We greatly appreciate the reviewers for their valuable comments, which have helped us improve the paper significantly. We have addressed all of the comments carefully, as detailed below. The original comments are in black, and our responses are in blue.

Anonymous Referee #1

The authors presented field observation results of PM2.5 mass concentration, water soluble inorganic ions in PM2.5, OC/EC analysis of PM2.5, as well as reconstructed light extinction coefficient during haze events in January 2013 in the city of Suzhou, China. (1) Sources and processes leading to PM components were discussed with both back-trajectory analysis and the potential source contribution function (PSCF) method. (2) To investigate the contribution of PM species to visibility reduction, the authors attempted to link the two with the revised IMPROVE algorithm and the Koschmieder equation. (3) Finally, gas-to-particle conversion in secondary inorganic species (sulfate and nitrate) formation was briefly discussed.

The dataset itself is definitely interesting and worth exploring, and the methodologies employed by the authors were also scientifically sound. There are two reasons I don't see this version of the manuscript is publishable yet. First, for the three points summarized in the first paragraph, point (1) is just standard and should be in a short description only; point (2) is totally unnecessary since it is a two-step linkage from chemical composition to visibility (to me, it looks more like a number game if no measured extinction coefficient is shown); point (3) should be explored in greater detail, together with the distinct feature of the third haze event (30, January). This first reason will be elaborated point-by-point below in the major comments. Second, the language of the current form of the manuscript needs lots of work to enhance its readability. This second reason will be supplemented with some minor comments below (for those I have spotted). With these reasons, I suggest a major revision before the manuscript can be published in ACP.

We have made major revisions of the paper based on the reviewer's comments. We'd like to illustrate a few points here:

(1) The discussion on the backward trajectory analysis results has been simplified and improved for easy reading. We kept this discussion as one subsection due to the following considerations. Firstly, we think it's important to investigate the effects of the regional sources to the aerosol pollution in Suzhou as it is located in the typical monsoon region. Secondly, we think the results of back trajectory analysis and PSCF do not conflict with the assertion that secondary formation was responsible for high PM level. The impact of regional areas to the studied site is multifaceted. The regional transport might bring precursors or catalytic agents which could promote the secondary formation and/or the generation of PM instead of directly bring PM or secondary components to the studied site. Besides, the results of PSCF in the present study showed that Suzhou was mainly affected by local and nearby areas. Thirdly, the analysis helped the understanding of the distinct feature of the third haze event.

(2) In the revised manuscript, the reconstructed light extinction coefficients were compared

with those derived from visibility and calculated using another model. Strong correlations were observed, confirming that the reconstructed b_{ext} from IMPROVE algorithm was reliable. It's necessary to investigate the dominant contributors to the light extinction which is directly linked to the visibility. The contribution of chemical specie to light extinction might be different from that to aerosol.

(3) The discussions about the formation of secondary inorganic species and the distinct feature of the third haze event were strengthened in the revised manuscript. We have done more comprehensive analysis of the data to explore the SIA formation mechanism in the revised manuscript, such as the roles of liquid water content in aerosol, meteorological parameters and gaseous pollutants. The results suggested that the gas-phase homogeneous reaction and the heterogeneous process both responsible for the formation of nitrate and sulfate. The discussion of the distinct characteristics of the third haze was also strengthened by making comparisons with the other two haze events from more aspects such as $PM_{2.5}$ composition, light extinction coefficient, sources, etc.. The language has also been modified by professionals to enhance its readability.

Major:

1. Section 2. (a) The authors used data of SO2, CO, and NOx quite a bit later in the Results and discussion section. Measurement of these criteria pollutants should be mentioned here as part of the methodology. (b) A brief description on URG 9000 IC would be beneficial to readers since it is not as standard as TOEM. (c) A brief description on OC/EC analyzer and what method of OC/EC splitting was used (thermal OC/EC or optical OC/EC)?

The instruments used for the measurements of gaseous pollutants and meteorological parameters have been described in the revised manuscript. A brief description of instruments applied for PM, water soluble ions and OC/EC determinations has been provided in the revised manuscript.

2. Section 3.4. In this section, the authors tried to link the sources with both chemical composition and light extinction by back-trajectory analysis and PSCF. I don't see a great value of this sub-section for the following two reasons. First, if this analysis is useful, then the results just basically invalidated the authors early assertion that secondary formation was the dominant "source" for high PM levels observed (which I believe in). Second, the discussion of this sub-section is just too confusing and difficult to follow. I would suggest to talk generally about air mass origins in the general characteristics sub-section while not pushing too far to pin-point sources of those mainly secondary species.

As mentioned earlier, we think this discussion is helpful and kept this sub-section due to the following considerations. Firstly, we think it's important to investigate the effects of the regional sources to the aerosol pollution in Suzhou as it is located in the typical monsoon region. Secondly, we think the results of back trajectory analysis and PSCF do not conflict with the assertion that secondary formation was responsible for high PM level. The impact of regional areas to the studied site is multifaceted. The regional transport might bring

precursors or catalytic agents which could promote the secondary formation and/or the generation of PM instead of directly bring PM or secondary components to the studied site. Besides, the results of PSCF in the present study showed that Suzhou was mainly affected by local and nearby areas. Thirdly, the analysis helped the understanding of the distinct feature of the third haze event. For your second reason, the discussion on the backward trajectory analysis results has been simplified and improved for easy reading.

3. Sub-section 3.2.3. The authors used revised IMPROVE algorithm to reconstruct light extinction coefficient and used Koshmieder equation to "reconstruct" visibility. The discrepancy is large (a factor of two), owning (in my opinion) to this two-step linkage with both steps involving a number of assumptions and uncertainties. If there is no measured extinction coefficient to support, I do not see what value this analysis would add to the manuscript.

It's meaningful to investigate the contributions of $PM_{2.5}$ component to light extinction, because the results directly manifest the dominant contributor to visibility reduction, and have policy implications. Additionally, the contribution of chemical specie to light extinction might be different from that to aerosol loading. Although the discrepancy in light extinction between those reconstructed using IMPROVE and those derived from visibility is large, the results provide useful knowledge in improving our standing of the topic. Therefore, in the revised manuscript, we have made comprehensive comparisons of light extinction coefficients calculated from using different models, and with added uncertainty analysis. The results confirmed that the reconstructed b_{ext} from the IMPROVE algorithm was reliable to a large extent.

4. Section 3.3. The discussion of secondary inorganic species formation is informative but need some cautions. (a) correlation between RH and SOR does point to the importance of aqueous-phase formation of sulfate, but RH is an indication of gas-phase water after all. It is suggested that liquid water content in PM to be estimated using E-AIM or ISORROPIA.

We agree that to get the conclusion about the importance of aqueous-phase formation to sulfate only based on the correlation between RH and SOR might hasty. So in the revised manuscript, we estimated the liquid water content in PM by using E-AIM and conducted more detailed analysis to explore the formation mechanism of nitrate and sulfate.

(b) the authors cited Pathak et al., 2004, Pathak and Chan, 2005 to back the statement that homogeneous reaction between HNO3 and NH3 was important in nitrate formation. But these two papers talked about sampling artefacts for filter sampling when particles and gases can interact for 24 hours, while the authors used continuous measurement technique to measure SNA. I don't see that is relevant.

We agree with the reviewer that it's inappropriate. We cited other references (Pathak et al., 2009; Jansen et al., 2014; He et al., 2012) to back that statement in the revised manuscript.

(c) excess ammonium is of course one way to look at nitrate formation, but partitioning

equilibrium between NH4NO3 and HNO3 and NH3 is also important to considered given the low temperature and high RH in the studied period.

We explored nitrate formation mechanism from more aspects and conducted more detailed analysis. The results indicated that heterogeneous chemistry, such as equilibrium partitioning between NH_4NO_3 and HNO_3 and NH_3 (HNO_3 (g) + NH_3 (g) ↔ NH_4^+ (aq) + NO_3^- (aq)) also contributed to the formation of nitrate just as the reviewer expected.

5. The authors briefly mentioned the uniqueness of the third haze events compared to the first two in a number of places, but did not elaborate them. From the high POC and dominated air mass origins of C2 (short circulating trajectories), I believe this event was mainly contributed by primary emission from the local (surrounding) areas. It is suggested that this episode to be discussed in contrast to the other two with respect to primary/secondary fractions and meteorological parameters.

Thanks for the useful suggestion. According to the reviewer's suggestion, the discussion about the distinct feature of the third haze event has been strengthened in the revised manuscript. The characteristics of the third haze event has been compared with the other two haze events from various aspects, such as weather conditions, aerosol compositions, light extinction contributions oxidation ratio, air mass origination and etc. The results indeed indicated that the carbon components from the primary emission in local and/or nearby areas instead of secondary formation inorganic ions might be relatively more important for the visibility reduction for the third haze event.

Minor

P33409, L3: change "public" to "the public".

It has been revised accordingly.

P33409, L15: what is "artificial sampling"? should be "filter sampling"?

The phrase "artificial sampling" has been changed to "filter sampling": "Most existing studies were based on filter sampling and off-line analysis and had limitations in providing detailed insight into the roles the major chemical species played during shorter haze periods."

P33409, L26: change "close" to "similar".

It has been revised accordingly.

P33410, L3: change "salt" to "salts".

It has been revised accordingly.

P33410, L22: change "investigate the" to "investigation of".

The sentence has been revised.

P33411, L1: suffered should be suffered from.

It has been revised accordingly.

P33414, L20: change little to slightly (also in P33416, L6).

It has been revised accordingly.

P33417, L14-16: this sentence is not convincing to me. It is stated that there is difference between Suzhou and Beijing. But here it is asserted that nitrate formation may be also affected by re-volatilization of NH4NO3 as that in Beijing (similarity?).

There are both difference and similarity between Suzhou and Beijing. The NO_3^-/SO_4^{2-} ratio was higher under worst visibility conditions in Suzhou while higher under better visibility in Beijing. But similar to Beijing, the ratios of (NO_3^-/SO_4^{2-}) to (NO_X/SO_2) were lower for worse visibility period, which could indicate the effect of the re-volatilization of NH_4NO_3 on the nitrate concentrations. We have modified this paragraph hopefully to make it more understandable.

P33418, L16: change migh related to might be related.

It has been revised accordingly.

P33418, L28: change “similar profile” to a profile similar”.

It has been revised accordingly.

L33149, L3: I don’t agree that all the secondary aerosol species were “affected” by O3. O3 is just one of those oxidants that can oxidize precursors and lead to SIA formation. In fact, it is the secondary nature of O3 that makes its diurnal profile some similar to those of the secondary aerosol species.

We agree with the reviewer. We now rephrase this sentence: “The diurnal profiles of the secondary species were similar to their precursors but obviously affected by other factors such as solar radiation, which could promote the oxidation of the precursors.”

P33149, L11: also responsible should be also be responsible.

It has been revised accordingly.

L33419, L12: level should be levels.

It has been revised accordingly.

L33419, L19: “because of” should be “be due to”.

It has been revised accordingly.

P33419, L21: favored for should be favored.

It has been revised accordingly.

P33419, L22: always southwest wind? The discussion later for the first and second haze events suggests otherwise.

We assume that the later discussion the reviewer refers to is the back-trajectory analysis. We think there is no certain link between wind direction and back-trajectory results. Wind direction refers to instantaneous local weather condition. But back-trajectory results reflect the migration of air mass on a much larger temporal and spatial scale. So we don't think the results conflict with each other.

P33420, L2-7: I don't see it is necessary to repeat the equation here. In fact, I don't see it is necessary to have this analysis of reconstructed light extinction coefficient, as shown above.

We agree with the reviewer that it's unnecessary to repeat the equation. Therefore, the revised equation was not showed in the new version of manuscript. But we do think it's meaningful to have the discussion about light extinction for aforementioned reasons.

P33420, L14: "similar temporary trend" and "significantly correlated" are referring to the same thing.

We agree with the reviewer. The sentence has been rephrased in the revised manuscript: "Nevertheless, they were significantly correlated with each other ($r = 0.71$, $p < 0.001$).".

P33420, L16: were should be was.

It has been revised accordingly.

P33420, L26: were reduced should be reduced.

It has been revised accordingly.

P33421, L28: how come only NO2 (not NO + NO2) was used in calculation of NOR?

The calculation of NOR by NO_2 and NO_3^- has been used to estimate the secondary formation of nitrogen in many studies (Wang et al., 2016; Ji et al., 2014; Jansen et al., 2014; Zhao et al., 2013; Squizzato et al., 2013; Fu et al., 2008). The pathways for nitrate formation mainly include heterogeneous hydrolysis of N_2O_5 and gas-phase photochemical oxidation of NO_2. N_2O_5 is mainly produced from NO_2 ($NO_2 + O_3 \rightarrow NO_3 + O_2$, $NO_2 + NO_3 + M \leftrightarrow N_2O_5 + M$). So we think it's reasonable to only use NO_2 in the calculation of NOR.

P33422, L4-6: should be two sentences.

This sentence has been split into two sentences: "The daily variation of NOR showed similar pattern as NH_4^+ and NO_3^-. Likewise, SOR had similar diurnal change as SO_4^{2-}.".

P33422, L11-14: show the correlation.

The correlation coefficient has been provided in the revised manuscript.

P33422, L27: a variety of cities (change to different cities?); a little (change to slightly?)

The phrase "a variety of cities" should be "several" and "a little" has been changed to "slightly". The whole sentence now is "Pathak et al. (2009) also reported an intercept value of 1.5 for several cities worldwide while Jansen et al. (2014) found a slightly smaller intercept value of 1.38 for Hangzhou.".

P33424, L27: remove "differently".

It has been revised accordingly.

P33425, L13: suggest should be suggests.

It has been revised accordingly.

P33425, L14: but not should be but does not.

It has been revised accordingly.

P33426, L10: severe should be efficient.

It has been revised accordingly.

P33426, L18: were should be was.

There are many changes in this section in order to highlight the main conclusions of this article. This sentence is not included in the revised manuscript.

P33426, L24: But distinctively should be However.

There are many changes in this section in order to highlight the main conclusions of this article. This sentence is not included in the revised manuscript.

P33435: what are the shaded areas with different colors?

The shaded areas in orange refers to periods when visibility lower than 10 km and accompanied by precipitation. The shaded areas in grey refers to haze periods. We have clarified this in the revised manuscript.

P33437: it is difficult to be convinced that CO is a precursor of aerosol species; O3 might be one type of oxidants in secondary aerosol formation, but not a precursor.

We agree with the reviewer that it might be inappropriate to refer to these species as precursors. So the title of this figure has been rephrased: "Figure 6. Diurnal profiles of meteorological variables, aerosol precursors (NO_X, SO_2), CO, O_3, $PM_{2.5}$, and major aerosol compounds (NO_3^-, SO_4^{2-}, NH_4^+, EC, POC, SOC) under different visibility conditions.".

P33439: if it is a ratio, then show a ratio, not a percentage.

It has been revised accordingly in the revised manuscript.

Anonymous Referee #2

Manuscript on "Characteristics of aerosol pollution during heavy haze events in Suzhou, China" evaluates the atmospheric concentrations of PM, its chemical composition, and light extinction properties to understand the characteristics of haze events at Suzhou. Their results showed that haze events are characterized by the presence of enhanced concentration of secondary aerosol species and their precursors; further, abundance of OM, (NH4)2SO4, and NH4NO3 led to visibility impairment during the haze events. The study is useful towards understanding the role of pollutants in haze formation and visibility degradation and could help policy makers in specific control measures. This is an interesting piece of work, but in

my view, the manuscript needs to be strengthened in terms of scientific analysis and its novelty. Authors should highlight in the "Introduction" and in their Results and Discussion, the gaps in understanding (what is known and what we still need to understand through this work) related to aerosol pollution during haze events and how objectives and analyses presented in the manuscript for a specific location of Suzhou is different than reported information in literature for other locations influenced by haze events, e.g. Beijing etc. Also, there is a difficulty in the readability of the manuscript, this needs to be improved checking sentences throughout.

We have made significant revisions based on the reviewer's comments to strengthen the paper. The novelty, originality and importance of this study can be summarized below. (1) Suzhou, an important city in the YRD, with annual haze days increased from only two days to more than 150 days from 1956 to 2011, has experienced the extremely serious aerosol pollution in Jan. 2013, but little is known about the chemical characteristics and sources of fine particles in this city. This paper provide basic data for implementing effective pollution control measures in Suzhou. (2) The discussion in the present study was based on high time resolution data which could provide detailed insight into the role the major chemical species in $PM_{2.5}$ played during shorter haze periods. (3) The dominant species in $PM_{2.5}$ and those responsible for the visibility reduction were identified. (4) The formation mechanism of sulfate and nitrate were explored as high secondary aerosol contributions to particulate pollution during haze events. (5) The impact of local and outside sources on aerosol pollution in urban Suzhou was discussed. We now highlighted these points in the manuscript and we have also strengthened the comparison of the haze characteristics between Suzhou and other locations such as Beijing etc. The language has also been proofread by professionals to enhance its readability.

Specific comments:

Section 2.1: Please discuss uncertainty involved in measurements using each of the equipments.

The uncertainties have been discussed for the measurement of each equipment in the revised manuscript.

Section 2.2.1: It would be helpful to the reader if you can include information on basis of "numbers" (such as 2.2 x fs(RH) and so on, what does 2.2 indicate?) used in the equation 1 (IMPROVE algorithm).

The information of the numbers have been included in the revised manuscript: "The constant numbers in the above equation are extinction efficiencies for each chemical species under dry condition."

Section 2.2.2: please provide reason for 48 h back trajectories calculation; typically, taking into account lifetime of aerosols of the order of seven-days in the lower troposphere, should you perform 7-day back trajectory calculations?

48-h back trajectory analysis has been used in many studies (Yu et al., 2014; Ji et al., 2014; Behera and Balasubramanian, 2014; Zhang et al., 2013; Huang et al., 2012; Huang et al.,

2010; Wang et al., 2005). We think the back trajectory analysis is mainly used to explore the origination of the air mass in the studied area. The transport of air mass might bring aerosols and/or precursors to the target area and consequently influence the aerosol pollution. The results of this analysis basically could imply the impact of regional transport on the existing aerosol in the studied area. So we think the back trajectory analysis don't mean to calculate the lifetime trajectory of the aerosol.

Section 3.2.1: How does the chemical composition observed at Suzhou compares with that at Beijing or other places where haze events are frequent. Please discuss if sources of aerosol species are distinct between Suzhou and others.

The chemical composition observed at Suzhou has been compared with that at Beijing or other places in the manuscript, such as "These secondary inorganic components in total constitute 93% of total WSIIs, close to the result in Beijing (Gao et al., 2015; Tao et al., 2015)." and "The carbonaceous species, constituting 22% of $PM_{2.5}$, were dominated by organic carbon, which was 22.8 ± 10.6 mg m^{-3} and 3 to 29 times of that of elemental carbon (2.79 ± 2.58 mg m^{-3}), similar to those in Beijing (Tao et al., 2015).". Other results in present study have also been compared to other places, for instance, "The relatively high ratios of OC/EC (10.6 ± 4.29), which were higher than the ratios in Beijing (7.1 ± 0.5) and Jinan (7.15 ± 1.78)", "The ratio of NO_3^-/SO_4^{2-} in this study was lower than that in Beijing, but higher than those in Shanghai (0.43), Qingdao (0.35), Taiwan (0.20), and Guiyang (0.13) (Wang et al., 2006; Yao et al., 2002; Hu et al., 2002; Fang et al., 2002; Xiao and Liu, 2004).", etc.. We agree with the reviewer that to compare the sources of aerosol species between Suzhou and other places might be interesting. However, specific emission sources could not be identified only based on the existing data of species (water soluble ion, OC, EC) in $PM_{2.5}$. Some tracers of emission sources would be needed other than existing components.

Section 3.2.2: Please replace 'AM peak' and 'PM peak' by other relevant words. "It seemed that low visibility: : :southwest wind" please explain the possible reason for this.

The "AM peak" and "PM peak" have been replaced by "morning peak" and "afternoon peak". The possible explanation has been provided in the revised manuscript: "This might related to the topography. There are mountains located on the southwest which is not conducive to the diffusion of pollutants."

Section 3.2.3: "In order to appoint: : :" please change the word "appoint". Your analysis show that while OM is the largest contributor to light extinction, but its contribution to PM during haze events is lower than rest of the water soluble constituents. Please discuss the possible reason for this.

(1) The word "appoint" has been changed to "determine" in the revised manuscript: "In order to determine the contribution of $PM_{2.5}$ constituents to the visibility degradation, light extinction (b_{ext}) was reconstructed based on the revised IMPROVE algorithm." (2) There might be two possible reasons. Firstly, OM refers to organic matter which is derived from multiplying OC concentrations by a factor of 1.8 to account for unmeasured atoms. Secondly, the extinction efficiencies for each species under dry conditions are different. The extinction efficiencies of OM (2.8 for Small OM and 6.1 for Large OM) are higher than $(NH_4)_2SO_4$ (2.2

and 4.8) and NH_4NO_3 (2.4 and 5.1).

Section 3.4: Please provide insights on emission sources corresponding to paths of clusters and PSCF analysis. Please discuss why a set of clusters are found to be different than the rest.

We agree with the reviewer that discussing the emission sources (such as biomass burning, coal combustion, vehicle emission, etc.) corresponding to paths of clusters and PSCF analysis would be of great interest. However, specific emission sources could not be identified only based on the existing data of species (water soluble ion, OC, EC) in $PM_{2.5}$. Some tracers of emission sources would be needed other than existing data. Besides, the current paper is already very long and focuses on the chemical contribution, formation mechanism and regional impact. Introducing additional analysis of emission sources would weaken the overall focus, increase the length, and decrease the readability. Trajectories are calculated on basis of meteorological parameters such as wind direction and wind speed. Different meteorological parameters would result in different trajectory. The pollution characteristics (such as aerosol pollution level, chemical composition, light extinction, etc.) in clusters might differ from each other. There might be many reasons, for instance, the trajectories pass over different areas and/or the local emission sources varied.

Anonymous Referee #3

Reviewer's comments for "Characteristics of aerosol pollution during heavy haze events in Suzhou, China" by Tian et al. The manuscript by Tian et al. presented a field study for PM2.5 mass concentration, PM2.5 chemical composition, and associated gaseous precursors during haze events in a major Chinese city located in the Yangtze River Delta. Like many other studies in China, this work highlighted the large contributions of secondary aerosol species in PM2.5. Source regions were analyzed based on the back trajectory calculations.

The new perspective (in my opinion) this paper brought to us is the results on the main contributors of the light extinction in PM2.5 components. These results are interesting and potentially have policy implications, because light extinction is directly linked to the visibility, which is one of the major public concerns in China. In the current manuscript, this assessment was based on the measured concentrations of chemical species weighted by the mass-extinction coefficients derived from the revised IMPROVE algorithm. The authors identified OM, sulfate, and nitrate as the major contributors of visibility impairment. However, I do agree with reviewer #1 that such analysis was not convincing without appropriate comparisons with other measurements. I would suggest the authors to strengthen this analysis before this paper can be published in ACP, since it seems to be crucial for the main conclusions.

A possible way to do this can be including a more comprehensive comparison between the light extinction reconstructed from chemical species with that derived from the visibility. A previous study (Chen et al. ACP, 2012) in North China has shown that the visibility-derived ambient light extinction can be well reconstructed by an optical model with measured number-size distribution, hygroscopicity, and RH. For the revision, a scatter plot and/or a time series plot could be included for comparison of the light extinction coefficients derived from both methods. Uncertainties should also be included. In the case of poor agreement,

several hypotheses can be tested, including: 1) Coefficients developed based the IMPROVE data are not suitable for the aerosol populations in China, e.g., due to the differences in size distributions. In this case better parameterizations are needed for the haze over China. 2) The RH (or other key parameters) measurement may not be accurate; e.g., Fig. 2 shows severe haze events are associated with high RH (_90%), where the RH sensor may have a large error. In this case the assessment can be biased for contributions between hygroscopic inorganic species and hydrophobic OM. Some caveats should be discussed.

We have compared the light extinction coefficients reconstructed by IMPROVE algorithm with that calculated by the optical model as the reviewer suggested. Strong correlations were observed (R^2 = 0.952) with a slop of 0.837, confirming that the reconstructed b_{ext} from IMPROVE algorithm were basically reliable. We also discussed the possible reasons for the deviations between the results.

Another suggestion is that the authors can also present the contributions of different species to PM2.5 mass concentration in addition to light extinction (e.g., in Fig. 5 and 9, Table 1). This analysis will be based on measured variables and thus less ambiguous. Such results can be useful in the context of aerosol health effect, which is another major concern related to the air pollution.

Thanks for the valuable suggestion. The aerosol compositions in the three haze events have been illustrated in the pie chart in figure 3 in the revised manuscript.

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