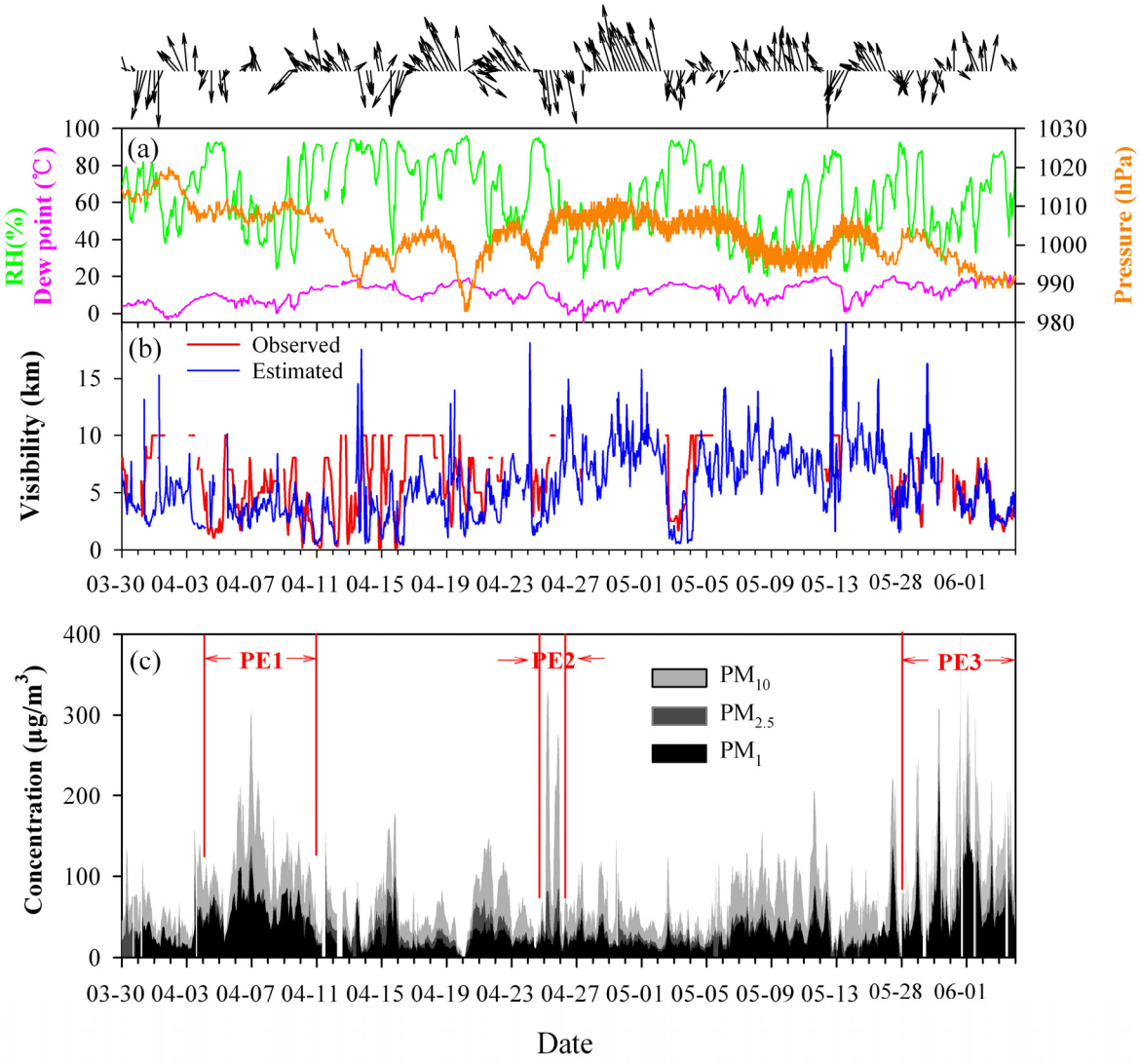
Response to the comment by Anonymous Referee #1

In this study, Huang et al. presented measurements of aerosol and trace gases at a site in Shanghai, China from April to June 2009. Three major haze episodes having distinct characteristics were observed during the experiment, under different meteorological conditions. Using the correlation between gaseous pollutants (CO, SO2, and NOx) and PM2.5, chemical tracers of dust (Al) and biomass burning (K+ and OC, EC), as well as satellite and ground-based lidar data, the authors were able to attribute the three episodes to anthropogenic/industrial pollution, long-range transport of dust from Gobi, and agricultural fires in eastern China. Overall, the manuscript is based on a quite comprehensive, original data set collected from a megacity known to have severe air quality issues. The topic is suitable for the special issue and should also be of interests to the wide science community. The results were presented in an organized fashion. On the other hand, this reviewer found the manuscript lacking quantitative, in-depth analysis, and missing some important details. Also the quality of the writing needs to be improved: there are numerous grammatical mistakes and typos in the manuscript (too many to list blow), making it difficult to follow at times. Major revisions are recommended before the manuscript can be accepted for publication in ACP.

We thank the reviewer for a thorough comment on this manuscript. In the revised manuscript, we made those corresponding corrections, following the comments point by point. In addition, we improved the quality of the writing and corrected those grammatical mistakes and typos. Below are the responses to all the comments one by one.

Specific Comments: In the manuscript, the authors focused on the impact of different sources on the three types of haze episodes. And this brings an interesting question: the sources and transport of dust and biomass burning emissions can well explain the episodic nature of episodes 2 and 3, then how about episode 1? The industrial and traffic emissions are not expected to change dramatically over two months, but why was episode 1 so much more polluted than the “normal period”? This is likely due to change in meteorological conditions, and the authors are encouraged to look into the meteorological mechanisms for the formation of major haze events.



Yes, thanks for the good suggestion of adding the meteorological parameters to look deeper into the formation of haze under different conditions. In the revised manuscript, we have added several meteorological parameters, i.e., wind speed and direction, relatively humidity (RH), dew point and the atmospheric pressure in Figure 1. During PE1, the local air was under a low pressure system turning from a high one several days ago as shown in Fig. 1a. Winds blew from various directions and the wind speeds were relatively low and even were stagnant sometimes, which were not favorable for the dispersion of pollution. There was an obvious decrease of relative humidity (RH) during PE1. Cold front from inland China probably contributed to the high pollution of PE1 to some extent. The similar meteorological conditions were also observed during PE3. While during PE2, winds dominantly blew from northwest with high speeds, consistent with the backward trajectories in Fig. 3 in the manuscript. Due to the cold front from northern China, dew point and relative humidity both decreased a lot. During the normal days, i.e, the relatively clean periods, we could find that winds blew from the southeast at most times, i.e. from the East China Sea. Also,

the wind speed in those normal days was usually higher than that in the haze events (except for PE2). Thus, we did find that the change of meteorological conditions could impact greatly on the formation of haze events as the reviewer suggested.
Actually, we did mention the role of meteorological conditions on the haze formation in some parts of Section 3.1. For instance, the three days air mass backward trajectory computed by the HYSPLIT model in Fig. 3 showed the wind direction and speed, which was consistent with the surface meteorological measurements in Fig. 1a. Also, the height of PBL layer shown in Fig.3 was another important parameter for assessing the convection of air pollution. During PE1 and PE3, the average daily PBL height was around 500 meters. Thus, we think that the low PBL layer could also contribute to the haze formation.
In the revised manuscript, Fig. 1 was revised, as shown above. Also, we added more detailed description of the possible role of meteorological conditions in the haze formation.

Section 2: a more detailed description should be given for the site – is it urban, suburban, or rural? Are there any major local emission sources (point sources, roads), and are they expected to have impact on the data set? And how is this mitigated? Is the site truly representative of a megacity? Also more detailed information is needed for gas analyzers – for example, what kind of calibration standard was used? Is zero check every week sufficient for the CO analyzer, among other instruments?

Yes, we have added the description of the observational site into Section 2.1 as follows: 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, Shanghai. Almost no high buildings are around this sampling site. This observational site is approximately 40 km inland from the East China Sea. The closest industrial sources are located approximately 10.5 km away and are primarily to the southeast and northwest (Li et al., 2011). 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).

The routine QA/QC procedures includes the daily zero/standard calibration, span and range check, station environmental control, staff certification, etc., according to the Technical Guideline of Automatic Stations of Ambient Air Quality in Shanghai based on the national specification HJ/T193 - 2005, which was developed following the technical guidance established by the U.S. Environmental Protection Agency (U.S. EPA) [1998]. The multi-point calibrations were applied weekly upon initial installation of the instruments. At least five points were used for calibration, consist of three or more test concentrations, including zero concentration, a concentration between 80% and 90% of the full scale range of the analyzer, and some intermediate concentrations. If some instruments have malfunction, the multi-point calibrations are also used to verify the linearity of instruments. On a daily basis, a two-point calibration method was applied, which is easier and quicker.
In the revised manuscript, we have added more detailed information about the QA/QC.

P21719, line 2, a fixed lidar ratio was used in spite of various aerosol types (industrial, dust, and biomass burning) observed at the site?

Yes, we admit that using a fixed lidar ratio under different types of haze is not very reasonable. Thus, in Section 3.4 of discussion about the aerosol vertical profile, we only quantitatively discussed the vertical profile of aerosol backscattering coefficient, while the vertical profile of aerosol extinction coefficient was only qualitatively discussed. We just want to state that a lidar ratio of 50sr was only applied to derive the extinction coefficient under normal times in Section 2.1.3,. While in some special haze events, i.e, dust and/or biomass burning, a different lidar ratio should be used. However, as the mixing proportion of dust, biomass burning aerosol, and secondary inorganic aerosol is difficult to accurately determined, we didn't look very deep into the lidar ratio under different haze events.

P21721, line 18: why made the comparison between two stations that are 32 km apart?

At the observational site in this study there was no instrument directly measuring the visibility. For the purpose of evaluating the estimated visibility by using the formula shown in Section 3.1 (now move to Appendix A), we chose the available visibility data from the meteorological station at Pudong of the closest distance to our observational site. Although our site is 32 km away from this station, we think that this comparison is still reasonable, as the visibility within a narrow area should be relatively homogeneously distributed in the haze events. However, in the future study we will try to use the datasets at the same location for comparison or evaluation.

P21722, line 1: are the PM1, PM2.5 and PM10 mean concentrations given above the average of hourly or daily data? 24-h mean should be used to compare to the 24-h standard. P21722, line 15: air mass usually refers to large body of air of similar characteristics (e.g., Arctic air).

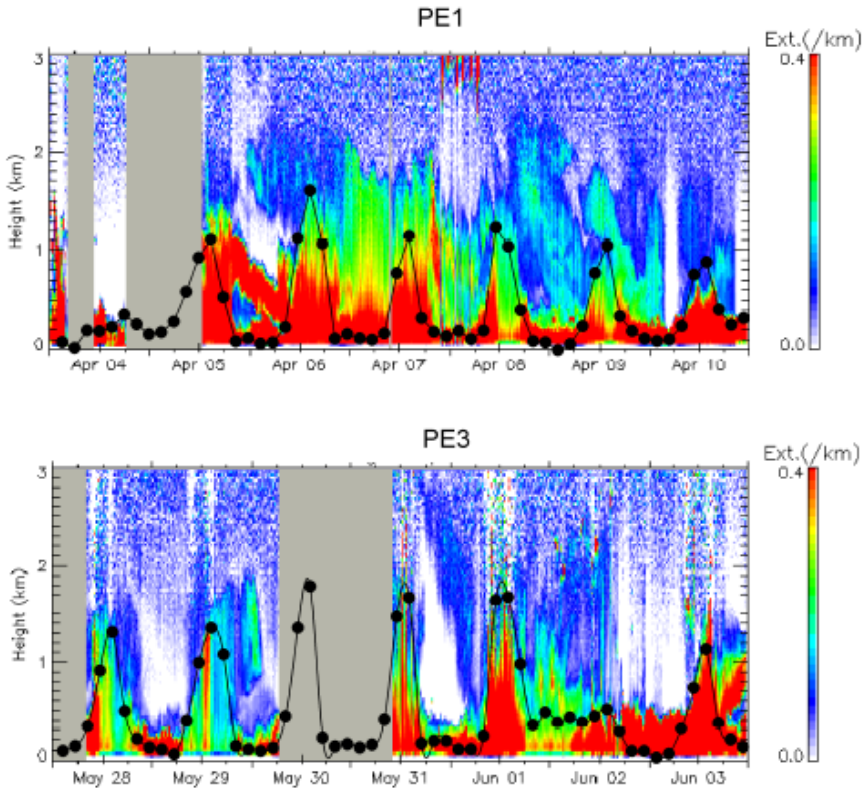
Yes, the PM_1, $PM_{2.5}$, and PM_{10} mean concentrations given are the average of 24-h concentrations. Yes, we have changed the term "air masses" into "air flows" here and elsewhere in the revised manuscript.

P21722, line 17 and Fig. 3: how was mixing height determined? Is it the mean along the trajectories? How does it compare to lidar data? Lower panel of Fig. 3b shows daily mixing height but the caption indicates it is 3-h average?

The mixing heights shown in Fig. 3 are computed from the NCEP Global Data Assimilation System (GDAS) model (http://ready.arl.noaa.gov/READYamet.php). The model outputs are 3-hr at the time resolution, thus the blue dots in Fig. 3 denoted the 3-h average mixing height. Also, we calculated the daily average mixing height as indicated by the pink horizontal step lines. In the revised manuscript, we have added the data source of the mixing height.

The modeled PBL heights are overlaid with the aerosol vertical profile measured by lidar. As

shown in the figures below, the temporal variations of modeled PBL heights (black dots) are relatively consistent with the PBL heights visualized from lidar at most times. This comparison suggests that both datasets are reliable.



P21723, line 18: April should be June?

Yes, this is typo. Thanks for pointing out this mistake. It was corrected in the new version.

P21724, line 11: the authors should not assume that the readers all have a good knowledge of the geography of China. Would recommend that the authors mark the important features/locations/regions on the map of Fig. 5.

Yes, thanks for the good suggestion of marking the major locations on the China map. However, we find it difficult to see clearly if too many descriptions are added on Fig. 5, as this figure is composed of various colors. Alternatively, we added the locations on Fig.4, as this figure has more blanks and it is easier to look at.

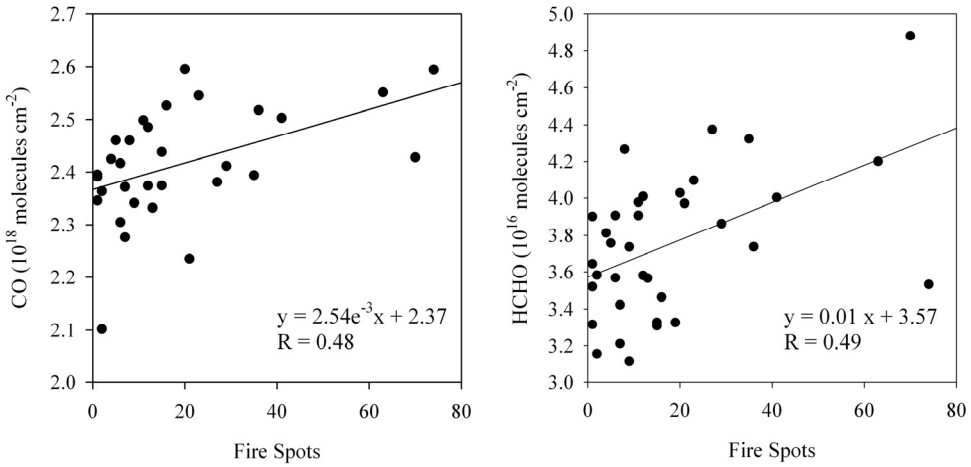
Section 3.2: overall, the analysis using satellite data is problematic. The authors should demonstrate that the Angstrom exponent from MODIS is a reliable product before using it to distinguish various aerosol types (refer to Levy et al., ACP 2010 for a recent validation study of the product). Otherwise, Angstrom exponent from ground-based sun photometers can be used.

There seem to be artifacts in the processing of the satellite data, as Fig. 5a-d look odd with stripes and small data gaps. Also has OMI row anomaly been removed from the HCHO data shown in Fig. 5g?

Yes, we agree to what the reviewer said here. The satellite products usually have uncertainties due to the algorithm assumptions, different retrieve methods, and the interferences from clouds, precipitation, etc. As pointed out by Levy et al. (2010), the Angstrom exponent product has greater uncertainty than the other MODIS products. However, its values are still within reasonable range as compared to the ground-based AERONET products. As the reviewer suggested, Angstrom exponent from ground-based sun photometers should be better used for charactering the aerosol sizes. Unfortunately, in this study there was no AERONET data available in the region of the Yangtze River Delta. Even now there is no AERONET data available in the whole mainland China (refer to the global map of AERONENT sites distribution in http://aeronet.gsfc.nasa.gov/cgi-bin/type_piece_of_map_opera_v2_new). Thus, we have no choice but only are able to use the satellite data. Additionally, Angstrom exponent retrieved from satellite still has its advantage compared to the ground-based instrument, as it can provide a clearer picture of the regional distribution of aerosol optical properties. For instance, from Figure 5d we could confirm that the pollution during PE2 was caused by the large-scale dust storm but not local pollution such as construction works near the sampling site. Also, in the discussion of the Angstrom exponent in the manuscript, we didn't tend to quantitatively analyze it as we know that this parameter is highly uncertain. Instead, we focused on the qualitative analysis, e.g, the comparison between regional distribution of Angstrom exponent during PE1 and PE2, which gave a visualization of the different aerosol size. In the future study, we will try to use more ground-based techniques to accurately characterize the type of aerosols.

As for the stripes and small data gaps in the satellite image, it was due to that some grids have no data owing to the satellite scan gaps and relatively sparse pixels and bigger footprints at the marginal granule. Also, the interference of clouds and/or precipitation could also shield some information of aerosol. Thus, stripes and small data gaps should not be from the artifacts in processing the satellite data.

P21725, line 20: a more quantitative demonstration of the spatial correlation between CO and fire counts would be helpful. Similarly for HCHO and fire counts mentioned in line 11, P21726.



Yes, we have demonstrated the spatial correlation between CO, HCHO and fire counts as the reviewer suggested. All the data are grided with resolution of 0.5 × 0.5 degree and then the linear regression was applied to all the data at the same grid. As shown in the figures above, CO and HCHO both had moderate correlation (R ~ 0.49) with the numbers of fire spots, indicating the impact from biomass burning on the trace gases. However, the correlations were not very significant, as the effect of meteorological conditions were not considered. Overall, the linear correlations were still reasonable. In the revised manuscript, we have added more quantitative discussion of the correlation between CO and HCHO with fire spots.

P21726, line 21: why would sea breeze clean aerosol but not CO or HCHO?

Actually, we observed that the sea breezes tended to clean more aerosol (AOD) and HCHO than CO as shown in Fig. 5. We attribute the different behaviors to their different life times. Aerosol contains a considerable fraction of soluble species, i.e, sulfate, nitrate, ammonium salts, and soluble organic aerosol, which are easily subject to be scavenged by see breezes with rich moisture. For HCHO, it is highly water soluble. Also, HCHO is easy to be oxidized by some oxidants such as OH radical, O_3, and etc. That's why we found that aerosol and HCHO were mostly scavenged over the East China Sea. For CO, its life time is much longer than aerosol or HCHO and it is relatively not easy to be oxidized. Thus, we could find that the transport distance of CO was longer than AOD and HCHO and the spatial distribution of CO is more dispersed.

P21728, line 28 and Fig. 6: this part is confusing. It appears that the Ca/Al ratio was also small for other days and is quite noisy – does not seem to be a good indicator for the dust source region.

The "noisy" Ca/Al ratios could be due to several causes. Shanghai is a fast developing city with frequent construction works almost all year around, such as housing demolition, municipal construction and road renovation. Enriched Ca content in aerosol in the urban cities mainly derived from the construction works (Wang et al., 2006; Yang et al., 2005). Also, the re-suspended dust lifted by the vehicles contained high-Ca aerosol. Compared to the Ca/Al ratio of 0.5 in the dust from the Gobi Desert, the Ca/Al ratios during the normal times were usually around or above

1.0, due to the enrichment of Ca. The Ca/Al ratios during the study period seemed noisy because the intensity of construction works and traffic activities varied intensively daily. Thus, the fluctuation of Ca/Al ratios was expected and reasonable. There did have several low Ca/Al ratio case, i.e, on May 4 and 12. However, the mineral (Al) aerosol concentration was relatively low, indicating that these days were not influenced by the transport of dust and were relatively clean episodes from the local sources.

We used the Ca/Al ratio as an indicator for the dust source region as it had a much lower value (0.5) in the Gobi Desert than in the local sources of Shanghai. When the local environment is influenced by the entrained dust, there would be an obvious decrease of the Ca/Al ratio. In this study, the Ca/Al ratio decreased to 0.75 when Al reached 13.7 μg m^{-3}. However, this ratio was not close enough to the source region (i.e, 0.5) as the reviewer questioned that there were also other days with small Ca/Al ratios. That's because the dust event in this study was a weak event, the entrainment of dust couldn't dominate over the local dust. During a stronger dust event on April 2, 2007 in Shanghai, the daily TSP concentration reached 806.2 μg m^{-3}, and the Ca/Al was lower with the value of 0.67 (Huang et al., 2010). Thus, the feasibility of using the Ca/Al tracer for the dust transport in downstream regions depended on the intensity of entrained dust and local anthropogenic dust. If the dust event is strong enough, we think it was a good choice to use the Ca/Al ratios to trace the dust source region.

P21729, line 29: what is the ratio between K+ and Cl-? Cl- can also be derived from coal burning.

In this study, the K^+/Cl^- ratio during biomass burning pollution was 2.09. Ryu et al. (2004) summarized the K^+/Cl^- ratios during biomass burning in various regions of the world, as shown in the table below (note that Cl^-/K^+ ratios are presented in the table). The K^+/Cl^- ratios ranged from 1.29 to 2.63 except the extremely low result in Africa savanna. Thus, the result of our study fell well into this range.

Table 6. Differences in chemical composition among the different types of biomass burning aerosol.

Region	Type	K^+/NO_3^-	K^+/SO_4^{2-}	K/BC	Cl^-/K^+	OC/EC	Remarks
Northeast Asia	Rice straw	0.11	0.11	0.24	0.71	8.3	This study, BBR
(agricultural waste)	Barley straw	0.33	0.24	—	0.75	7.3	This study, BBB
Europe (Spain)		1.3	0.41	—	0.62	—	Ezcurra et al.[25]
(cereal waste)							
Africa (savanna)	Airborne	0.97	0.61	0.62	3.89	2.7	Andreae et al.[8]
	Flaming	—	—	0.2	—	—	Maenhaut et al.[5]
	Smoldering	—	—	0.17	—	—	
Tropical forest	Flaming	0.15	1.13	0.11	0.38	—	Yamasoe et al.[9]
	Smoldering	0.3	0.98	0.1	0.5	—	
Brazil (Amazon basin)	—	—	—	0.6	—	—	Ferek et al.[26]
	Flaming	4.84	4.17	0.24	0.73	—	Yamasoe et al.[9]
	Smoldering	3.71	3.94	0.2	0.77	—	

Cl^- could be derived from coal burning as the reviewer said, because coal has always contributed to the biggest share of the energy structure in Shanghai. The background (i.e, when fossil fuel

combustion dominated) K^+/Cl^- ratio in Shanghai ranged from 0.10 to 0.46 based on a seasonal analysis (calculated from Table 3 in Wang et al. (2006)). During PE3 in this study, K^+/Cl^- ratio was much higher than its background ratio, indicating the dominance of biomass burning source. More detailed source profile is needed to quantitatively analyze the contribution from various sources. In the revised manuscript, we have added discussion on the possible source of Cl^- from coal burning.

P21730, line 9-14: from the results of Yamaji et al. (cited above), the OC/EC ratio during MTX2006 was closer to 3 (Zhang et al., 2008) than to the results given here.

Yes, there seems inconsistence between this study and Yamaji et al. (2010) on the issue of OC/EC ratios from biomass burning. However, from the table above (Ryu et al., 2004), they did also observe higher OC/EC ratios during biomass burning in northeast Asia, which was consistent with our results. We thought that the relatively low OC/EC ratio in Yamaji et al. (2010) could be due to the mixing of local anthropogenic sources (e.g, industrial, traffic) with biomass burning. Shandong province, where the MTX 2006 campaign took place, is a big province of China with heavy anthropogenic emission. The background aerosol composition is high, which could probably shield the signature of biomass burning.

P21731, line 8-17: the enrichment of As in agricultural fire emissions is a very important result – any more data on As emissions from agricultural fires?

Yes, the enrichment of As in agricultural fire emissions is an important issue due to its adverse effects on human health and ecosystem. We do have multi-year aerosol As data at several sites of China, i.e, Eastern China, North China Plain and Western China. The next step of our study will be to gather more cases to investigate the relation between biomass burning and As enrichment in aerosol.

Section 3.4: from Fig. 6 one can tell that soil or dust was a sizable part of aerosols on April 6, however this was not reflected in the lidar data?

Yes, dust was a significant part in the total suspended particles (TSP) on April 6. However, the secondary aerosol also presented the highest concentration on the same day as shown in Fig. 6. Thus, the abundant fine particles probably disguised the characteristics of coarse particles as coarse particles had much smaller backscattering efficiency than fine particles. As indicated from the depolarization ratio detected by the lidar, its values were almost lower than 5% on the whole day of April 6, indicating the dominance of spheric particles.

P21734, line 1, PE3 should be PE1?

Yes, this is a typo. We corrected “PE3” to be “PE1” in the new version.

P21735, line 5: recent studies show that SO2 emissions might have started decreasing in China (refer to Li et al., GRL, 2010 and Lu et al., ACP, 2010).

Yes, thanks for proving these two useful references. We have added into the discussion.

P21735, line 19: NO3-/SO42- not SO42-/NO3-?

Yes, it should be NO_3^-/SO_4^{2-} , not SO_4^{2-}/NO_3^-. We did correct it in the new version. Thanks for pointing out this mistake.

P21737, line 10: there should be updated emission inventory for the region.

Yes, we replaced the original emission inventory with the latest INTEX-B emission inventory in Asia (Zhang et al., 2009) and re-calculated the results.

P21738, line 27: use of coal should be more or less correlated with utility generation and could be on the rise in China over the past decade or so.

Yes, we agree to what the reviewer said here. The emission from power plants are the main source of SO_2 in China and the utility generation from coal usage has been increasing over the past decade.

Based on the two reviewers' concern on Section 4, they both believe that this section seems not very closely linked to the main theme of this paper. The authors decided to remove this section out in the new version to make the whole paper more focused on its main theme. Thanks for this reviewer's hard work on this section.

P21739, line 14: there are plenty of minerals as shown in this and previous studies, even if NH3 emissions are reduced.

Yes, we agree with the reviewer's statement. Minerals are also important for the formation of secondary aerosol as it can serve as alkaline surface for the heterogeneous reaction. In this study, most minerals accumulated in coarse particles. Minerals in fine particles (i.e, $PM_{2.5}$) only accounted for about 11% in the total suspended particles (TSP). Thus, control of minerals should be beneficial for the mitigation of coarse particles. In those coastal urban cities in China, such as Shanghai, minerals mainly derive from the construction works and sometimes from the dust transport. Our recent results show that PM_{10} concentration during the 2010 Shanghai Expo had been significantly reduced due to the temporary shutdown of construction sites. However, to

reduce the fine particle emission, control measures on the NH_3 emission may be more efficient as sulfate ammonium and nitrate ammonium are mainly formed in fine particles, not in coarse particles. A very recent study applied the Response Surface Modeling (RSM) technique in the Community Multi-Scale Air Quality Model (CMAQ) to study the response of inorganic species to the change of anthropogenic emissions (i.e., NH_3, SO_2, NOx) (Wang et al., 2011). They found that in the most polluted areas in China such as NCP (North China Plain), and YRD (Yangtze River delta), NH_3 was sufficiently abundant. The 90% increase of NH_3 emissions during 1990 - 2005 resulted in ~50-60% increases of NO_3^- and SO_4^{2-} aerosol concentrations. It is suggested that the most effective pathway is to adopt a multi-pollutant strategy to control NH_3 emissions in parallel with current SO_2 and NOx controls in China.

Section 4: it would really be useful for the authors to give some statistics on different types of haze events: how many inorganic ones, how many biomass burning ones, and how many dust ones? That way, the quality of the paper will be much improved, and the results would be more helpful for the policy makers.

Yes, thanks for the very good suggestion on summarizing the statistics on different types of haze events. At this stage, however it is difficult to finish this task during a very short time. In Shanghai, we started seasonal aerosol sampling from late 2004 till now. To analyze and differentiate from case to case of total 6 years' data will be a really big project in a limited time. In addition, the chemical analysis in laboratory is still ongoing for recent sampling periods (e.g, 2011). This paper could be regarded as a pilot study for the typical haze types in Eastern China. In the further study, as the reviewer suggested, a more comprehensive and long-term study of this issue will be conducted and the results will be expected to contribute to the community and be helpful for the policy makers.

P21741, line 1: a paper on the 2010 dust recently came out in Atmos. Environ. (Wang et al., First detailed observations of long-range transported dust over the northern South China Sea, 2011).

Yes, thanks for providing this reference.

Fig. 1b, may consider using different colors for PM2.5 and PM10. Also it would be helpful if the three episodes can be marked in the figure.

Yes, we have used different colors for better visualization and the three episodes are marked in the

revised figure.

Fig. 2, is gas concentration for standard temperature and pressure?

Yes, it is for standard temperature and pressure.

Fig. 7, the high EF of V for PE1 is interesting, and may deserve some discussion.

The EF of V during PE1 is about 11, indicating slight enrichment. It could be from the re-suspension of local soils and road pavement erosion (Amato et al., 2009).

Fig. 9, a and c are average profiles for the entire day? Fig. 10, what do the error bars stand for?

Fig. 9a and 9c represent average profiles of several hours on a certain day. We have added the detailed time information into the manuscript.

Error bars in Fig. 10 represent one standard deviation of the gaseous concentrations during different periods. We added the illustration of error bars in the caption of this figure.

Fig. 12. The CO/PM2.5 ratio seems to be higher in PE1 than in PE3, can you give more discussion on that?

The CO level was similar between PE1 and PE3 as shown in Fig. 10. However, the average $PM_{2.5}$ concentration was 84.0±48.4 μg m^{-3} during PE3, higher than that of 63.6±16.4μg m^{-3} during PE1 as discussed in Section 3.1. Thus, the higher $CO/PM_{2.5}$ ratio during PE1 was due to the higher $PM_{2.5}$ concentrations during PE3. This probably meant that biomass burning could produce higher aerosol mass than the industrial/traffic emission as most of the biomass burning events are incomplete combustion, which couldn't effectively oxidize the hydrocarbon to CO_2 or CO.

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