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

Response to referee#1

The manuscript by Sun et al. had a detailed characterization of composition, sources and processes of submicron aerosols during wintertime in Beijing with an aerosol chemical speciation monitor and various collocated instruments. Positive matrix factorization was performed to investigate the sources and composition of organic aerosol. In addition, the effects of meteorology on PM pollution were also explored. Some results in this study are interesting and important to improve our understanding the air pollution in summer and winter in Beijing. For example, the authors found that coal combustion organic aerosol plays a major role in leading to PM pollution during wintertime. Sulfate however is more important during high relative humidity periods because of aqueous-phase processing. This manuscript is well written and the topic itself is appropriate for ACP. Thus, I recommend publication of the manuscript after some minor revisions.

Comments:
1. P 2079, line 24: spell "OA" out when it shows up for the first time

"OA" was spelled out.

2. P2080, line 25: add related references on receptor models

The related references of Zheng et al. (2005) and Song et al. (2006) on CMB and PMF receptor models, respectively, were added.

3. P2089, line 15: remove "a.m." and "p.m."

"a.m." and "p.m." were removed.

4. P2090, line 8: the authors concluded that the larger evening peak of COA during wintertime is due to enhanced cooking activities, which might not be the fact. The cooking activities at night between summer and winter should not be quite different. Instead, the enhanced COA emissions because of low temperatures might be the major reason.

We agree with the reviewer's comments and revised the explanation accordingly. Now, it reads "The diurnal cycle of COA is characterized by two pronounced peaks corresponding to lunch and dinner times. Similar diurnal behavior of COA was also observed at PKU site in Beijing in 2008 (Huang et al., 2010). Note, at the same site of IAP, the COA shows larger differences between noon and evening peak in winter than summer. Given that cooking activities are expected to be similar in the two seasons, the enhanced COA concentration at night during wintertime might be mainly due to the low temperature, which facilitates gas-particle partitioning of semi-volatile OA species, and hence enhance the COA emissions."

5. P2095, line 16: "increasing mass" to "mass increase"

"increasing mass" was changed to "mass increase"

Response to referee#1

This paper by Sun et al. reported a wintertime study in Beijing by an aerosol chemical speciation monitor. The authors had a thorough analysis of the chemical composition, diurnal variations, day-night variations, and sources and processes of organics aerosols, and then compared with the results obtained in summer. Very different composition and diurnal variations of aerosol species between summer and winter were observed. Also the roles of aerosol species in PM pollution are quite different between summer and winter. Compared to the dominance of secondary organic aerosol (OA) in summer, primary organic aerosol (OA) dominated OA with coal combustion OA contributing the major fraction during wintertime. The meteorology plays an important role in PM pollution. In particular, fog processing to form secondary inorganic products during high RH periods is important. The paper is quite clearly written and was easy to review. This paper could be published on ACP after addressing the issues below.

Detailed comments:

1. The coal combustion organic aerosol (CCOA) is interesting. The identification of this factor is important. However, the authors inferred this factor mainly based on the comparisons with biomass burning OA from previous studies. The mass spectra of coal combustion OA and biomass burning OA might be quite different, so I am wondering if the authors can provide more evidence, like mass spectra of CCOA from Chinese coal stoves.

We agree with the reviewer that the mass spectra of BBOA and CCOA might be different. In fact, a recent study by Wang et al. (2013) reported the mass spectra of OA from pulverized coal combustion. As expected, the mass spectrum of CCOA in this study shows more similarity to that of OA from pulverized coal combustion under certain oxygen/carbon ratios in comparison to standard BBOA spectrum (Ng et al., 2011a). Also consistent with our results, low *m/z* 60 and 73 (AMS markers for biomass burning) were observed in OA spectra of pulverized coal combustion. Additional GC/MS analysis of fine particulate matter from the coal combustor even did not detect the biomass burning marker – levoglucosan (Wang et al., 2013), further supporting the low *m/z* 60 and 73 in the OA spectra. Therefore, the recent study by Wang et al. (2013) , which was included in the discussions in the revised manuscript, provides strong evidence for the presence of CCOA during wintertime in Beijing.

2. The authors use NO_3^- / SO_4^{2-} to explore the RH effects on formation mechanisms. I note that the nitrate concentration is often ~twice higher than sulfate in both summer and winter, which is not typical in Beijing. Although ACSM shows good agreements with TEOM measurements, I am still interested if the authors have other measurements, like filter packs, PILS etc. to compare with ACSM NO_3^- and SO_4^{2-} measurements.

We thank the reviewer's suggestions. Unfortunately, we currently don't have such collocated species measurements for inter-comparison. Nevertheless, an inter-comparison between ACSM and PILS-IC measurements was reported in NYC filed intensive campaign in 2009 (Ng et al., 2011b). The aerosol species (e.g., sulfate, nitrate, ammonium, and chloride) measured by ACSM agreed well with those measured by PILS-IC system.

Most previous works on characterization of aerosol particle composition in Beijing are based on filter measurements. Recent studies have found significant evaporative loss of semi-volatile ammonium nitrate particles during filter sampling, particularly in summer (Ianniello et al., 2011;Dong et al., 2012), whereas such effect is minor for real time on-line measurements. This might be one of the major reasons that we observed higher concentration of nitrate than previous studies.

3. In the abstract and the text, the author concluded that high concentration of sulfate under high RH condition is due to fog processing, what is the mechanism? With respect to the aqueous phase formation of sulfate, there are many literature reporting the formation mechanism, could the authors give more detailed mechanism explanation, which is very helpful for readers to understand the current heavy PM episode in Beijing.

The aqueous-phase oxidation of SO_2 to form sulfate mainly include three mechanisms, i.e., oxidized by 1) H_2O_2, 2) O_3, and 3) O_2 catalyzed by Fe/Mn (Seinfeld and Pandis, 2006). The oxidation rates depend on the droplet pH and the concentration of oxidants. While H_2O_2 is found to be the most important oxidant at low pH (e.g., $pH < 4$), O_3 plays more important roles in oxidation of SO_2 at high pH (e.g., $pH > 6$). The aqueous oxidation of SO_2 during fog periods can be very fast, especially when pH is above 6 (Reilly et al., 2001;Kaul et al., 2011). Although we don't know the mechanism dominating SO_2 oxidation because of the absence of fog pH measurements in this study, the rapid oxidation of SO_2 associated with fog processing are expected, which is supported by the reduction of SO_2 during fog events. Similar rapid fog processing of SO_2 was also observed previously in North China Plain (Zhang and Tie, 2011)

Following the reviewer's suggestion, we expanded the discussions on the formation mechanisms of aqueous-phase processing of SO_2 in section 3.6. Now it reads: "Aqueous-phase oxidation of SO_2 can be very fast, and the oxidation rates depend on the fog droplet pH and oxidants (e.g., H_2O_2, O_3, and O_2/Fe/Mn) (Reilly et al., 2001;Seinfeld and Pandis, 2006). While H_2O_2 is the most important oxidant at low pH (e.g., $pH < 4$), O_3 plays more important roles in oxidation of SO_2 at high pH (e.g., $pH > 6$) (Shen et al., 2012). The rapid fog processing of SO_2 was also observed in the North China Plain (Zhang and Tie, 2011), however, the dominant oxidation pathway is not known yet because of the absence of fog pH measurements. Despite this, our results suggest that aqueous-processing, mostly fog processing in this study, has played a significant role in the formation of sulfate."

4. I also found that the organic fraction in the current study was the major species in both summer and winter, accounting for around 50% of the total measured compounds, which is in *contrast to the case reported previously by Zhang et al 2007 in GRL (GRL 34, L13801, doi:13810.11029/12007GL029979, 2007), their results showed that organic matter is relatively small, accounting for <30% of the total mass. Could the authors give some explanation for such aerosol composition changes in Beijing during the past years? And what is its implication?*

The contribution of organic matter to NR-PM_1 shows large variations among different studies, depending on sites, sources, seasons, and meteorology. Zhang et al.(2007a) and Sun et al. (2010) reported that organic aerosol (OA) accounted for 35% of NR-PM_1 in summer, 2006 in Beijing. This contribution is close to 40% observed in summer, 2011 in our study (Sun et al., 2012), and also 38% in summer, 2008 (Huang et al., 2010). It should be noted that the results reported by Zhang et al. (2007) was based on two-week AMS measurements. Even during the two weeks, the contribution of OA to NR-PM_1 had large variations, for example, when the air mass was from the north, the OA contribution was up to ~70%. Similarly, Huang et al. (2010) also observed much higher OA contributions when the air masses were from the north and the northwest than those from the south. Therefore, our summer results are overall consistent with previous AMS studies in Beijing. In comparison to summer, winter shows much enhanced OA. This is mainly due to coal the combustion emissions during heating season. The enhanced contribution of organic matter in winter has been widely reported in Beijing (Dan et al., 2004;Duan et al., 2006;Zhang et al., 2007c;Lin et al., 2009;Yang et al., 2011).

Because most previous AMS studies in Beijing (from 2006 to 2011) were conducted in different days, sites, and seasons, it's hard to evaluate if the aerosol composition change is because of source emissions or meteorology. But we agree with the reviewer that it's important to know how the aerosol composition changes during the past few years and how much these changes are related to sources emissions. Currently, we are deploying an ACSM for long-term measurements of aerosol particle composition at the urban site of IAP in Beijing, and we expect to address the reviewer's question in a few years.

5. I noted that in this paper, and also in many other ACSM measurements, there is no data about bisulfate HSO_4^-, *I am not sure if this means that bisulfate is negligible and/or cannot be detected due to the instrument limitation? Since the author argued that aqueous-phase oxidation of* SO_2 *to produce* SO_4^{2-} *is important in Beijing especially in winter, one would expect that bisulfate is probably abundant in aerosol-phase.*

The aerosol mass spectrometer cannot distinguish bisulfate (HSO_4^-) from sulfate because the two species have very similar fragmentation ion patterns. Because the aerosol particles in this study are neutralized for most of time based on the comparisons between measured NH_4^+ and predicted NH_4^+ (= $SO_4^{2-}/96 \times 36 + NO_3^-/62 \times 18 + Chl/35.5 \times 18$ (Zhang et al., 2007b)), the concentration of bisulfate is expected to be very low. During the fog periods, the aerosol particles become acidic because of oxidation of SO_2 $NH_4^+{}_{meas.}/ NH_4^+{}_{pred.} < 1$). We expect some bisulfate in the aerosol particles, however, given that the lowest $NH_4^+{}_{meas.}/ NH_4^+{}_{pred.}$ is ~0.8, the concentration of bisulfate could not be significant in this study.

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