, as stated by this reviewer. The other reviewer also pointed out that, based on limited VOCs measured, we cannot achieve a conclusion that whether the O3 formation is VOCs-sensitive or NOx sensitive. Therefore, in the revised manuscript, we only pointed out the idea as below in section 3.2:

"…As a result, although the reduction in emissions of urban anthropogenic sources leads to a large decline of NOx and VOCs, this reduction does not mitigate the average ambient O_3 concentration, which implies that the concentration ratio VOCs/NOx play an important role in controlling O_3 concentration."

Specific comments:
1. Page 7 Lines 137-139: 2016. Are "all fragments" of m/z 44 and m/z 57 measured by AMS are the tracers of oxygenated organic aerosol and primary hydrocarbon organic aerosol, respectively? Or "most of fragments" are?

REPLY:

We have corrected in the revised manuscript as below:
"Most of fragments of m/z 44 and m/z 57 are the tracer of oxygenated organic aerosol and the tracer of primary hydrocarbon organic aerosol (Zhang et al., 2005), respectively, which are measured by AMS."

2. Page 11 Lines 212-213: This sentence is confusing to me. In addition to emissions and sinks, the concentration of air pollutants is also dominated by meteorological conditions, especially PBL and wind field. In Table 1, meteorological conditions in NSFT and NSFM are not much different (similar wind speed and no precipitation), and it may be the major reason for the small difference in the concentrations of most air pollutants between the two periods. It is not suitable to conclude that meteorology has only a small impact on their concentrations. Please provide a more appropriate interpretation.

REPLY:

To be more rigorous, this sentence is changed to:
“The decreasing ratios of various species during SF when compared with the NSFT and NSFM periods are similar, which suggests that the meteorological variations might not be the dominant reason for the species decreasing during SF.”

3. Page 15 Lines 282-283: “emissions are greater on holidays than on non-holidays” and “In addition, emissions were higher during the 2008 Beijing Olympic Games” Do the authors mean “emissions of O_3”? If so, I suggest using the word "concentrations" instead of "emissions".

REPLY:

We have corrected in the revised manuscript.

4. Page 15 Lines 287 and 288: VOC/NOx ratio gives an important idea that it tends towards a NOx-sensitive or VOC-sensitive environment. However, VOCs are a very complex mixture of compounds with large difference in reactivity with respect to ozone formation. The true impact of VOCs to ozone formation is more relevant to the total reactivities of VOC species rather than to the total amount of VOCs. Furthermore, the VOCs reported in the study (Table S1, measured by PTR–MS) include only a total of 13 masses. TVOC/NOx ratio in the study should be carefully used to explain its effect on the ozone level due to above-mentioned concerns.

REPLY:

We agree that based on limited VOCs measurements, it is not reasonable to get the conclusions that whether the O_3 formation is NOx-sensitive or VOCs-sensitive. Therefore, in the revised manuscript, we only state as below:
“As a result, although the reduction in emissions of urban anthropogenic sources leads to a significant decline of NOx and VOCs, this reduction does not mitigate the average ambient O_3 concentration, which implies that the concentration ratio VOC/NOx plays an important role in controlling O_3 concentration.”

5. Page 15 Lines 288 and 289: Do the authors mean the chemical regime at noon during the SF period was NOx-sensitive? If so, the authors should provide a clearer explanation to support the statement.

REPLY:

Following the reply to the above question, we are not making the conclusion about whether the O_3 formation is NOx-sensitive or VOCs-sensitive.

Referee #3’s comments

General comments:

The paper presents an assessment of the effect of the Chinese Spring Festival on urban air quality in a southern China city. Various trace gases and aerosols were measured over three consecutive winters (2014-2016), including both the Spring Festival (SF) and non-Spring Festival (non-SF) periods, at an urban site of Shenzhen

city. By comparing the concentrations of these pollutants in different periods, the authors show that the decreases for some pollutants are significant (by 50%-80%) in the SF with respect to the non-SF while the decreases of others pollutants are smaller. They suggest that such differences in the reduction extent of pollutant concentrations be used to judge their sources, i.e., predominantly from local emissions or from regional transport. This study, together with a valuable dataset, should be a welcome addition to the literatures on the holiday effects on air quality. The manuscript can be accepted for publication after the following issues have been well addressed.

As the main purpose of the study is to distinguish the sources of urban pollution between the local emissions and regional transport, the local and regional pollution sources should have been clearly defined. It would be great if the authors could provide a map showing the geographical distributions of emissions rates of major air pollutants in Shenzhen city and its surrounding areas during wintertime. It is also suggested that if possible, the geographical extent of pollution reduction during the SF in relative to the non-SF period be identified, perhaps by using population density or satellite product.

The study identifies the primary source of each pollutant, i.e. the local or regional origin, using the reduction extent of a pollutant (in relative percent) from the non-SF to SF period. This methodology is fine for primary pollutants (such as NOx and BC), and it may also work for some secondary pollutants (perhaps SO_4^{2-}); but it might not entirely applicable to O_3 since ozone production changes non-linearly with an increase (or a decrease) of NOx. A stronger evidence is needed before a conclusion on the origin of ozone is given in the manuscript.

REPLY:

As to the definition of the local and regional pollution sources, please see our reply to your specific comment #14. In the revised manuscript, we have tried to define them as below:

"Apparently, the dominant sources for most of these pollutants are primarily local emissions in the urban scale, such as combustion sources for BC, m/z 57, and NOx…"; "The species in this group are either typical regional air pollutants mostly from beyond the urban scale, such as CO, which has a long lifetime…"

As to a map showing the geographical distributions of emissions rates of major air pollutants and the geographical extent of pollution reduction during the SF, they need a lot of supporting data sources, which are not easily available, and such work could be much beyond the scope of this paper. Alternatively, we have cited a paper of emission inventory in PRD (Zheng et al., 2009b) in section 2.1 to describe the relative location of the sampling site in terms of pollutant emissions in PRD, as below:

"A highly resolved temporal and spatial emission inventory for PRD indicates that the sampling area is characterized by lower SO_2 emissions but higher NOx and VOCs emissions in comparison with other areas in PRD (Zheng et al., 2009b)."

As to the conclusion on the origin of ozone, we agree on your specific comment #12 that based on limited VOCs measurements, it is not reasonable to get the conclusions that whether the O_3 formation is NOx-sensitive or VOCs-sensitive. Therefore, in the revised manuscript, we only state as below:
"As a result, although the reduction in emissions of urban anthropogenic sources leads to a significant decline of NOx and VOCs, this reduction does not mitigate the average ambient O_3 concentration, which implies that the concentration ratio VOC/NOx plays an important role in controlling O_3 concentration."

Specific comments:
1. Line 12 and Line 49-53: During the SF, the power plants are generally not shutdown in the megacities of China. Are there any power plants in Shenzhen? Where the industrial area of Shenzhen located in reference to the measurement site? Are the wind roses of various pollutants in the SF different from those in the non-SF period?
REPLY:
There are few power plants in Shenzhen. We now cite a paper (Zheng et al., 2009b) in section 2.1 to describe the relative location of the sampling site in terms of pollutant emissions in PRD, as below:
"A highly resolved temporal and spatial emission inventory for PRD indicates that the sampling area is characterized by lower SO_2 emissions but higher NOx and VOCs emissions in comparison with other areas in PRD (Zheng et al., 2009b)."
Since the data points on each wind direction are limited, wind rose analysis may not be a solid evidence. However, the analysis of relationship between pollutant concentrations and wind speeds during SF and NSF in section 3.3 can already well reveal that Group LD was highly influenced by wind speed.

2. Line 19-21: Here it might not be suitable to say "decreasing of regional pollutants" since there is an increase of O_3 by 6%.
REPLY:
We rephrased the sentences by "The concentration variation of species mostly from regional or natural sources, however, is found to be much less, such as for bulk $PM_{2.5}$."

3. Line 47: It might not be fully suitable to say so. The reported emission reductions could be verified by comparisons of different approaches, e.g., ground measurements, satellite observations, and model simulations with different emission inventories.
REPLY:
"…so the reported emission reductions cannot be verified" changed to "…so the air quality monitoring campaigns cannot be repeated."

4. Line 77-79: Figure S1 provides only geographical position of measurement site. How far away from the main traffic roads the site is? How about regional distributions of pollutants' emissions?

REPLY:

We have provided the relevant information in the revised text as below:

"PKUSZ is located in the western urban area of Shenzhen, and there are no significant anthropogenic pollution sources nearby except a local road ~100 m far from the sampling site. A highly resolved temporal and spatial emission inventory for PRD indicates that the sampling area is characterized by lower SO_2 emissions but higher NOx and VOCs emissions in comparison with other areas in PRD (Zheng et al., 2009b)."

5. Line 135: Figures S3-S5 provide more detailed information and can be moved into the formal part of the manuscript.

REPLY:

After careful consideration, we still believe that Figures S3-S5 are too large to be moved into the main text. Therefore, supporting information could be a better choice when considering that ACP is freely available on the internet.

6. L174: CO is NOT a typical SECONDARY regional air pollutant as most of CO in the urban and polluted areas are mostly probably to be primarily emitted.

REPLY:

It is a typo. "secondary" is now deleted.

7. L183-184, L252-253, L261-267, L309-312 and L337-340: According to the study, isoprene and DMS measured in Shenzhen have anthropogenic sources. But they are repeatly described as "natural" gases; on the other hand, sometimes they are classified as "pollutants". These vague expressions should be corrected.

REPLY:

We have checked all the manuscript and rephrased the words to say that they are "mainly emitted by natural sources"

8. Line 191-192: It should noted that there is an increase of O_3 to a small extent.

REPLY:

We have rephrased the sentences as below:

"The group of pollutants with smallest decrease in concentration (hereinafter called "SD") includes SO_2, $PM_{0.8–2.5}$, and O_3 (8h) in the case of comparison with NSFM. The magnitude of the average percent change is less than 20% relative to the two NSF periods. It is interesting to note that there was even concentration increase in other O_3–related cases."

9. L195-202: Are there any power plants in Shenzhen and nearby areas? A plot of the regional emission distributions of SO_2 would be helpful for the reader to follow the discussion here. It would also be great if the wind rose or trajectory analysis result could be given.

REPLY:

There are few power plants in Shenzhen. We now cite a paper (Zheng et al., 2009b) in section 2.1 to describe the relative location of the sampling site in terms of pollutant emissions in PRD, as below:
"A highly resolved temporal and spatial emission inventory for PRD indicates that the sampling area is characterized by lower SO_2 emissions but higher NOx and VOCs emissions in comparison with other areas in PRD (Zheng et al., 2009b)."
Since the data points on each wind direction are limited, wind rose analysis may not be a solid evidence. However, the analysis of relationship between pollutant concentrations and wind speeds during SF and NSF in section 3.3 can already well reveal that Group LD was highly influenced by wind speed.

10. L212-215 and L300-301: The sentence "meteorology has only a small impact on their concentrations" is misleading. Note that only for the period average values are the result consistent.

REPLY:

To be more rigorous, this sentence is changed to:
"The decreasing ratios of various species during SF when compared with the NSFT and NSFM periods are similar, which suggests that the meteorological variations might not be the dominant reason for the species decreasing during SF."

11. L231, Figure 2: Ticks for 10 nm and 1000 nm could be given.

REPLY:

Our instrument (SMPS) determined the particle number size distribution in the size range of 15–615 nm.

12. L288-289: In-depth analysis should be performed if you insist that the lack of NOx at noon during the SF period hinders the generation of O_3. Figure 3B shows that the levels of NOx in the SF and NSFM at noon are comparable. It seems that the difference in VOCs might play an important role.

REPLY:

We agree that based on limited VOCs measurements, it is not reasonable to get the conclusions that whether the O_3 formation is NOx-sensitive or VOCs-sensitive. Therefore, in the revised manuscript, we only state as below:
"As a result, although the reduction in emissions of urban anthropogenic sources leads to a significant decline of NOx and VOCs, this reduction does not mitigate the average ambient O_3 concentration, which implies that the concentration ratio VOC/NOx plays an important role in controlling O_3 concentration."

13. L300: What are the wind fields look like? It might be more appropriate to say the wind field patterns are the same.

REPLY:

The wind rose plots can be seen in Figure S2. We take this suggestion to use "wind field patterns"

14. L368-370: The concept of regional air pollutants is unclear. How are they defined? Even for NOx, it can also result in regional pollution.

REPLY:

In this study, we studied urban air quality and urban emissions, and thus regional pollutants refer to species from outside the urban areas. Yes, NOx can also be from regional transport. However, based on our results, NOx is found to be mostly from local urban emissions. In the revised text, we have added the words like “air pollutants mostly from regional or natural sources” to be more rigorous.

Differentiating local and regional sources of Chinese urban air pollution based on effect of Spring Festival

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Abstract: The emission of pollutants is extremely reduced during the annual Chinese Spring Festival (SF) in Shenzhen, China. During the SF, traffic flow drops by ~50% and the industrial plants are almost entirely shut down in Shenzhen. To characterize the variation in ambient air pollutants due to the "Spring Festival effect", various gaseous and particulate pollutants were measured in real time in urban Shenzhen over three consecutive winters (2014–2016). The results indicate that the concentrations of NOx, volatile organic compounds (VOCs), black carbon (BC), primary organic aerosols, chloride, and nitrate in submicron aerosols decrease by 50%–80% during the SF period relative to the non-Spring Festival periods, regardless of meteorological conditions, which suggests that these pollutants are mostly emitted or secondarily formed from urban local emissions. The concentration variation of species mostly from regional or natural sources, however, is found to be much less, such as for bulk $PM_{2.5}$. More detailed analysis of the Spring Festival effect reveals an urgent need to reduce emissions of SO_2 and VOCs on a regional scale rather than on an urban scale to reduce urban $PM_{2.5}$ in Shenzhen, which can also produce some use for reference for other megacities in China.

Key words: Spring Festival effect; local emissions; regional pollution; $PM_{2.5}$; ozone

1 Introduction

The rapid economic development and urbanization of China over the recent decades has brought with it the consequence of severe atmospheric pollution, especially in the key economically developed regions, such as the Beijing–Tianjin–Hebei region (Sun et al., 2013, 2015; Guo et al., 2014), the Yangtze River Delta (Huang et al., 2013), and the Pearl River Delta (PRD), as well as their densely populated megacities (Hagler et al., 2006; Zhang et al., 2008; He et al., 2011). Great efforts have been made to determine the sources and formation mechanisms of fine particles ($PM_{2.5}$) in these region. Previous studies indicate that $PM_{2.5}$ forms from primary fine particles and through secondary formation from gaseous precursors (Zhang et al., 2008; Zheng et al., 2009a; Huang et al., 2014), and the sources of local production and regional transport are both important (Huang et al., 2014; Huang et al., 2006, 2011; Li et al., 2015).

The causes of air pollution in urban atmosphere in China are particularly complicated, and bring great challenges to management strategies for protecting human health (Parrish and Zhu, 2009). To explore the causes of urban air pollution in China, previous studies have focused on monitoring and comparing the reduction in emissions during special events, such as the 2008 Beijing Olympic Games (Huang et al., 2010), the 2010 Guangzhou Asian Games (Xu et al., 2013), the 2014 Asia Pacific Economic Cooperation conference (APEC) (Chen et al., 2015; Sun et al., 2016; Zhang et al., 2016) and the 2015 China victory day parade (Zhao et al., 2016). During such events, the air quality improved remarkable because of short-term limitations on traffic and industrial activity (Huang et al., 2010; Wang et al., 2010; Xu et al., 2013; Sun et al., 2016; Zhao et al., 2016). However, these limitations were temporary, non-repeatable measures, so the air quality monitoring campaigns cannot be repeated. Actually, a

spontaneous reduction in emissions occurs every year in China during the Spring Festival (SF), which is the single most important holiday in China. During the week-long holiday (in January or February every year), the urban emission patterns depart significantly from the usual patterns: traffic decreases in the mega cities because most people are not working, and most of the industries, stores, and production sites are closed in the city except for the infrastructure (e.g., power plants) that cannot be shut down (Qin et al., 2004; Feng et al., 2012; Shi et al., 2014). Tan et al. (2009) reported that the concentrations of NOx, CO, NMHC, SO_2, and PM_{10} were lower in the SF periods than in the non-Spring Festival (NSF) periods in the metropolitan area of Taipei over 1994-2006, while the variation of O_3 was in a reversed trend. Jiang et al. (2015) found that the ambient concentrations of VOCs had a sharp decline by ~60% during the SF in Shijiazhuang.

This study focuses on Shenzhen as a special example to evaluate the effect on urban air pollution of the SF. Shenzhen is in the eastern Pearl River Delta (PRD) and is the fourth largest economic center in China, with a total residential population of over 10 million and a fleet of civilian vehicles of more than 3.1 million (Shenzhen Yearbook of Statistics, 2015). Known as the country's city of most floating population, Shenzhen owns 7.4 million immigrants in 2014, which accounts 70% of the city's total population (Shenzhen Yearbook of Statistics, 2015). During the SF period, over 50% of the residents in Shenzhen are used to travel back to their hometowns (http://sz.gov.cn). It is reported that the traffic flow in Shenzhen during the SF of 2016 (Feb 7–13) was only the half before the SF period (http://sz.gov.cn). Additionally, industrial activities are almost totally suspended in Shenzhen during the SF period. To characterize the air quality during such extreme reductions of anthropogenic activities during the SF period in Shenzhen, various air pollutants in Shenzhen urban areas were

comprehensively and systematically monitored in real time in winter for three consecutive years (2014–2016). The annual SF in Shenzhen thus provides an excellent spontaneous control experiment for local emissions, which could provide unique and valuable information regarding the sources of urban air pollution.

2 Experimental methods

2.1 Monitoring sites and meteorological conditions

The monitoring site (22°36′N, 113°54′E) was on the roof (20 m above ground level) of an academic building on the campus of Peking University Shenzhen Graduate School (PKUSZ) (Figure S1). PKUSZ is located in the western urban area of Shenzhen, and there are no significant anthropogenic pollution sources nearby except a local road ~100 m far from the sampling site. A highly resolved temporal and spatial emission inventory for PRD indicates that the sampling area is characterized by lower SO_2 emissions but higher NOx and VOCs emissions in comparison with other areas in PRD (Zheng et al., 2009b). The sampling schedule ran roughly from late January to early March over 2014–2016, which includes the official SF holiday period and the prior and following periods. Our definition of the SF period follows that of the statutory public holiday calendar in China, and it is continuous seven days in each year. While the seven days immediately before or after the holidays are actually the transition periods between the holidays and normal days (called the Tran. periods hereafter), when people begin to move from the city (or their hometowns) to their hometowns (or the city), the typical non-spring festival (NSF) periods are better defined as the 7–14 days close to the SF period (called the NSFT period hereafter, where T indicates time similar). The specific dates and the average meteorological parameters are listed in Table 1, and Figure S2 shows wind rose plots. The data in Table

1 show that the meteorology differs among the SF, NSFT, and Tran. periods. To control for the influence of meteorology on the evaluation of emissions, we selected another 7-day period each year when the meteorology is similar to that of the SF period (called the NSFM period hereafter, where M indicates meteorology similar); the detailed parameters are listed in Table 1 and Figure S2. The meteorological data for the SF period are fairly similar to those of the NSFM period, suggesting similar meteorological conditions.

Table 1. Summary of meteorological conditions at sampling site during the SF, NSFT, NSFM and Tran. periods of 2014–2016.

		SF	Tran.	NSFT	NSFM
Data period	2014	Jan 31–Feb 6	Feb 7–Feb 13	Feb 14–Feb 20	Feb 20–Feb 26
	2015	Feb 18–Feb 24	Feb 11–Feb17	Feb 4–Feb10	Jan 24–Jan 30
	2016	Feb 7–Feb 13	Feb 14–Feb 20	Feb 21–Feb 27	Feb 27–Mar 4
Meteorological parameters	Temperature (℃)	19.0±4.7	14.1±5.3	14.1±4.0	18.1±3.8
	RH (%)	68.1 ±17.8	69.3±18.4	64.9±16.7	67.4±14.7
	Wind speed ($m\ s^{-1}$)	0.88 ±0.57	0.81±0.49	0.83±0.48	0.86±0.55
	Dominant wind direction	NW	NW and NE	NW and NE	NW
	Precipitation (mm)	0	0	0	0
	UVA ($W\ m^{-2}$)	5.4±8.5	2.5±4.3	3.8±6.7	5.0±8.0
	UVB ($W\ m^{-2}$)	0.24±0.40	0.11±0.25	0.16±0.32	0.22±0.38

2.2 Instrumentation

For the ambient sampling in this study, the measuring instruments were placed in a room on the top floor of a four-story teaching building at PKUSZ. A high-sensitivity proton transfer reaction mass spectrometer (PTR–MS) (Ionicon Analytik GmbH, Austria) was used to measure the selected volatile organic compounds (VOCs). The PTR–MS measured a total of 25 masses in the selected ion mode at

a time resolution of 30 s. Background checks were done for 30 of every 300 scan cycles with an activated charcoal trap at 360 °C, which can remove VOCs from the ambient air without changing water content. The VOCs reported here (Table S1) may be broadly classified into three categories: oxygenated VOCs [OVOCs: methanol, acetone, methyl ethyl ketone (MEK), acetaldehyde, and acetic acid], aromatics (benzene, toluene, styrene, C8 and C9 aromatics), and three types of tracers [isoprene, acetonitrile, and dimethyl sulfide (DMS)]. The PTR–MS was calibrated every 5 to 7 days by using a TO15 mixture standard (Air Environmental Inc., US) and permeation tubes (Valco Instruments Co. Inc., US) (de Gouw and Warneke, 2007).

An aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR–ToF–AMS) (Aerodyne Research, US) was deployed to measure non-refractory PM_1 (NR-PM_1) (Canagaratna et al., 2007) in the period 2014–2015 with a time resolution of 4 min. An aerosol chemical speciation monitor (ACSM) (Aerodyne Research, US) was used in 2016 with a dynamic resolution of 10 min. The detailed description of the ACSM is available in the recent review (Ng et al., 2011). The HR-ToF-AMS and ACSM were calibrated every month following the standard protocols (Ng et al., 2011; Jayne et al., 2000).

An aethalometer (AE–31) (Magee, US) was used for simultaneous detection of refractory black carbon (BC) with a time resolution of 5 min. In addition, a Scan Mobility Particle Sizer (TSI Inc., US) system was used to determine the particle number size distribution in the size range 15–615 nm (Stokes diameter) with a time resolution of 5 min. The stokes diameters of 15–615 nm is converted to aerodynamic diameters of 22–800 nm, and then $PM_{0.8}$ mass concentration can be calculated with the

142 particle density assumed according to the AMS measurement results of species.

143

144 To measure the $PM_{2.5}$ mass concentration, we used a Thermo Scientific TEOM 1405–D monitor. The
145 trace-gas instruments included a 43i sulfur dioxide (SO_2) analyzer, a 42i nitric oxide (NO)–nitrogen
146 dioxide (NO_2)–nitrogen oxide (NOx) analyzer, a 49i ozone (O_3) analyzer, and a 48i carbon monoxide
147 (CO) analyzer (Thermo Scientific, US). A meteorological station, also located on the roof of the same
148 building, measured the main meteorological parameters, such as temperature, relative humidity, and
149 wind speed (see Table 1).

150

151 **3 Results and Discussion**

152 **3.1 The NSF–SF differences for major air pollutants**

153 The results of observations from 2014 to 2016 appear in Figures S3–S5. Figure 1 shows the averaged
154 percent changes in the concentrations of major air pollutants of the SF periods relative to the two NSF
155 periods and Tran. period over 2014–2016. Most of fragments of m/z 44 and m/z 57 are the tracer of
156 oxygenated organic aerosol and the tracer of primary hydrocarbon organic aerosol (Zhang et al., 2005),
157 respectively, which are measured by AMS. The notation O_3–8h refers to the average maximum O_3
158 concentration over a continuous diurnal 8 h and $PM_{0.8–2.5}$ refers to the difference between the
159 concentrations of $PM_{2.5}$ and $PM_{0.8}$.

160

161 We can divide these air pollutants into three classes based on their percent changes: The group with
162 the largest drop (hereinafter called "LD") in concentration includes the aromatics (−50% to −88% for
163 the various species, see Figure S6), OVOCs (−40% to −85% for the various species, see Figure S6),

NOx, chloride (Chl), nitrate (NO_3^-), BC, and m/z 57. The concentrations of these pollutants all decrease by over 50% during the SF period compared with both the NSF periods. Apparently, the dominant sources for most of these pollutants are primarily local emissions in the urban scale, such as combustion sources for BC, m/z 57, and NOx (Zhang et al., 2005; Kuhlbusch et al., 1998; Lan et al., 2011), and vehicle, industrial and solvent use for aromatics (Liu et al., 2008). As detailed in the following section, the diurnal patterns and relationships with respect to wind speed further confirm the sources of these pollutants. The dramatic decrease in the ambient concentrations of these species is consistent with reduction in local anthropogenic activities in Shenzhen during the SF period. The SF causes a 50% decrease in urban traffic and temporarily closing of almost all local industrial plants. The nitrate and chloride measured by AMS or ACSM are actually ammonium nitrate (NH_4NO_3) and ammonium chloride (NH_4Cl), which are typical secondary air pollutants. These are thought to form via reversible phase equilibria with gaseous ammonia (NH_3), nitric acid (HNO_3), and hydrochloric acid (HCl) (He et al., 2011; Huang et al., 2011; Zhang et al., 2007). Typically, the formation of NH_4NO_3 from NOx and the reaction between HCl and NH_3 occur quickly in the atmosphere (Stelson and Seinfeld, 1982; Baek et al., 2004), suggesting that the concentrations of NO_3^- and Chl in winter in Shenzhen depend largely on the emission of precursors such as HCl and NOx. Therefore, the significant decline in the ambient concentrations of NO_3^- and Chl during the SF period and indicates that their precursors also have local origins, similar to the case for primary pollutants (this is also supported by the discussion in the following sections). The huge decline in the ambient concentration of OVOCs during the SF period shows that the source of these pollutants is (i) mainly from local emissions, including vehicle and industrial emissions (Schauer et al., 1999; Singh et al., 2001) and (ii) from secondary reactions involving local primary VOCs (Liu et al., 2015). Thus, in the LD group, the

186 significant reduction in local sources of pollutants strongly impacts the concentration of air pollutants.

187

188 The pollutants in the next group undergo a medium drop in concentration during the SF period
189 (hereinafter called "MD"). These are $PM_{2.5}$, NR–PM_1, $PM_{0.8}$, organic aerosol, m/z 44, sulfate (SO_4^{2-}),
190 ammonium (NH_4^+), isoprene, acetonitrile, DMS, and carbon monoxide (CO), and their percent change
191 varies from −20% to −55% when comparing the SF periods to the NSFT and NSFM periods. The
192 species in this group are either typical regional air pollutants mostly from beyond the urban scale, such
193 as CO, which has a long lifetime and is a tracer for combustion sources, acetonitrile from rural biomass
194 burning (de Gouw et al., 2003; Le Breton et al., 2013), m/z 44 representing secondary organic aerosols,
195 SO_4^{2-} from SO_2 oxidation (He et al., 2011; Huang et al., 2011), or typical tracers mainly emitted by
196 natural sources, such as isoprene from vegetation (Guenther et al., 1995) and DMS from marine source
197 (Dacey and Wakeham, 1986). In winter, the northeastern monsoon prevails in the PRD and transports
198 significant amounts of various air pollutants from the northern inland, increasing air pollution of the
199 PRD to the highest levels through the year (Huang et al., 2014). In particular, the small drop in CO
200 concentration during the SF period puts it in this group and indicates that the contribution to regional
201 air pollution does not decrease significantly during the SF period. Note that, the significant declines of
202 the concentrations of isoprene and DMS imply that they have anthropogenic sources, which will be
203 supported in the following sections. The other air pollutants in this group are the reflection of the
204 overall effect of the reduction of relevant air pollutants: OA is the whole of the two types of organic
205 aerosol represented by m/z 44 and m/z 57, NH_4^+ is represented by SO_4^{2-}, NO_3^- and Chl, and NR–PM_1
206 is the sum of all species measured by AMS or ACSM (their average chemical compositions during
207 different periods are shown in Figure S3–S5).

The group of pollutants with smallest decrease in concentration (hereinafter called "SD") includes SO_2 and $PM_{0.8–2.5}$, and O_3 (8h) in the case of comparison with NSFM. The magnitude of the average percent change is less than 20% relative to the two NSF periods. It is interesting to note that there was even concentration increase in other O_3–related cases. The average concentration of SO_2 was only 2.8 ppbv in Shenzhen in 2015 (http://www.szhec.gov.cn/), which is much lower than that in Beijing (4.7 ppbv) and elsewhere in China (http://www.zhb.gov.cn/). This result is partly attributed to the negligible coal consumption in Shenzhen, which instead relies mainly on natural gas and liquefied petroleum gas (Shenzhen Yearbook of Statistics, 2015). The emission inventory indicates that power plants and international marine container vessels are the dominant source of SO_2 in Shenzhen (Wang et al., 2009; Zheng et al., 2009b). According to official statistics, the Shenzhen port piloted 401, 568, and 521 ships during the SF period in the years 2014–2016, respectively, which is quite similar to numbers for the NSF periods (http://www.pilot.com.cn). As infrastructure, power plants are not fully shut down during SF. On the other hand, a piece of evidence for the regional origin of SO_2 is from the newly established 356 m meteorological and environmental monitoring iron tower in Shenzhen. The ambient SO_2 concentrations were similar at the highest platform (ave.=7.4 ppbv@325 m) and the lowest platform (ave.=7.2 ppbv@60 m) during January–February, 2017, indicating that SO_2 was already well mixed in the atmosphere and the local contributions should be minor. In contrast, the concentrations of NOx, which belongs to Group LD, had a 56% higher concentration at the lowest platform than at the highest platform (Zhuang, 2017). The small decrease of SO_2 is thus a reasonable result of the stable emissions during the SF periods and the primarily regional origin. The small decline of $PM_{0.8–2.5}$ during the SF period suggests that the reduction of more aged particles of lager sizes in $PM_{2.5}$ is much lower than fresher particles of smaller size. This can be also confirmed by particle number concentration (PNC)

measurement by SMPS, as shown in Figure 2. The largest difference of the PNC between the SF and NSFM periods exists mainly in a smaller size range (20–40 nm), which is recognized as the nucleation mode or second Aitken mode that represents fresh combustion emission (Ferin et al., 1990). In terms of chemical composition of $PM_{0.8–2.5}$, implications can be found in our previous size distribution measurement of aerosol chemical composition, using a ten-stage micro orifice uniform deposit impactor (MOUDI), during the fall to winter in Shenzhen (Lan et al., 2011). The results clearly indicate that smaller fine particles (e.g., 0.18–0.56 μm) contains relatively more BC (BC/SO_4^{2-}=0.83), while larger fine particles (e.g., 1.0–1.8 μm) contained a higher proportion of SO_4^{2-} (BC/SO_4^{2-}=0.17). The SO_4^{2-} in $PM_{2.5}$ in Shenzhen has been well proved to be mostly a regional pollutant, with similar concentrations at various sites including both urban and rural sites (Huang et al., 2014). Therefore, the very small decrease of $PM_{0.8–2.5}$ during SF should be closely related to its enrichment of secondary regional species like SO_4^{2-}. Contrary to other pollutants, the concentrations of O_3, present small increasing during the SF period (except a little decline when comparing O_3–8h with the NSFM period), which could be attributed to the different drop rates for O_3 precursor species, i.e. NOx and VOCs (Qin et al., 2004), and will be discussed in more detail in section 3.2.

The decreasing ratios of various species during SF when compared with the NSFT and NSFM periods are similar, which suggests that the meteorological variations might not be the dominant reason for the species decreasing during SF. This means that the strong decrease in the concentrations of air pollutants in group LD and MD is mainly due to the abatement of local sources. The larger decline in the SF period when compared to NSFT than to NSFM is associated with the lower temperature and stronger winds from the polluted northwest inland of the PRD during the NSFT period. In addition, the effect

of the SF on the concentrations of the various species is almost identical each year (see Figure S7), which further confirms that the pollutant concentrations are determined primarily by the activity of the sources. In Figure 1, the percent changes of pollutants of the SF periods relative to the Tran. periods are also presented, and it is found that the three-group classification defined above is also applicable, while the decrease levels are lower. For example, the average decrease percent of Group LD for the Tran. period case is 61%, while those for the NSFT and NSFM cases are 71% and 63%, respectively. This result is consistent with the fact that the SF travel of people occurred mostly during the seven days before and after the SF holidays (http://sz.gov.cn), and thus the city became much emptier even in the Tran. periods. In order to make a deeper and valid comparison for revealing the SF effect, the following discussion will only take the NSFM periods and SF periods for comparative analysis due to their more similar meteorology.

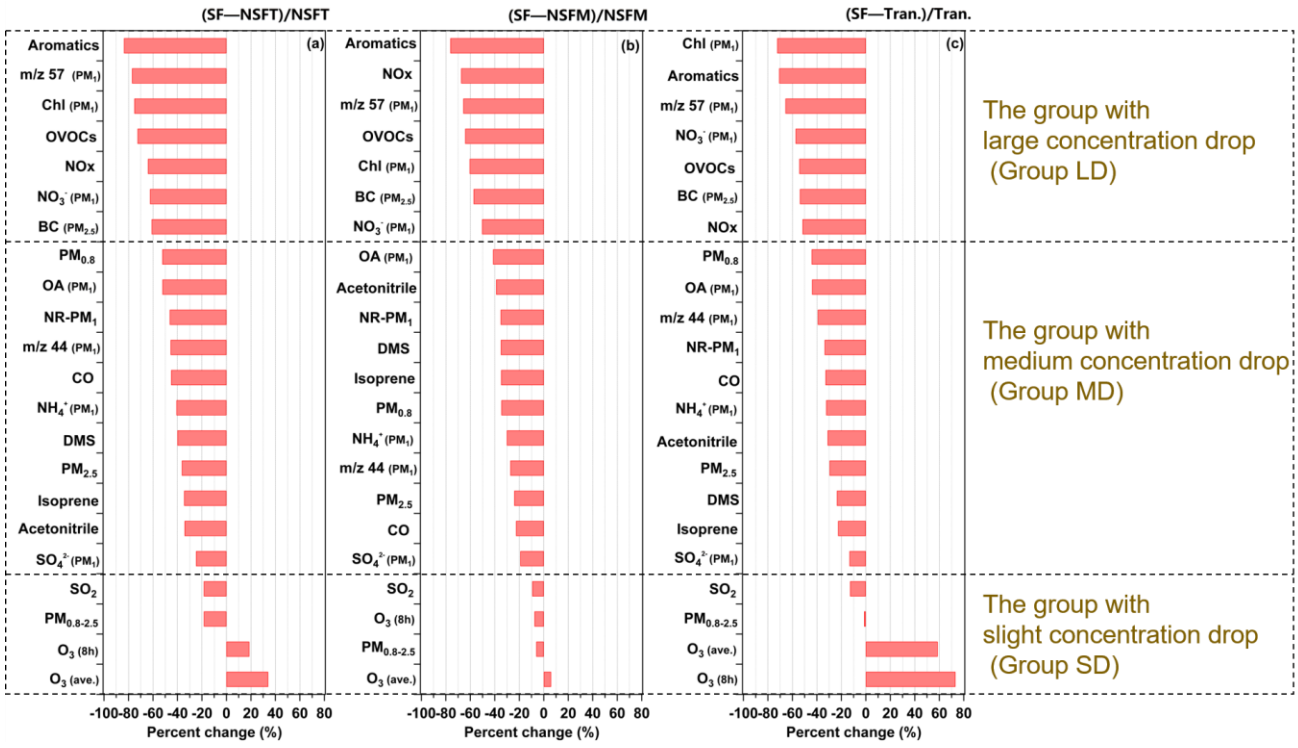
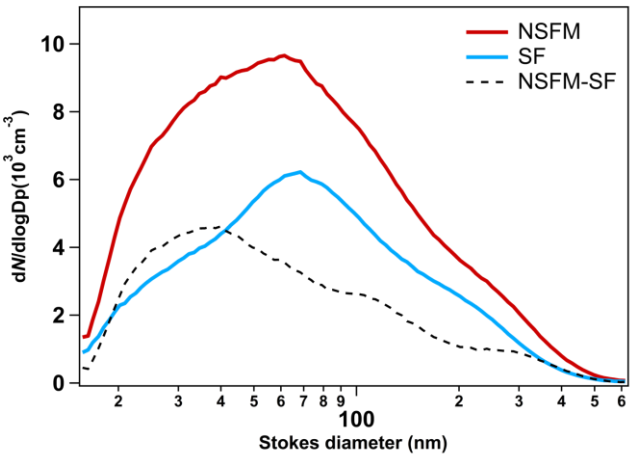


Figure 1. Percent change in concentrations of major air pollutants during the SF period relative to (a)

265 Tran., (b) NSFT and (c) NSFM periods averaged over 2014–2016.



266

267 **Figure 2.** Distribution of particle number concentration in the 15–615 nm size range during the SF

268 and NSFM periods.

269

270 **3.2 The diurnal variation of major air pollutants**

271 As shown in Figure 3, the diurnal cycles of all LD pollutants (except for the OVOCs) reveal significant

272 peaks in concentration around 8–9 am in the NSFM period, which is attributed to the low planetary

273 boundary layer (PBL) in the morning and local rush hour traffic emissions. The evening rush hour

274 peak, however, is not apparent for all the species, which is attributed to the higher ambient temperature

275 and thus the higher PBL at that time than in the morning. During the SF period, the concentrations of

276 all pollutants are far lower over the entire day. In particular, the rush-hour peaks become much smaller

277 or disappear altogether, which is consistent with the large reduction in local vehicle emissions during

278 the SF period. Although the sources of Chl remained uncertain in previous studies (Huang et al., 2011;

279 Aiken et al., 2008), the maximal reduction (80%) in this pollutant during the morning rush hour during

280 the SF period implies that local traffic emissions account for a significantly fraction of this pollutant

281 in Shenzhen (Figure 3E). Contrary to other species in this group, the concentration of OVOCs is high
282 in the daytime and peaks in the morning after the morning rush hour time during the NSFM period
283 (Figure 3D), suggesting that photochemical production and/or daytime industrial activities may be
284 important sources of OVOCs. The concentrations of different aromatics and OVOCs usually follow
285 similar diurnal variations (Figure S8).
286
287 The diurnal variations of the MD pollutants are relatively smooth except for two VOCs mainly from
288 natural sources (isoprene and DMS; see Figures 3L and 3M), which indicates that these pollutants
289 predominantly come from regional sources and are dispersed more uniformly over a larger scale. The
290 apparent difference of the diurnal variations of those anthropogenic air pollutants between the SF and
291 NSFM periods also exists in the rush hours (except for acetonitrile; Figure 3J), however, the reduction
292 in local sources has a relatively weak effect on the overall concentrations of these pollutants.
293 Acetonitrile, which is a tracer of biomass burning, is more concentrated during the daytime and its
294 peak concentration occurs after the rush hours during the NSFM period (Figure 3J), which is similar
295 to the result obtained for OVOCs and may be attributed to the influence of daytime anthropogenic
296 activities, for example, industrial biomass boilers. Isoprene is primarily emitted by vegetation as a
297 function of light and temperature, so the concentration of this pollutant goes through a broad peak that
298 spans the daytime hours during both the NSFM and SF periods. The percent change in isoprene
299 concentration between the SF and NSFM periods is approximately −40% (Figure 3M), despite the
300 NSFM and SF periods having similar temperature and solar radiation, which implies that the
301 contribution of anthropogenic sources to isoprene cannot be overlooked in Shenzhen. Many studies
302 have reported isoprene from vehicle exhaust, especially in cold seasons (Barletta et al., 2005; Borbon

et al., 2001). DMS is reported to be a marine tracer (Dacey et al., 1986), its peak concentration occurs in the morning during both the NSFM and SF periods (Figure 3L), which is presumably related to the minimal PBL. The concentration of DMS decreases by 30%–50% during the SF period, which reflects the reduced DMS emissions from anthropogenic sources. As reported in the literature, industrial activities can make significant emissions of DMS (Schafer et al., 2010).

The diurnal variations of $PM_{0.8–2.5}$, SO_2 and O_3 demonstrated more similar concentrations and trends in the SF and NSFM periods, respectively (Figure 3T–3V). For $PM_{0.8–2.5}$, a small difference is found in the afternoon, which is supposed to be a result of more aged larger particles formed through stronger photochemical reactions during the NSFM period. Though, slight differences appear in SO_2 concentration, mainly during the nighttime when the PBL is low. These data suggest a minor role of local near-ground SO_2 sources, such as vehicles. Although the daytime peak concentration of O_3 during the NSFM period is slightly greater than that during the SF period, this trend reverses from the evening to the midmorning hours. Similar phenomena have also been observed in other emission-reduction studies of urban areas (i.e., O_3 concentrations are greater on holidays than on non-holidays) (Qin et al., 2004; Tan et al., 2009). In addition, O_3 concentrations were higher during the 2008 Beijing Olympic Games (Chou et al., 2011), during which strict controls were imposed. The lower peak concentration of O_3 in the afternoon (13:00-16:00 LT) during SF suggests that the large reduction on precursors can also help mitigate the daytime O_3 concentration. However, the O_3 concentration at night during SF was higher than that during NSFM, which could be attributed to the oxidation reaction with NO of higher concentrations during NSFM, producing a titration effect and thus destroying O_3 (Qin et al., 2004; Tan et al., 2009). As a result, although the reduction in emissions of urban anthropogenic sources

leads to a large decline of NOx and VOCs, this reduction does not mitigate the average ambient O_3 concentration, which implies that the concentration ratio VOCs/NOx play an important role in controlling O_3 concentration.

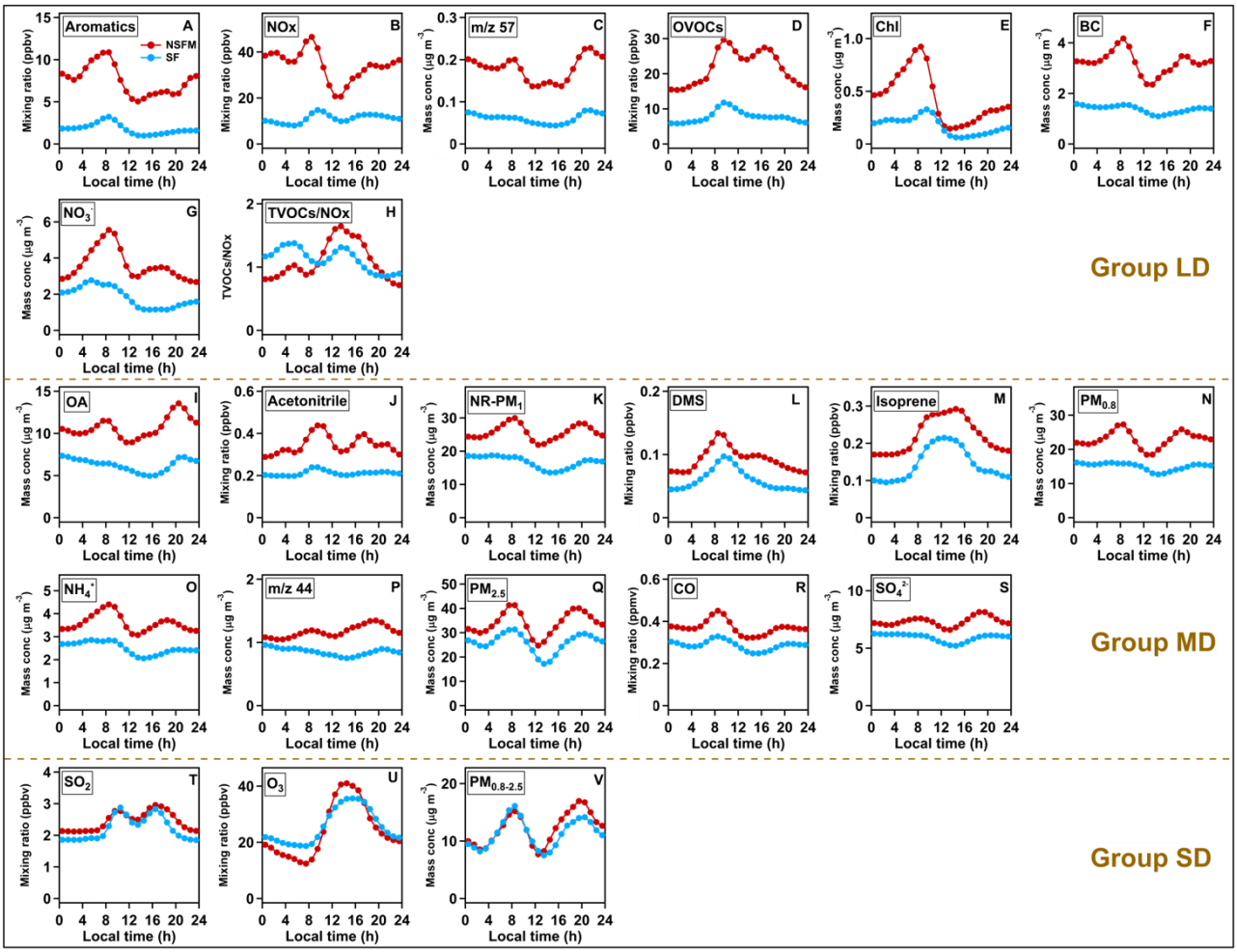


Figure 3. Diurnal variations in concentrations of major air pollutants at PKUSZ site over the SF (blue dots) and NSFM (red dots) periods.

3.3 Influence of wind on observed air pollutants

Wind plays a crucial role in the dilution and transport of air pollution. The wind field patterns are quite similar between the SF and NSFM periods (Figure S2). In general, the concentrations of LD air

pollutants depend strongly on wind speed during the NSFM period, whereas this dependence becomes much weaker during the SF period (Figure 4). The difference in the concentration of LD air pollutants (including various aromatics and OVOCs, see Figure S9) between the NSFM and SF periods is maximal (50%–80%) under conditions of low wind speeds (<1 m/s) because local pollution can more easily accumulate under these conditions. These results confirm that the concentration of air pollutants mainly from local sources is strongly reduced during in the SF period.

Compared with the LD pollutants, the concentrations of CO, SO_4^{2-}, m/z 44, isoprene, DMS, and acetonitrile do not vary significantly with wind speed during the NSFM period, providing further evidence that these pollutants primarily come from regional or natural sources and are consequently more evenly distributed in the atmosphere.

In the Group SD, SO_2 is generally little influenced by wind speed during the SF period, while some higher concentrations appeared under low wind speeds during the NSFM period, indicating again small contribution of urban local sources to SO_2. The fluctuation of $PM_{0.8–2.5}$ both in the SF and NSFM periods does not reveal a clear relationship with wind speed, suggesting again it is not a typical locally emitted air pollutant. The variations of O_3–8h display the opposite trend to other air pollutants both in the SF and NSFM periods, growing smoothly as wind speed increases, which could be possibly attributed to more regional transport and/or the higher VOCs/NOx ratio under high wind speeds (Figure 4H). Note that, when the proportion of regional transport relative to local emission becomes bigger under higher wind speeds, the concentrations of NO_3^-, SO_4^{2-}, m/z 44, $PM_{0.8–2.5}$, and O_3–8h are even slightly higher in the SF period than in the NSFM period, implying that regional photochemical

production during the SF period is not weakened.

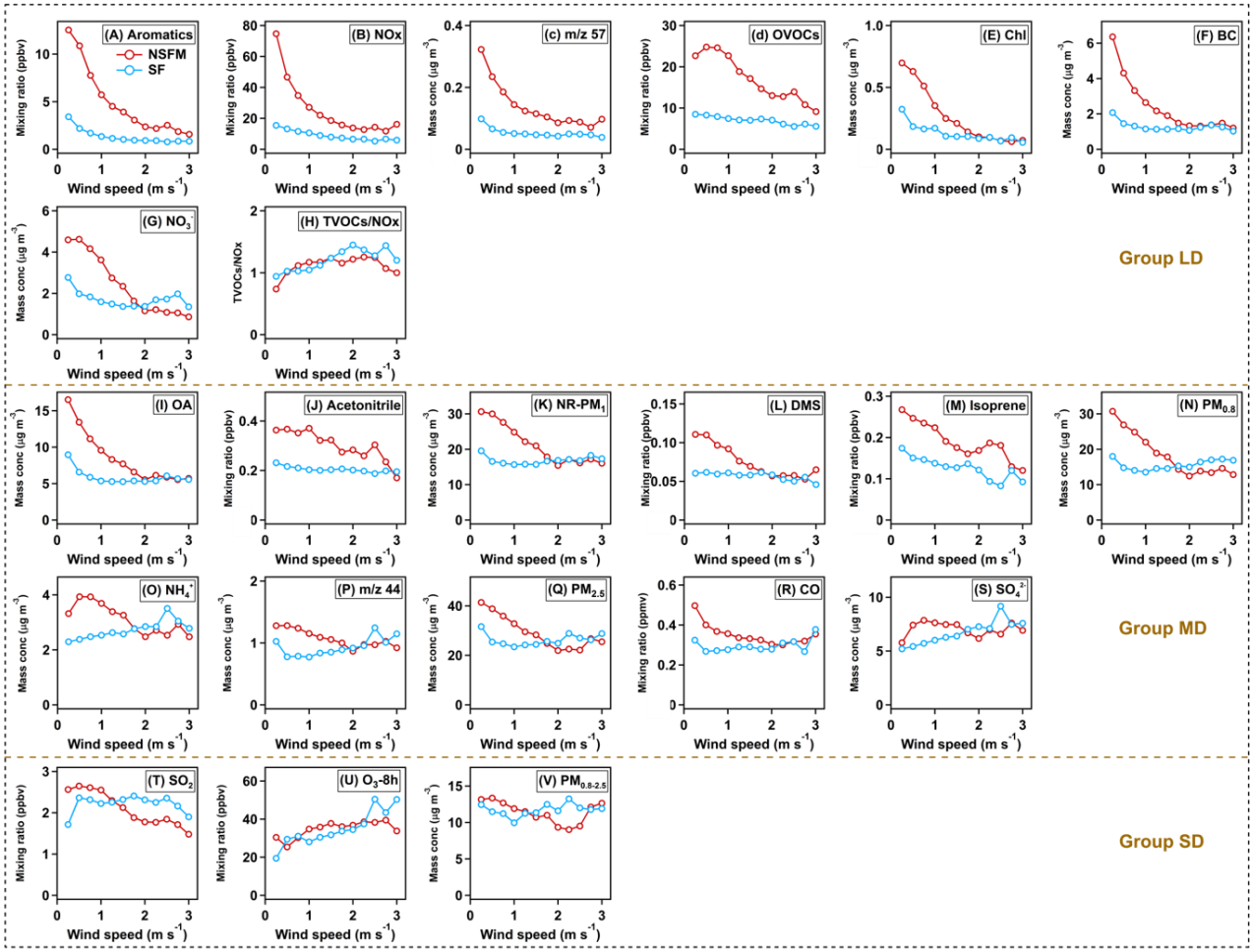


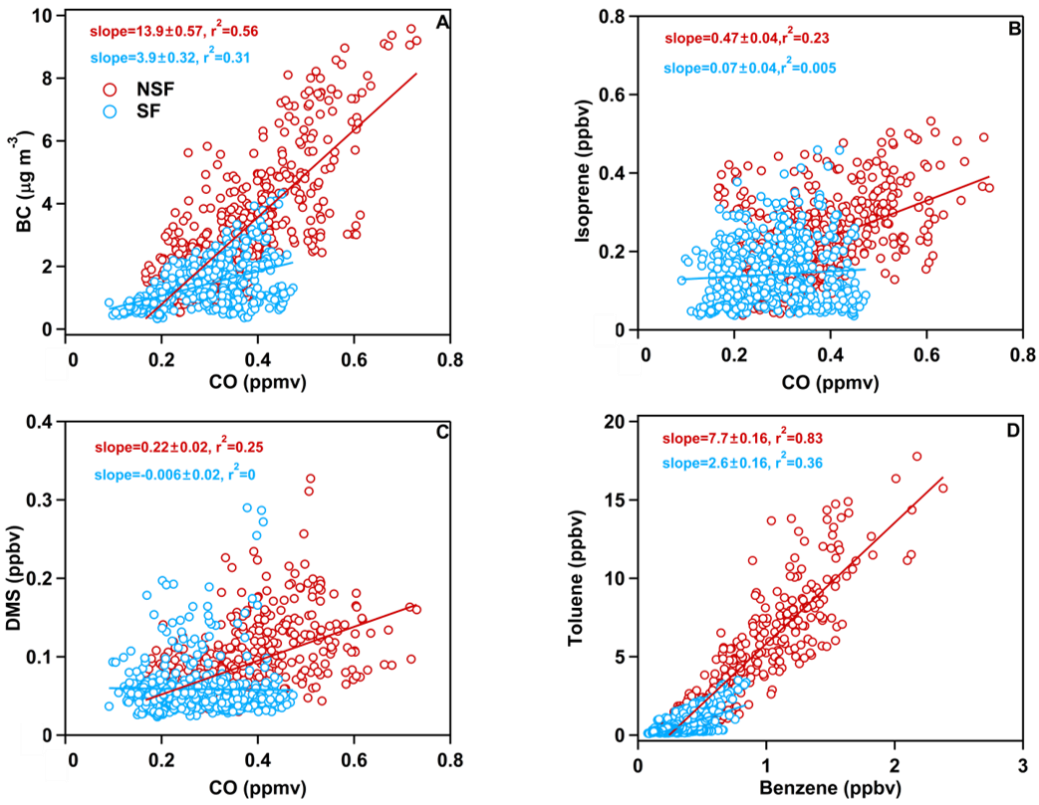
Figure 4. Concentrations of major air pollutants as a function of wind speed during the SF and NSFM periods.

3.4 Emission ratio analysis

Several groups of special correlations were applied to analyze the source characteristics of air pollutants in Figure 5. CO and BC are both products of incomplete combustion (Subramanian et al., 2010), but gaseous CO can travel farther because of its longer atmospheric lifetime (approximately a month for CO vs a week for BC) (Khalil et al., 1990; Ogren et al., 1983). As shown in Figure 5A, the

correlation coefficient and slope between BC and CO during the NSFM period (r^2 = 0.56, slope = 13.9) is greater than during the SF period (r^2 = 0.31, slope = 3.9), suggesting that local combustion sources make a much greater contribution during the NSFM period, but decline significantly during the SF period (He et al., 2011). The concentrations of isoprene and DMS, are not correlated with CO during the SF, whereas their correlation with CO is non-negligible during the NSFM period (Figures 5B and 5C), suggesting again that these pollutants have an anthropogenic source during the NSFM period.

The toluene/benzene ratio can be used to estimate the contribution of traffic emissions (Schneider et al., 2005). Generally, a value of 1.2–3 is found to be characteristic of vehicular emission in many urban areas (Nelson et al., 1984; Wang et al., 2002; Araizaga et al., 2013). The lower ratio of toluene to benzene (ave.=2.6) in the SF period suggests that the dominant source is vehicle emission. This ratio in the NSFM period, however, is much higher (ave.=7.7), indicating more complicated sources of VOCs like huge amount of toluene solvent usage in industrial activities in PRD (Barletta et al., 2005, 2008; Chan et al., 2006). This finding is well consistent with the temporary closure of industrial plants in the SF period, which leads to little toluene emission.



382

383 **Figure 5.** Correlation between air pollutants (A) BC and CO (B) isoprene and CO, (C) DMS and

384 CO, and (D) toluene and benzene during the SF (blue circles) and the NSFM (red circles) periods.

385

386 **3.5 Conclusions**

387 This study uses the SF in Shenzhen to investigate how the urban air quality reacts to significant,

388 temporary reductions in emission. During the winters of 2014 to 2016, the air quality was observed

389 continuously at Peking University Shenzhen Graduate School, from which we obtained the percent

390 change in the concentrations of various air pollutants during the SF periods with respect to the

391 comparable NSF periods. The analysis of these data shows that, despite meteorological variations, the

392 Spring Festival clearly and consistently influences the urban concentrations of various air pollutants.

393 The air pollutants can be divided into three groups: the large-decrease (LD) pollutants are those with

394 a percent change in concentration of −50% to −80% during the SF period and include aromatics, NOx,
395 m/z 57, OVOCs, Chl, BC, and NO_3^-. These results are consistent with the variation in urban emission
396 sources during the SF, suggesting that these pollutants are mostly directly locally emitted or formed
397 from secondary reactions between locally emitted pollutants. The medium-decrease (MD) pollutants
398 are $PM_{2.5}$, NR–PM_1, $PM_{0.8}$, organic aerosol, m/z 44, SO_4^{2-}, NH_4^+, isoprene, acetonitrile, DMS, and
399 CO; the concentrations of these pollutants decrease by 20% to 55% during the SF, which indicates that
400 the extreme reduction in urban emissions during the SF period has limited effect on air pollutants
401 mostly from regional or natural sources. Finally, the slight-decrease (SD) pollutants include SO_2,
402 $PM_{0.8–2.5}$, and O_3. The average percent change in the concentrations of these pollutants during the SF
403 period is less than 20%, which indicates that a significant reduction in urban emissions does not
404 significantly affect their concentration. Of particular interest is the origin of $PM_{0.8–2.5}$, which is almost
405 completely regional.
406
407 The results of this study show that the extreme reductions in urban emissions of Shenzhen only affects
408 the concentration of smaller fresh particles, such as $PM_{0.8}$, whereas the reduction of $PM_{2.5}$ is only
409 slightly affected because of the weak influence on aged, larger particles such as $PM_{0.8–2.5}$. The
410 concentrations of SO_4^{2-} and secondary organic aerosols are hardly affected by local reductions in
411 emissions. Therefore, reducing the emissions of SO_2 and VOCs on a regional scale is critical for
412 reducing their concentrations and achieving the goal of reducing concentrations of $PM_{2.5}$, at least for
413 South China. On the other hand, O_3 has recently become an increasingly important air pollutant in
414 China, especially in the PRD. However, the large reduction of O_3 precursors (NOx and VOCs) during
415 the SF period only lead to small variation of O_3 concentrations. Consequently, further investigations

416 are required to control not only the emissions of VOCs and NOx but also their concentration ratio.

417

418 ## Acknowledgements

419 This work was supported by the National Natural Science Foundation of China (U1301234 &
420 41622304), the Ministry of Science and Technology of China (2014BAC21B03), and the Science and
421 Technology Plan of Shenzhen Municipality.

422

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