We are thankful to the two reviewers for their thoughtful comments and suggestions that help improve the manuscript significantly. We have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to each reviewer's comments

Response to reviewer #1

This manuscript reports one-year measurements of non-refractory submicron particles by Aerodyne Aerosol Chemical Speciation (ACSM) at an urban site in Beijing. Temporal variations of the particle concentration and composition as well as their associations with meteorological conditions are explored. The authors also provided footprint analyses for the potential source regions of aerosol components. Overall, this paper is well written and clearly describes the analysis. The paper addresses relevant scientific questions within the scope of ACP. Some results however seem to be over-interpreted and need additional constraints. New scientific findings also need more emphasis in comparison with previous work. I recommend this manuscript be published after the following specific comments are addressed.

Specific comments:

(1) In Abstract and Conclusion, it is not clear to me what the new findings are compared to previous studies.

We present the first long-term, highly time-resolved measurement of fine particle composition in Beijing, China. Compared to previous studies that either focused on limited aerosol species, relied upon weekly filter samples, or used one month's worth of data to represent an entire season (Zhang et al., 2013a; Zhang et al., 2013b), this study provides a full spectrum of seasonal variations and diurnal patterns of aerosol species in Beijing, China. This is important for a better understanding of the compositions, sources, and evolution processes of severe haze pollution among the different seasons and hence mitigating fine particle pollution in Beijing. New findings were also obtained in this study, for example, we observed insignificant seasonal variations of secondary inorganic aerosol which is different from the results reported in previous studies (Zhang et al., 2013a). The reasons for such inconsistency were elucidated in the manuscript. We also identified high potential source areas for different aerosol species during the four seasons which were rarely reported in previous studies. These results provide evidence and demonstrate the importance of regional transport in the formation of severe haze pollution in Beijing. In addition, we had a more comprehensive investigation of the meteorological effects, particularly relative humidity and temperature on the formation of different aerosol species with the advantage of 1 year data.

(2) Section 2.4: It is unclear how the footprints of air masses are converted to potential source concentrations of aerosol species. The authors should add a bit more details about the PSCF method.

Following the reviewer's comments, we expanded the PSCF method in section 2.4 in the revised manuscript.

(3) Page 14557, line 16-18: Many possible reasons can lead high mass concentration of NR-PM_1 in June compared to July and August. If the authors really think this is biomass burning impacts, they should provide evidence to prove, for example, fire product near the site or upwind. Mass spectral marks of biomass burning OA may also help.

Good point, we do have evidence. As shown in Fig. R1, a large number of fire spots were detected in north China plain including Beijing surrounding regions during the late June, suggesting the ubiquitous agricultural burning during the summer harvest season. In addition, PMF analysis was also performed on organic aerosol (OA) in June, and three factors including a biomass burning OA (BBOA) were identified (Fig. R2). The BBOA spectrum shows obvious *m/z* 60 and 73, which are two marker *m/z*'s for biomass burning (Alfarra et al., 2007; Cubison et al., 2011). Note that the signal intensities of *m/z* 60 and 73 were not as high as those from fresh biomass burning (Cubison et al., 2011), indicating that BBOA in this study was aged when transporting from surrounding regions to the Beijing city. The time series of BBOA in June (Fig. R2b) showed high concentrations during 18 – 22 June, further supporting the large impact of biomass burning on aerosol loadings.

Following the reviewer's suggestions, the plot of MODIS fire spots was added in supplementary to support our conclusions.

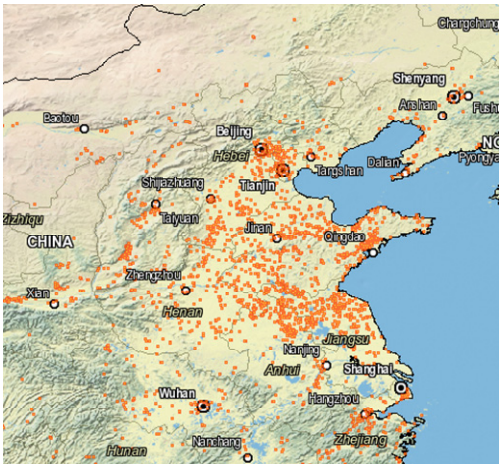

Fig. R1. Fire spots in north China plain during 15 – 30 June, 2012 (https://firms.modaps.eosdis.nasa.gov/firemap/).

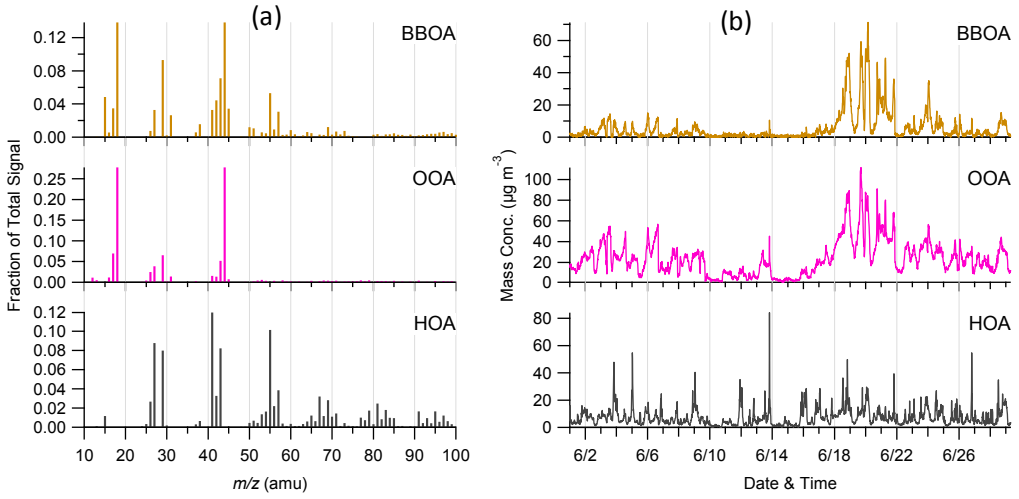

Fig. R2. (a) Mass spectra and (b) time series of three organic aerosol factors in June 2012.

(4) Page 14558, line 8-9: What are the definitions of moderately or heavy polluted days herein? Is it based on daily average, or day-time average, periodical spikes (e.g., plume) or consistent high NR-PM1 loadings? Figure 4 needs clarification about how the data is treated. High frequency of data points does not necessarily represent polluted days.

We thank the reviewer's comments. There was no exact definition for moderately and heavily polluted days. They were used for qualitative purpose which is 30 – 60 μg m^{-3} for moderately polluted days and > 150 μg m^{-3} for heavily polluted days in this study. According to the Chinese National Ambient Air Quality Standard, it is defined as heavy pollution when the daily-average $PM_{2.5}$ concentration is above 250 μg m^{-3} (Air quality index = 300). Assuming that NR-PM_1 is approximately 60 % of $PM_{2.5}$, the NR-PM_1 concentration during heavily polluted days is expected to be ~150 μg m^{-3}. The frequency in Fig. 4 was calculated with 15 min average data. The major purpose of Fig. 4 is to investigate the frequency of different NR-PM_1 mass loadings during the four seasons, particularly the highly polluted events. This has important implications for health studies. Using daily average data could significantly underestimate the exposure of people in the time with high PM levels.

We agree with the reviewer that it's not accurate to use highly time-resolved data to describe "days". We revised this paragraph by changing "days" to "events", and also deleted "moderately polluted days" because of no accurate definition.

(5) Figures 3 and 4 show duplicate information. I suggest combining them into one figure.

We thank the reviewer's suggestion. Fig. 3 shows the time series of aerosol species and average chemical composition during four seasons, while Fig. 4 presents the frequency of NR-PM_1 mass loading for each season. We kept these two figures considering it is difficult to read when combining them into one figure.

(6) Page 14559, line 3-4 and page 14560, line 4-5: Similar to comment #3, the authors should provide evidence to support the conclusion that "due to the impacts of agricultural burning in these two months".

See our reply to comment #3.

(7) Page 14561, line 6: Is the particle-phase ammonium sufficient to neutralize inorganic species? It may be better to show the ion balance information to support the forms of inorganic species.

We checked the ammonium balance in winter by comparing the measured NH_4^+ with that requires to fully neutralize sulfate, nitrate, and chloride (Zhang et al., 2007). As shown in Fig. R3, aerosol particles in winter were overall neutralized ($NH_4^+{}_{predicted}$ /$NH_4^+{}_{measured}$ = 0.93) although periods with slight acid particles were also observed. Here we mainly address the effects of temperature on gas-particle partitioning of ammonium chloride in winter compared to other seasons, therefore we didn't show ion balance information in the manuscript.

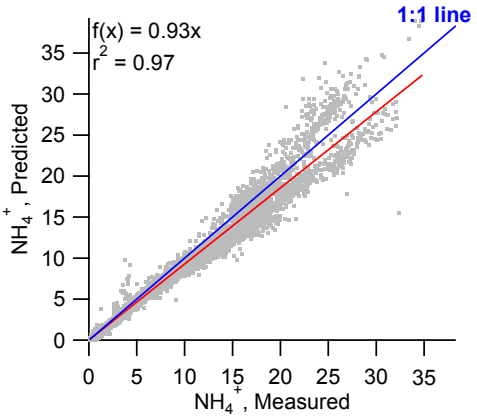

Fig. R3. Scatter plot of NH_4^+ predicted (= $2\times18/96\times SO_4^{2-} + 18/62\times NO_3^- + 18/35.5\times Cl^-$) vs. NH_4^+ measured in winter.

(8) Section 3.4: I am not convinced that the data for weekdays and weekend may really suggest anything about the emission strength without good constraints on meteorological conditions, atmospheric processing, and transport, life styles and so on in Beijing.

We agree with the reviewer that weekend effects can be affected by multiple factors including meteorological conditions, atmospheric processing, and even transport. However, for most of time, the meteorology conditions (temperature, relative humidity, and winds) and solar radiation won't change significantly in a week except special events, the comparisons between weekdays and weekends therefore could reflect, to some extent, the source emissions, e.g., primary aerosols, and also photochemical production of secondary species. In fact, such an approach has been widely used in many previous studies (Forster and Solomon, 2003; Pollack et al., 2012; Warneke et al., 2013; Wang et al., 2014). In this study, to better evaluate the weekend effects, the periods with low aerosol loadings (NR-PM_1 < 20 μg m^{-3}) that were usually from different source areas compared to high mass loading periods, were excluded in the analysis, which can reduce the effects of meteorological conditions to a certain degree.

A more detailed evaluation of weekend effects needs to involve modeling work which is beyond the scope of this study.

Technical remarks: In-text citations should be displayed in a proper format, for example, should be "Sun et al., 2013a" instead of "Sun et al., 2013b" in page 14552, line 5 and

"Zhang et al. (2013)" instead of "R. Zhang et al. (2013)" in line 27. First-name initials appeared in many other places, which should not.

The initials before some references in the text serve to distinguish this reference from other references with the same publication year, but a different first author with the same last name. Such a format is suggested by ACP.

Page 14553, line 6: Please provide references after "entire season".

The references Zhang et al. (2013b) and Zhang et al. (2013a) were provided.

Page 14553, line 15 and later text: The word "organics" doesn't exist in dictionary. Replace with "organic material" or "organic species".

Thanks the reviewer's suggestion. Considering "organics" is a common word that is widely used in the community, we keep it in the manuscript to be consistent with previous studies.

Page 14556, line 20: Metals are also refractory.

Yes, we revised this sentence as: "the ACSM cannot detect refractory black carbon, mineral dust, and metals."

Page 14558, line 2: Add "compared to other places in China" after "in Beijing".

Added.

Page 14560, line 25-26: Define "POA" and "SOA".

"POA" and "SOA" was defined in page 14553, line 17.

Page 14563, line 4: Replace "_ 8:00 until _19:00" by "about 8:00 to 19:00".

Replaced.

Response to reviewer #2

This manuscript reports results obtained with an Aerodyne Aerosol Chemical Speciation Monitor (ACSM) during a long-term measurement period (1 year) at Beijing, China. The authors describe the seasonal variations of non-refractory submicron particles (NR-PM1). Then, they discuss the properties of NR-PM1 as a function of the meteorological conditions and the air mass origin. This manuscript is very interesting and of prime importance, and is completely within the scope of Atmospheric Chemistry and Physics. Therefore, I highly recommend its publication after the authors address the following comments.

We are thankful to the reviewer for his/her positive comments on this manuscript.

Specific comments:

Page 14555, lines 5-20: when I read the calibration procedure of the ACSM in this paragraph and in the two first papers published by the group on this study (Sun et al., 2012; Sun et al., 2013), it seems that the RIE sulfate has not been calibrated with $(NH_4)_2SO_4$. This is important, because the RIE sulfate may vary a lot (up to a factor of 3) between different instruments. Therefore, if the default value has been used, there can be an important uncertainty on the accuracy of the sulfate concentrations. The authors can try to use the parameterization presented in Budisulistiorini et al. (2014) to estimate the RIE sulfate, and see if there is a high deviation from the default value. If it is the case, it can have an incidence on the NO_3^-/SO_4^{2-} ratio discussed later by the authors (page 14559, lines 14-26).

The reviewer is right. We didn't calibrate ACSM with $(NH_4)_2SO_4$ in this study because such an approach was only proposed recently. Following the reviewer's suggestions, we checked the RIE_{SO4} using the method presented in Budisulistiorini et al. (2014). The slopes of predicted SO_4^{2-} versus measured SO_4^{2-} varied from 0.95 to 1.41 during four seasons. As a result, the RIE of SO_4^{2-} varied from 1.1 to 1.6, leading to an uncertainty of 5% - 35% for sulfate quantification. Such an uncertainty is overall consistent with that (28%) from a recent comprehensive evaluation of ACSM measurements by Crenn et al. (2015). It should be noted that the approach suggested by Budisulistiorini et al. (2014) assumes that aerosol particles are fully neutralized. Considering that aerosol particle acidity in Beijing varied largely between different seasons (Liu, 2012), such a approach might introduce additional uncertainties for sulfate quantification. Therefore, we kept the default RIE of SO_4 in this study. Nevertheless, we expanded the discussions on RIE_{SO4} in the experimental section 2.2, which helps readers know the uncertainties in sulfate quantification in this study.

Page 14561, lines 14-18: the authors mention that in winter, the sulfate concentration increased because of a significant increase of precursor SO_2, which can be oxidized to form sulfate via either gas-phase oxidation or aqueous-phase processing. However, the RH was very low in winter (< 40%), compared to the other seasons, so maybe the aqueous-phase processing had a very limited influence on the sulfate formation in winter.

Thank the reviewer for pointing this out. Although the average RH for the entire winter was low, there were many episodes with high RH levels (> 60%) including fog events during wintertime, particularly when air masses were from the south (Sun et al., 2013b). SO_2 with high concentration can be rapidly oxidized to form sulfate via aqueous-phase processing (Sun et al., 2013a).

Page 14562, lines 8-19: there are a few discrepancies between the information given in this paragraph and PMF results presented in the two previous publications of the authors on this study. For instance, the authors mention that the noon peak of organics was primarily caused by cooking emissions, while the evening peak was driven by different primary emissions (cooking, traffic, and coal combustion emissions) among the different seasons. However, according to Sun et al. (2012), only OOA and HOA factors were identified in summer, no cooking or coal combustion related emissions. It is only in winter that the authors identified cooking and coal combustion organic aerosols (Sun et al., 2013). Obviously, the HOA factor identified in summer was a mixture of HOA and COA (Sun et al., 2012), but if the noon peaks, which were more significant in summer than in the other seasons, were mainly driven by cooking activities, a COA factor would certainly be easily identified in summer.

They were consistent. In summer, PMF analysis of the unit mass resolution (UMR) spectra cannot separate cooking aerosol from traffic-related HOA. This was similar to a study in summer 2006 in Beijing by a Quadrupole Aerosol Mass Spectrometer (Sun et al., 2010). The COA and traffic-related HOA were also not separated by PMF analysis of UMR spectra. The reasons for this might be either due to the similar spectra of COA and HOA that cannot be easily separated by PMF analysis of UMR spectra, or COA and HOA in summer are well mixed due to high temperature and turbulence. However, COA and HOA can be separated by PMF analysis of high resolution mass spectra of OA due to richer chemical information from ion fragments (Huang et al., 2010). Although COA is more significant in summer, it's unlikely to be resolved as an individual factor if UMR spectra are used for PMF analysis. Therefore, HOA in this case would be a mixture of traffic and cooking aerosols.

Coal combustion OA (CCOA) was only significant during the heating season. Indeed, CCOA was never resolved in summer in Beijing, even for the PMF analysis of high resolution mass spectra of OA (Huang et al., 2010), indicating that coal combustion is a minor source of OA in summer. In contrast, coal combustion OA can be easily resolved during wintertime due to substantial emissions for domestic heating (Sun et al., 2013b; Sun et al., 2014; Zhang et al., 2014).

Moreover, I am not sure to understand why cooking activities are reduced in winter (also mentioned on page 14565, lines 8-9). Normally, cooking activities should be more or less the same during the different seasons.

The largest difference in cooking activities between summer and winter is charbroiling. While charbroiling is popular at both noon and evening time in summer, it is significantly reduced in winter due to the low temperature outside.

Page 14563, lines 24-25: how did the authors determine that chloride was present under the form of ammonium chloride? In Sun et al. (2012), the authors mention that chloride measured by the ACSM is primarily ammonium chloride, since the ACSM does not detect sea salt. However, non refractory chlorides include also other compounds, such as HCl and organic chlorides. It should be very difficult to determine this point with the scatter plot of NH4 measured vs NH4 predicted, since the contribution of chloride to the total inorganic species is small compared to sulfate and nitrate.

Chloride mainly exists in the form of NH_4Cl because ACSM is insensitive to refractory NaCl and/or KCl at its vaporizer temperature of 600°C. There's possibility that chloride was also partly from HCl and organic chlorides. Organic chlorides could not make a large contribution to the total chloride since no organic chlorides were reported in Beijing aerosols to date.

ACSM detects particle phase chloride, whereas HCl is volatile, which has an equilibrium with NH_4Cl ($NH_4Cl \leftrightarrow NH_3 + HCl$). The chloride showed pronounced diurnal patterns with low concentration in the afternoon due to the evaporative loss of particle phase NH_4Cl to gas phase HCl. Therefore, chloride detected by the ACSM would be primarily ammonium chloride. We agree with the reviewer that it is difficult to distinguish the forms of chloride using ammonium balance plot.

We revised this sentence as "Chloride in this study was primarily detected as ammonium chloride because ACSM is insensitive to refractory NaCl and/or KCl at its vaporizer temperature of 600°C"

Technical corrections:

5) Page 14571, line 7: "in the formation of serve PM pollution".

Corrected.

6) Figure 12: the text "(c) NO_3^- " seems to be opaque, since it hides the label of the x-axis just on the top of it. Please make it transparent, or move it a little bit to the bottom.

Corrected.

Response to reviewer #3

Recommendation: Reject Reason: It has been stated clearly in "Aims and scope" of this journal that "The journal scope is focused on studies with general implications for atmospheric science rather than investigations that are primarily of local or technical interest." Apparently, this manuscript is a local interest in a single site of China.

We conducted the first long-term, highly time-resolved measurement of fine particle composition in Beijing, China. The formation mechanisms and evolution processes in driving the seasonal variations and diurnal cycles of aerosol species have general implications for atmospheric science. The impact of meteorological conditions (e.g., relative humidity and temperature) on aerosol formation mechanisms also has general implications for atmospheric community. In addition, the long-term data this study serves as an important contribution to modelers for evaluating regional and even global models. Also note that the air pollution in Beijing has attracted worldwide attention since the 2008 Olympic Games. Therefore we disagree with the reviewer that this manuscript is only a local interest, and we believe that our study fits well within the scope of ACP.

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