Response to Reviewer’s Comments from Anonymous Referee #2

This study presents measurements of gaseous pollutants and aerosols made at a site in Shanghai during the Chinese Spring Festival in 2009. The authors focused on the change in pollution before, during, and after the week-long national holiday, when significant fluctuation in anthropogenic emissions was expected. It was suggested that the elevated pollution levels before and after the holiday was mainly due to the traffic flow out of and into Shanghai, respectively, while fireworks contributed to the pollution episode on the Chinese New Year’s day. The authors also attempted to attribute aerosol light extinction to different species as well as the atmospheric moisture. It is a good idea to estimate the impact of anthropogenic emissions on air quality through such “virtual atmospheric experiments”, but there exist several major issues in both experimental method and data analysis in this study. It is this reviewer’s opinion that the manuscript needs major revisions before meriting publication in ACP.

We appreciate the reviewer for a very thorough comment on this manuscript and we do find it useful for the improvement of this manuscript. In the responses below, we have made the corresponding corrections following the comments point by point. Below are the responses to all the comments one by one.

Major Comments:

1) Methodology

a. The authors provided little information about the site, only indicating that it was on the campus of a university. It appears that the site was in a fairly urban setting. Is it possible that the reduction in traffic emissions near the campus was greater than that in other parts of the city – since the university could be in winter or holiday break? If this is indeed the case, then the representativeness of this site for the study is questionable. In any case, information or estimate on the change in anthropogenic emissions in the study area would be useful for interpret the data.

Yes, we now provide more detailed information on the monitoring site as shown below following the reviewer’s suggestion.

2.1.1 Observational site

The observational site (31.3°N, 121.5°E) in this study is on the roof (~20 m high) of a teaching building on the campus of Fudan University in Yangpu district of Shanghai. Almost no high buildings are around this sampling site. This observational site is approximately 40 km inland from the East China Sea. About 1.3 million residents are living in this area (SMSB, 2011). This site could be regarded as a representative of the megacity Shanghai, standing for the mixing of residential, traffic, construction, and industrial sources (Wang et al., 2006).

References:

SMSB: Shanghai Municipal Statistics Bureau, Shanghai sixth national census in 2010 Communiqué on Major Data (in Chinese), 2011.

Wang, Y., Zhuang, G. S., Zhang, X. Y., Huang, K., Xu, C., Tang, A. H., Chen, J. M., and An, Z. S.: The ion chemistry, seasonal cycle, and sources of PM2.5 and TSP aerosol in Shanghai, Atmos. Environ., 40, 16, 2935-2952, 2006.

To clarify the reviewers' concerns on whether the holiday break in university would potentially impact on our analysis, we have explained more about this site as below:

First, students (also most staffs) in universities of China don't drive to school, which is quite different from that in other countries such as United States or Europe. As they live on campus dormitories, they rely on bicycles, which is almost zero pollution emission. Public transportation (e.g, bus, metro) is still the first choice for most people to travel in Shanghai. Thus, we don't think the holiday break due to school break would have great impacts on the traffic emission since school activities contributed little to the traffic emission throughout the year. Alternatively, we think our site is a suitable site for evaluating the impact due to variation of emission sources. The Inner Ring Road and Middle Ring Road (means more populous areas) traverses through the Yangpu District where the university campus is located. And the university is linked to various metro lines, e.g, Line 3, 4, 8 and 10. This area is predominantly composed of residential communities and its population ranked the 7th among the 18 districts of Shanghai. Thus, we think this site could reflect well the change of air quality due to emission changes such as the spring festival in this study.

We compare the daily concentrations of SO_2, NO_2, and PM_{10} measured in our monitoring site to the reported data from Shanghai Environmental Monitoring Center (http://www.envir.gov.cn/airnews/) as shown in the figure below. FD represents our data and SH represents the average values from all the monitoring stations in Shanghai. It could be seen that our data generally varied consistently with the other dataset and also their magnitudes were close to each other, indicating that this site could be indeed a representative of the Shanghai area.

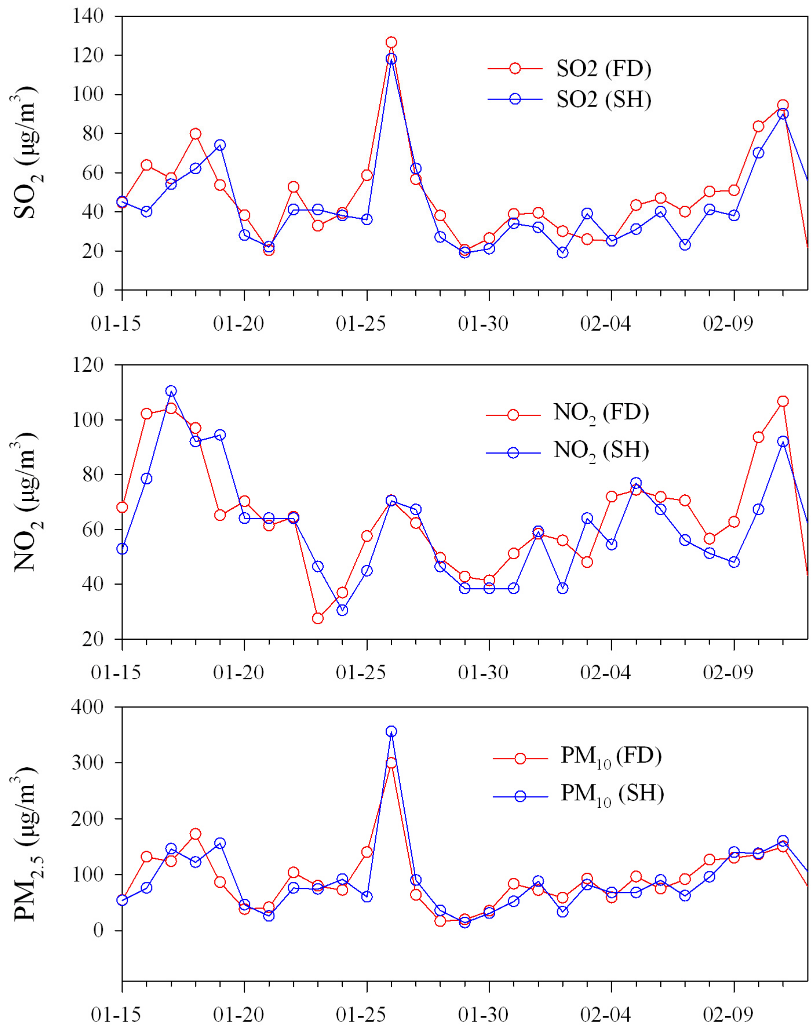

Fig. R1. The daily concentrations of SO_2, NO_2, and PM_{10} measured in our monitoring site (FD) and average values (SH) of Shanghai from Shanghai Environmental Monitoring Center

b. Given the daytime peak in SO2, it is likely that the measurements around noon could represent relatively more regional sources, instead of local emissions. The authors may want to also analyze these data to gain some insight into the regional sources.

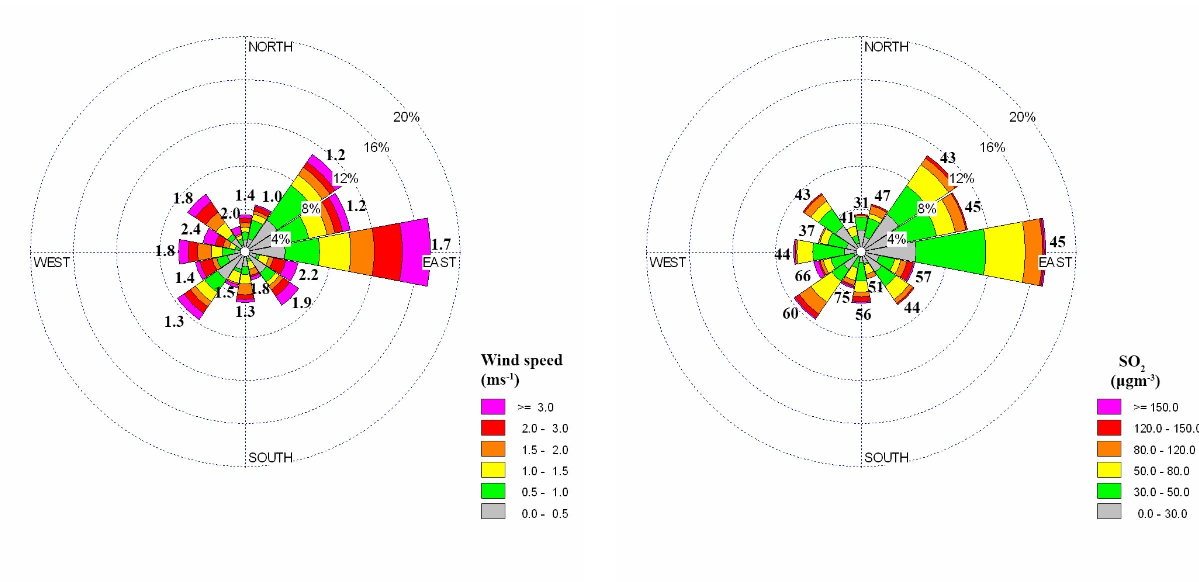

Fig. R2. Rose plots of wind speed (ms^{-1}) and SO$_2$ (μgm^{-3}). Each circle represents the percentage of the winds from a particular direction, and different colors represent the ranges of various parameters. The mean values are plotted at the end of each directional line.

Thanks for the suggestion. Now we use the rose plots of wind and SO$_2$ concentrations as shown above to analyze the possible regional transport. The wind rose plot (left) shows higher frequencies of winds coming from the northeast to the east (clockwise) than the other directions. And higher wind speeds mainly derived from the east to the southeast and from west to north. Generally, the average wind speeds from all directions were lower or close to 2.0 m/s, indicating that the atmospheric convection was relatively weak during this study period. For the rose plot of SO$_2$, higher concentrations mainly derived from south to west (180° – 247.5°). Lower wind speed should be one of the causes as the dispersion of air pollutants was not favored. Another cause could be from the regional transport. Fig. R3 shows the distribution of power plants over the YRD region in 2009. On the pathways to our observation site via the south and west winds, there were a number of power plants located in Shanghai and northern Zhejiang province. Thus, the regional transport of power plant emission from these areas could be an important reason for the higher SO_2 concentrations. Although there was also a considerable amount of power plants located to the northwest of the sampling site (e.g, southern Jiangsu province), relatively low SO_2 concentration was still observed and we attributed it to the higher wind speeds from these directions (see the wind rose plot). Since almost no power plants located in the east, relatively lower SO_2 concentrations were also observed from the direction of 0° – 180°.

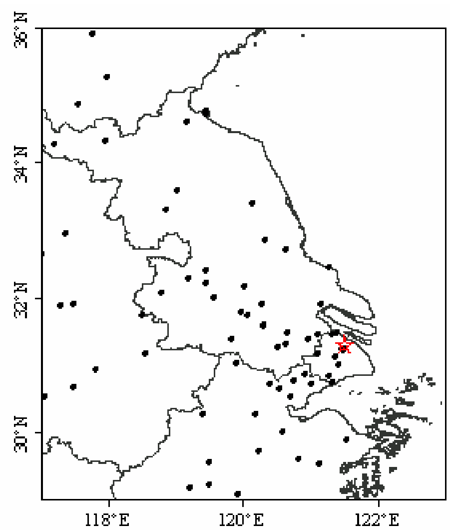

Fig. R3. Distribution of power plants (black dots) over YRD in 2009 (Source: CARMA). The red star shows the location of our monitoring site.

c. How did the TEOM instrument perform under the quite humid conditions encountered during the study? Was the sample flow heated, and to what temperature?

During this study period, the instrument (the inner body and the tube that connected the $PM_{2.5}/_{10}$ sampler) was set at a temperature of 50 °C to avoid the interference of moisture on the calculation of aerosol concentrations. We have addressed this in the revised paper.

d. Any measures to address the potential interference of NOy species on NO2 measurements?

Yes, we do have some measures to eliminate the potential interference of NOy species on NO_2 measurements as much as possible. We used filter to prevent particulate nitrates (e.g, ammonium nitrate) and to adsorb some water soluble gases, e.g, nitric acid and ammonia. In addition, the NO_2 concentration was corrected by subtracting the concentration of HONO measured by DOAS at the same location (Due to use rights, we don't present any DOAS data in this study) to decrease the interference of this NOy species on NO_2 concentrations. However, other NOy species such as PAN, PNN, nitric acid and etc. were not measured due to limitation of the instruments. We have addressed this issue in the revised paper.

e. Was the forward scattering accounted for in the aerosol scattering measurements?

Yes, the forward scattering was included in the total scattering.

f. It is highly likely that the aerosol absorption was overestimated by the Aethalometer, due to multiple scattering and also filter loading effect. Empirical correction method has been proposed in several previous studies (e.g., Arnott et al., 2005). A comparison between absorption and EC may also help.

Thanks for pointing out this data issue of the Aethalometer due to multiple scattering and filter loading problems. Now we have corrected all the aerosol absorption data that used in our study by using a practical algorithm that proposed by Arnott et al. (2005) (Equation #27 in that paper). After applying this correction function, we found that the original aerosol absorption coefficients were indeed overestimated as the reviewer assumed. The corrected aerosol absorption was about 21% lower on average than the original data without correction. And the average absorption efficiency after correction was calculated as 8.56 m^2g^{-1}, which was close to the value in the Pearl River Delta region of China (8.28 m^2g^{-1}, (Wu et al., 2009)).
In the revised paper, we added this correction method into Section 2.3.2. Also, we have made a thorough change of all the context and figures, which were related to the aerosol absorption (e.g, the values of b_{ap} and SSA).

References:
Arnott, W. P., Hamasha, K., Moosmuller, H., Sheridan, P. J., and Ogren, J. A.: Towards aerosol light-absorption measurements with a 7-wavelength Aethalometer: Evaluation with a photoacoustic instrument and 3-wavelength nephelometer, Aerosol Science and Technology, 39, 1, 17-29, 2005.
Wu, D., Mao, J. T., Deng, X. J., Tie, X. X., Zhang, Y. H., Zeng, L. M., Li, F., Tan, H. B., Bi, X. Y., Huang, X. Y., Chen, J., and Deng, T.: Black carbon aerosols and their radiative properties in the Pearl River Delta region, Science in China Series D-Earth Sciences, 52, 8, 1152-1163, 2009.

g. There is no mentioning of the measurement method and location of visibility in the manuscript.

Yes, now we have added the description of visibility measurement into Section 2.2 of the revised manuscript as shown below.
The visibility was measured by using a Vaisala Present Weather Detector (Model: FD12P). Its sensor that combined the functions of a forward scatter visibility meter and a present weather sensor could measure visibility (meteorological optical range, MOR), precipitation intensity, and precipitation type. The detector measured visibility by using the principle of forward scatter measurement within a range of 10 – 50000 m (Vaisala, 2002). All the instruments used in this study were co-located at the same measurement site.

References:
Vaisala: Weather Sensor FD12P User's Guide. Vaisala Oyj, Helsinki, Finland, 2002.

2) Data analysis
While the authors stated that an objective of the study was to evaluate the impact of anthropogenic emissions on air quality in Shanghai (also reflected in the title of the manuscript), they have not separated the effects of meteorological conditions and change in emission sources. For example, the lower aerosol concentrations during the holiday may be related to the stronger precipitation. The higher concentrations of secondary species after the holiday may be due to extended period with high RH. Very stagnant conditions between Jan 26 and 27 may as well have contributed to the pollution episode.

We do agree with the reviewer that meteorological condition is definitely an important factor influencing the air quality. In the writing of Section 3.1, we did discuss the impact of meteorological conditions on air quality though not quantitatively. For example, Line 3-5 in Page 17160 of the original manuscript discussed the stagnant environment during the "pre-holiday pollution episode". And Line 12-13 in Page 17161 discussed the potential impact of high RH on the low visibility after the holiday. For the effect of meteorology during the holiday, we have revised Line 22-23 in Page 17160 as "Afterwards, particle concentrations dropped a lot, probably due to the substantial decrease of fireworks burning and also stronger precipitation on January 29 – 31 (total rainfall amount: 22.1 mm).

Table R1. Pearson linear correlation coefficients between each pair of aerosol, trace gases and meteorological parameters.

	PM_1	$PM_{2.5}$	PM_{10}	Vis[a]	RH[b]	Precip[c]	Temp[d]	Pres[e]	WS[f]	SO_2	NO_2	O_3	CO
PM_1	1.00												
$PM_{2.5}$	0.87**	1.00											
PM_{10}	0.81**	0.89**	1.00										
Vis	-0.57**	-0.54**	-0.37**	1.00									
RH	0.03	0.03	-0.18**	-0.55**	1.00								
Precip	-0.19**	-0.18**	-0.23**	-0.07	0.21**	1.00							
Temp	0.22**	0.30**	0.12**	-0.33**	0.14**	0.04	1.00						
Pres	0.00	0.02	0.02	0.12**	0.20**	0.01	0.05	1.00					
WS	-0.14**	-0.07	0.00	0.30**	-0.38**	-0.02	0.22**	0.07	1.00				
SO_2	0.52**	0.48**	0.56**	-0.06	-0.31**	-0.20**	0.13**	0.00	0.03	1.00			
NO_2	0.52**	0.51**	0.54**	-0.32**	0.05	-0.13**	0.13**	-0.05	-0.27**	0.62**	1.00		
O_3	-0.02	0.03	0.02	0.15**	-0.35**	-0.03	0.38**	0.02	0.48**	-0.02	-0.43**	1.00	
CO	0.53**	0.49**	0.61**	-0.22**	-0.04	-0.15**	-0.16**	0.01	-0.23**	0.49**	0.63**	-0.31**	1.00

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

[a]Visiblity [b]Relative Humidity [c]Precipitation [d]Temperature [e]Atmospheric Pressure [f]Wind Speed

To separate the effects of meteorological conditions and change in emission sources, the best solution is probably using a regional air quality model with a scenario of Shanghai's emission 100% cut off. However, the real emission inventory during this study period is difficult to quantify, especially for the temporal profile as the local activity data (e.g, traffic flows) is hard to predict. Also, modeling is not the main scope of this paper. Instead, we use the linear correlation analysis to qualitatively evaluate the effect of meteorological conditions as shown in the table above. Pearson correlation analysis was conducted on each pair of parameters with significances indicated. Relative humidity, rainfall, temperature, atmospheric pressure, and wind speed were the main meteorological parameters evaluated in the correlation analysis. As shown in Table R1, most significant correlations were found between particles and pollution gases, i.e, SO_2, NO_2, and CO. While particle and gas concentrations showed weaker correlations with the meteorological parameters, suggesting air quality was more dominated by emission than meteorology. RH showed moderately negative correlations with most of gases and almost negligible correlations with particles. Instead, it had a significant correlation with visibility with a correlation coefficient of -0.55. This corroborated with the results in Section 3.4.1 that the contribution of water vapor to the visibility could reach up to around 50%. Precipitation had moderately negative correlations with almost all the gases and particles, especially for PM_{10} due to the more efficient wet scavenging of coarse particles. Temperature had moderately positive correlations with most pollutants, indicating higher temperature facilitated the atmospheric chemical processing. Atmospheric pressure was found almost not correlated to any pollutant. Wind speed seemed to have more impact on the pollution gases while little on aerosol. Overall, we found out that air quality during this study period was still dominated by the emission intensity, and meteorological conditions also exerted impacts on the chemical processing, deposition of air pollutants and possible regional transport.

The manuscript also lacks focus. For example, the part on the diurnal cycle is at best only very loosely related to other parts of the study. Readers can be easily distracted here.

Thanks for pointing out this. In order to make all the sections more closely related to the aim of this study, we have deleted the original section about diurnal variation and made the following changes:

1. We added the correlation analysis between various pollutants and major meteorological parameters to assess the role of emission and meteorology on air

quality (Section 3.2.1 in the revised manuscript). The writing of this section is mainly from the response to the above question.

2. To explore the potential impact of regional transport, we added another section (Section 3.2.2 in the revised manuscript) for this discussion:

The figure below shows the rose plots of wind speed, SO_2, NO_2, and $PM_{2.5}$ by corresponding the hourly wind directions to the other parameters measured at the same time. 16 wind directions were generated. Each circle represents the percentage of the winds from a particular direction, and different colors represent the ranges of various parameters. The mean values are plotted at the end of each directional line. The wind rose plot (Fig. R4a) illustrated that higher frequencies of winds derived from northeast to east (clockwise) than the other directions. And higher wind speeds generally derived from east to southeast and from west to north. The average wind speeds from all directions were lower or close to 2.0 m/s, indicating that the atmospheric convection was relatively weak during this study period. For the SO_2 rose plot (Fig. R4b), higher concentrations mainly derived from south to west (180° – 247.5°). Lower wind speed should be one of the causes as the dispersion of air pollutants was not favored. Another cause could be from the regional transport. Figure R3 below shows the distribution of power plants over the YRD region in 2009. On the pathways to our observation site via the south and west winds, there was a number of power plants located in Shanghai and northern Zhejiang province. Thus, the regional transport of power plant emission from these areas could be an important reason for the higher SO_2 concentrations. Although there was also a considerable amount of power plants located to the northwest of the sampling site (e.g, southern Jiangsu province), relatively low SO_2 concentration was still observed and we attributed it to the higher wind speeds from these directions (see the wind rose plot). Since almost no power plants located in the east, relatively lower SO_2 concentrations were also observed from the direction of 0 – 180°. Compared to SO_2, the NO_2 rose plot (Fig. R4c) showed a different pattern with relatively higher NO_2 concentrations from both north to east (22.5° - 90°) and south to west (157.5° – 247.5°). This was attributed to the widespread traffic networks even when winds came from the ocean. Compared to the gaseous pollutants, the concentrations of $PM_{2.5}$ showed more even distribution from most directions (Fig. R4d) and independent on wind speeds, indicating that local emission should be more responsible for this rather than the regional transport. As shown in Table

R1, $PM_{2.5}$ presented negligible correlations with wind speed while SO_2 and NO_2 both had moderate negative correlations with wind speed, corroborating the different rose patterns among these species in Figure R4. One common characteristic found in Figure R4 was that higher pollutant concentrations mainly derived from south to west, possibly indicative of the impact of regional transport on the air quality of Shanghai.

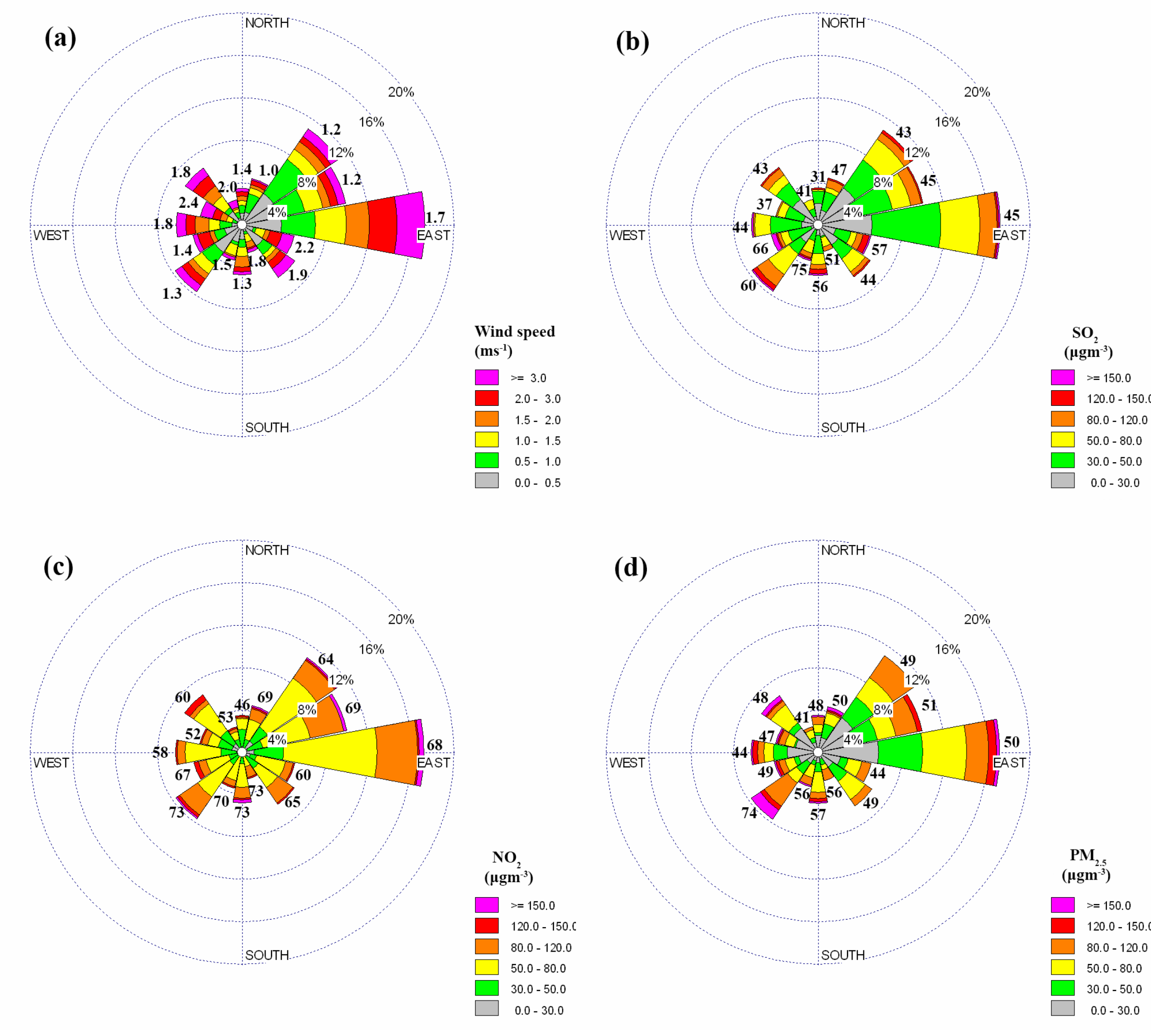

Fig. R4. Rose plots of wind speed (ms^{-1}), SO_2, NO_2, and $PM_{2.5}$ (μgm^{-3}). Each circle represents the percentage of the winds from a particular direction, and different colors represent the ranges of various parameters. The mean values are plotted at the end of each directional line.

PCA was used to determine the major sources of aerosols in Shanghai. The temporal change of these different derived sources or factors during the experiment can probably shed light on the change in anthropogenic emissions during the holiday. The same group of authors recently published a paper also on air quality in Shanghai. The authors may want to highlight in this paper the differences between the two studies, and what new conclusions can be drawn from this study.

Thanks for the good suggestions of adding the temporal variations of different sources that determined by the PCA analysis. Now we show the time-series of selected typical elemental tracers in $PM_{2.5}$ from the five major sources determined by PCA as shown in Figure R5 below. The time-series of mineral source (PC1: Al, Ca, Fe, Na) showed high concentrations before the holiday. On the one hand, busy transportation before the holiday caused more suspension and re-suspension of road dust. On the other hand, relatively low humidity (Figure 1 in the original manuscript) didn't favor the deposition of road dust. It was noticed that highest peaks of mineral elements occurred from 23 to 25, January. If we check the meteorological conditions in Figure 1, we could find that RH had a sharp decrease from 23 January to around 20% in the next few days. In figure 1e, the $PM_{2.5}/PM_{10}$ ratio was observed to be the lowest ratio of 0.15 during the whole study period. Thus, this typical meteorological condition and characteristics of aerosol inferred that there was a cold front brining floating dust which was mainly composed of coarse particles rich in mineral elements. After the holiday, the levels of minerals stayed at low concentrations at most circumstances. Higher humidity may be one of the causes for lowering the mineral aerosol. The traffic source (PC2: Mn, Mo, Ni) generally had a consistent temporal variation with the traffic related gases (e.g, NOx, CO) in Figure 2, corroborating the effect of intensified traffic emission on aerosol chemical species during the two specific periods of the spring travel rush as discussed in Section 3.1. Tracers for the fireworks burning (PC3: K, Ba, Sr) had small fluctuations during the study period except the obvious enhancement on January 25, which was exactly the New Year's Eve. As for the industrial source (PC4: Cd, Sb), higher concentrations occurred before the holiday and then it decreased during the holiday. After the holiday, it gradually increased again due to re-opening of factories. The temporal variations of the coal burning source (PC5: As, Se, Ge, S) coincided well with that of SO_2 (Figure 2) and particulate SO_4^{2-} (Figure 4). Higher elemental concentrations mainly occurred after the holiday and this was attributed to the higher electricity demand when people were heading back home.

In the revised manuscript, we have added these discussions as part of Section 3.3.2 following the section of the PCA analysis.

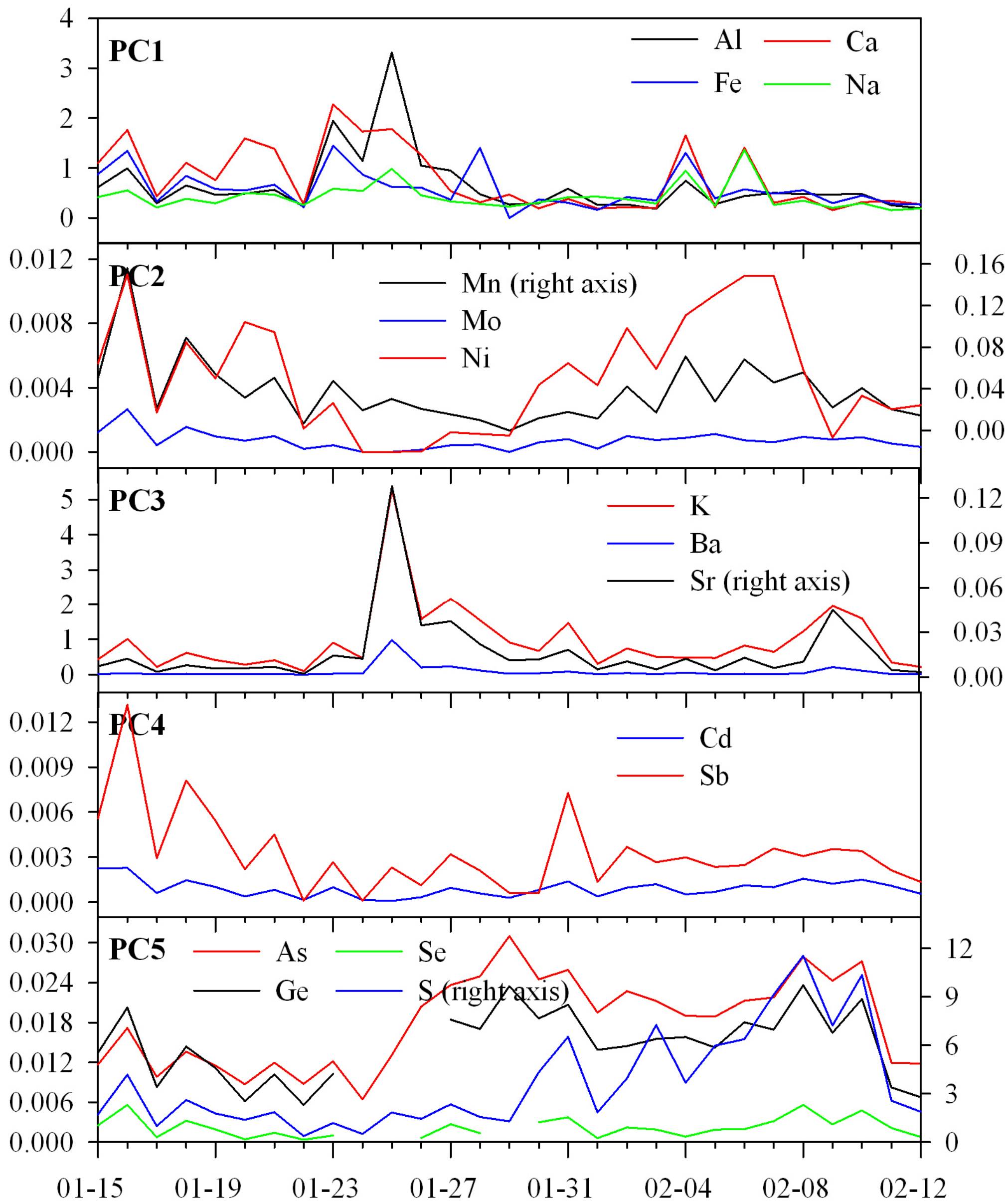

Figure R5. Time-series of typical elements from five major sources as determined by the Principle Component Analysis (PCA).

Also, we now highlighted the difference between this paper and our previous paper (*Huang et al., 2012.* Title: Typical types and formation mechanisms of haze in an Eastern Asia megacity, Shanghai) in the Introduction part. Please refer to the revised manuscript for the changes.

Page 17162, Line 15-24: as already mentioned, the daytime peak in SO2 was likely caused by boundary layer processes, not the change in utility generation.

Please refer to the response to the Major Comments 1b in this text.

At last, we would like to thank sincerely the Anonymous Referee again for his/her efforts in improving this manuscript.