Dear editors,

There is a response to reviews of our manuscript “Insights into a historic severe haze weather in Shanghai: synoptic situation, boundary layer and pollutants” (acp-2015-665). We thank very much for anonymous reviewers, and their suggestions are helpful for improving our manuscript. According to reviewer’s suggestions, we make revision to the manuscript in detail, all of revision have been marked in red in the new manuscript. The following is a point-to-point answer to comments.

For Referee 1

Question 1: The paper compared several controlling factors for their correlations with visibility. These factors include RH, PM, and particle number concentration at different sizes. However, these correlations were not based on the physical relationships. Directly computing the correlation coefficients for visibility (km) may not be appropriate, because the visibility values are generally low during hazy days and the correlation coefficient would be largely determined by the high visibility data. The authors could consider using the inverse of visibility as a proxy of extinction coefficient (at ambient RH) to do such analysis.

Answer: The extinction parameter can be converted to Vis using an equation such as from Stoelinga and Warner (1999) as Vis.=-ln(0,02)/Ext. (Stoelinga M.T. and Warner T.T. Nonhydrostatic, mesobeta-scale model simulations of cloud ceiling and visibility for an East Coast winter precipitation even, Journal of Applied Meteorology, 1999, 38: 385-404). However, in this paper, Vis. is used for a direct proxy of haze identification, influenced by various factors of particle mass (PM2.5), RH, particle number and sizes, particle chemistry, etc. We have analyzed the correlations between Vis. and these factors to only understand their possible linking or contributions to haze formation. In the next paper, we will make a deeper analysis of their relationships using data of Sca and Abs parameters measured by nephelometer, cavity ring-down spectroscopic instrument and aethalometer.

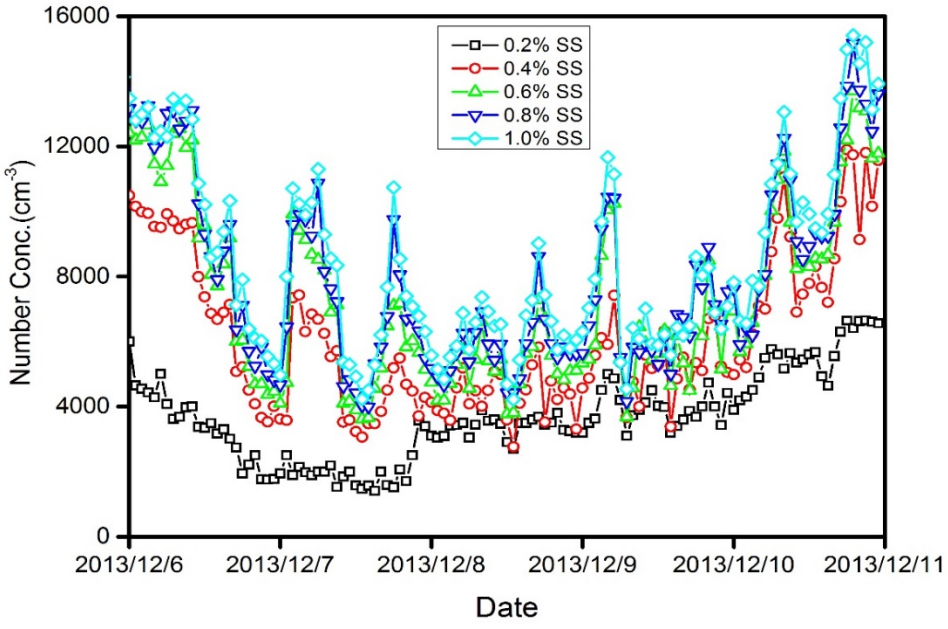
Question 2: The authors used the measured inorganic ions to calculate the hygroscopicity. The contribution of water-soluble organic carbon (WSOC) was entirely omitted in this analysis. WSOC can contribute 5%-40% percent of the water content for urban aerosols, depending on the particle size and composition. The value of kappa was therefore underestimated. In addition, the authors simply added sulfate, nitrate, and ammonium ions and assigned a kappa value of 0.6 (ammonium sulfate) to this group. This treatment was somewhat oversimplified. In some conditions, the bisulfate or sulfuric acid can present when the particles are acidic. The contribution of H+ to hygroscopicity would be omitted in this simplified treatment. Although this may not be the case for current study (because the concentration of nitrate is high, as an indication of excess ammonium), the method used in current manuscript can be very misleading for readers. The data should be analyzed in a more rigorous way, e.g., by considering ion balances or using a thermodynamic model.

Answer: It is notable that the contribution of water-soluble organic carbon (WSOC) shouldn’t be ignored in estimating values of hygroscopicity. In fact, the contribution (percent) of WSOC to hygroscopicity varies highly with time and region. In this paper, we simply considered sulfate, nitrate, and ammonium ions and assigned a kappa value of 0.6 (ammonium sulfate) to this group because of a lack of the available data of organic carbon during this period. The value of kappa

maybe somewhat underestimated. On the other hand, we mainly aimed to estimate the potential influence of RH to atmospheric visibility impairment by evaluating the contribution of inorganic ions to hygroscopicity. In the following paper, we will take more focuses on the impacts of particle chemistry and mixing or aging to hygroscopicity including a thermodynamic model. We have added some description about it in the revised manuscript.

Question 3: CCN data. For about half of the CCN data shown in Fig. 6, the CCN number concentration for 0.2% SS is higher than that for 0.4% SS.

Answer: It's really a fatal mistake that we neglected to select the valid CCN data of each supersaturation. The continuous-flow CCN counter (CCN-100) manufactured by Droplet Measurement Technologies (DMT, USA) was utilized to measure the CCN activation concentrations. Five supersaturations (e.g. 0.2, 0.4, 0.6, 0.8, and 1.0%) made up a cycle of half an hour, taking 10 min for 0.07% and 5 min for other supersaturations. Between the switch of different two supersaturations, it need a short period time to change into stable situation and the data should be abandoned. Therefore, that is we ignored before which is fatal to the result. We have corrected this mistake in the new figure 6. The figure shows as below.



(**Figure 6:** Time series of 1-h mean CCN concentration (NCCN) at supersaturations (SS) of 0.2-1.0% from 6 to 10 December.)

Question 4: Criteria of haze. In section 3.1.1, the authors state that " It has been widely accepted that the key criterion for discerning a haze event is to identify an apparent decrease of atmospheric visibility less than 10 km, and ambient relative humidity (RH) below 80% lasting for several hours (Fu et al., 2008; Du et al.,2011). When 80% < RH< 90 %, the event is referred to as a complex of haze-fog co-occurring or transition, and it is also classified into hazy episode in the present study (Leng et al., 2014a)."These definitions are vague and not widely accepted. A classification guideline for fog and haze using RH and visibility might be acceptable as an operational definition.

Answer: We agree with you that the definitions of criterions for discerning a haze event are vague

and not widely accepted. We just use these criterions to classify fog and haze events. In recent years in China, our government pay great attention to the environmental issues which are really thorny problems, and the document of haze pollution discriminant standard (trial) has been compiled by many researchers in China. Therefore, we use RH and visibility as an operational definition to discern haze events. Furthermore, we are inclined to consider other additional data (like PM2.5) to discern. We have added more descript in the new manuscript about it.

Minor comments:

Question 5: Abstract: the abstract should be revised. Please use short and clear sentences to improve the readability.

Answer: We have revised the abstract in the new manuscript.

Question 6: The language, in particular terminology, should be carefully checked throughout the whole manuscript to ensure that it is precise. Here are a few examples: …

Answer: We thank you for your good comments and suggestions, and have specified them in our revised manuscript.

Question 7: **Section 2.2**. Specify the principle of MAGRA. Are the gases measured by MAGRA also analyzed? Was the size distribution measured at dry condition? "LPS was calculated ..." How LPS was calculated? By Mie model? Or it is just a calibration using the PSL particles? “Without obscuration due to relatively lower aerosol loading and well mixed atmosphere". What does this sentence mean? Why lower aerosol loading can obscure the atmosphere? Specify the operation principle of PM monitors. Does the RH affect the measurements?

Answer: The principle of MARGA has added in the paper using red highlight. The data has been compared with other teams in our department before used, revealing the relative error was within 10% for SO_4^{2-} and NH_4^+. The hourly-averaged conc. of gases was from the Shanghai Environmental Monitoring Center (SEMC), which was measured by the precision instruments. For the WPS, of course, aerosol flow pass through a silica-gel Diffusion Drier before get into the WPS. In that case, the relative humidity can be controlled. According to the user manual, we learn the theory of WPS and the description of LPS theoretical response calculation is described as follow. "The LPS infers the particle size distribution from measurements of light scattering. It draws an aerosol into a sensing chamber, where the aerosol is illuminated with a beam of laser light. The light scattered by the aerosol is then collected by a photomultiplier tube. The amount of scattered light is the proportionally converted into a voltage. This voltage is then multiplied by an internal calibration factor to yield the particle size information. It should be noted that for light scattering-type aerosol instrument (e.g., LPS), the scattered light is dependent upon instrument properties (optical design, illumination source, etc) and particle properties (size, refractive index, shape, etc). The light scattered by individual spherical particles can be calculated using Mie theory (Hulst, 1981). "And well, we also used the PSL particles before and after the observation experiment. The operation principle of PM monitors have been displayed in the paper which is highlighted in red. The PM monitor (FH62C14) measures the relative humidity immediately upstream of the sample filter-tape assuring a representative measurement of the aerosol conditioning prior to real-time mass determination. So it is no need to worry that the RH affect the measurements.

Question 8: I would suggest to combine fig. 1 and fig. 2 and label the panels as a, b, and c. It is difficult to align low visibility and high RH in separate figures.

Answer: We have specified them in our revised manuscript. The figure shows as below.

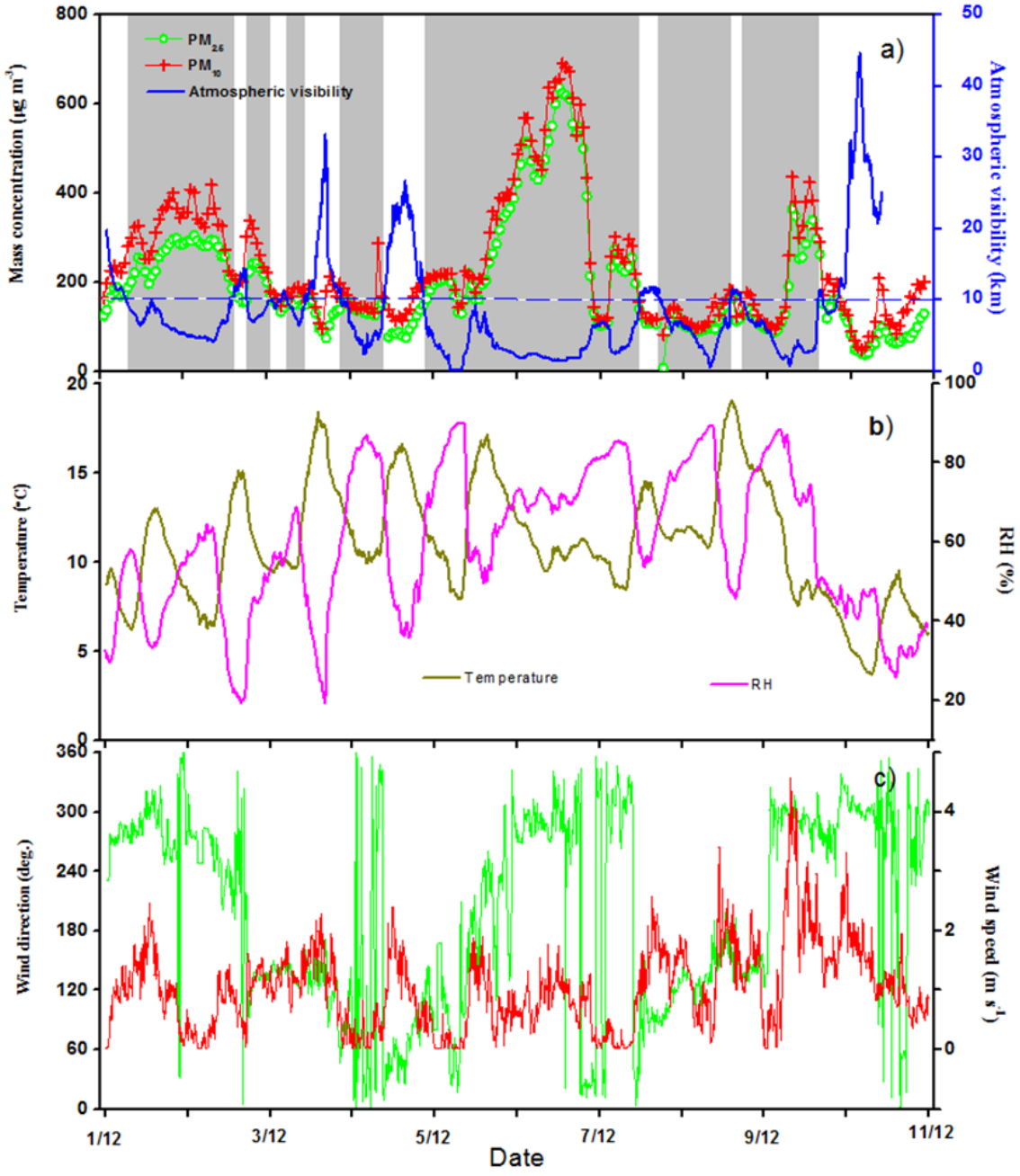


Figure 1. Temporal variations of (a) PM2.5, PM10, atmospheric visibility (Vis) and (b, c) meteorological parameters measured in Shanghai from 1 to 10 December 2013. The dash line is Vis at 10 km.

Question 9: It seems that the record-breaking haze event was regional rather than a local event, because the PM concentrations in other cities in the same region were also high. Consider moving the discussions for AOD maps closer to this paragraph and make this point clearer to readers. When did the PM2.5 measurements start in Shanghai? The 600 ug/m3 was a record since when?

Answer: We have moved the discussions for AOD maps (**section 3.2.3**) after this paragraph as **section 3.1.4** and have renew the order of following sections**.** We started our measurement using PM monitor at late November, but until the machines were steady and stable, we used the

data of PM. The record 600 ug/m3 was observed during several hours since the midday in 6th Dec.

Question 10: An implicit assumption of the absorption coefficient calculation is that an averaged mixing state of black carbon was used. This should be discussed. Specify the wavelength of data reported. (532 nm?) Also include the single-scattering albedo (SSA) in the analysis?

Answer: An implicit assumption of the absorption coefficient calculation is that an averaged mixing state of black carbon was used. These explain and the wavelength have been added in new **section 3.1.5**. We also added SSA in the new manuscript.

Question 11: P32575, L14 "suggesting that atmospheric oxidation of NO_2 and SO_2 contributed significantly to the formation of nitrate and sulfate". This sentence is confusing. Shouldn't nitrate and sulfate be entirely contributed by NO_2 and SO_2, respectively? Did the authors mean "suggesting that atmospheric oxidation of NO_2 and SO_2 contributed significantly to the formation of particulate matter"? The authors used nitrate/sulfate ratio to discuss contribution of motor vehicles. Nitrate concentration should also depend on the acidity. Abundance of ammonia can play a key role. As suggested in my previous comment, the inorganic species data should be analyzed in a more sensible way by analyzing ion balance or using a thermodynamic model.

Answer: We have specified them in new **section 3.1.7** in our revised manuscript.

Question 12: Include citations for the original data source of weather charts in Fig. 8 and 9. (KMA?)

Answer: We obtained the weather charts (Fig. 8 and 9) from the products of Regional atmospheric and oceanic short-term real-time forecasting system 9.0. One part of this system put the data of Korea meteorological Administration (KMA) on this system to analysis surface weather. We have also corrected the mistake "clod" into "cold".

Question 13: Would the mass concentration calculated by integration of size distribution consistent with measured PM2.5 and PM10? Consider the difference between aerodynamic diameter and mobility diameter in the calculation. This kind of closure calculation can be useful for validating data quality. "...no significant correlation was derived between atmospheric visibility and aerosol size of 10–600nm and 1.4–10 μm ". What does this sentence mean? Were the number concentration used in the regressions?

Answer: Before we started our monitor, the WPS had been calibrated according the user manual carefully. We also have compared with other teams to control the data's precision and validity. In new **section 3.2.5,**this sentence we wanted to say that the aerosols in these bins（10–600nm and 1.4–10 μm） have no significant correlations with visibility. Only those aerosols within the diameter interval from 0.6 to 1.4 μm show the expected reciprocal relationship. And the number concentrations used in the regressions display a normal distribution.

Question 14-16: in Figures, Please use discernible colors other than red and pink.

Answer: We have specified them in new figures in our revised manuscript. The figures show as below.

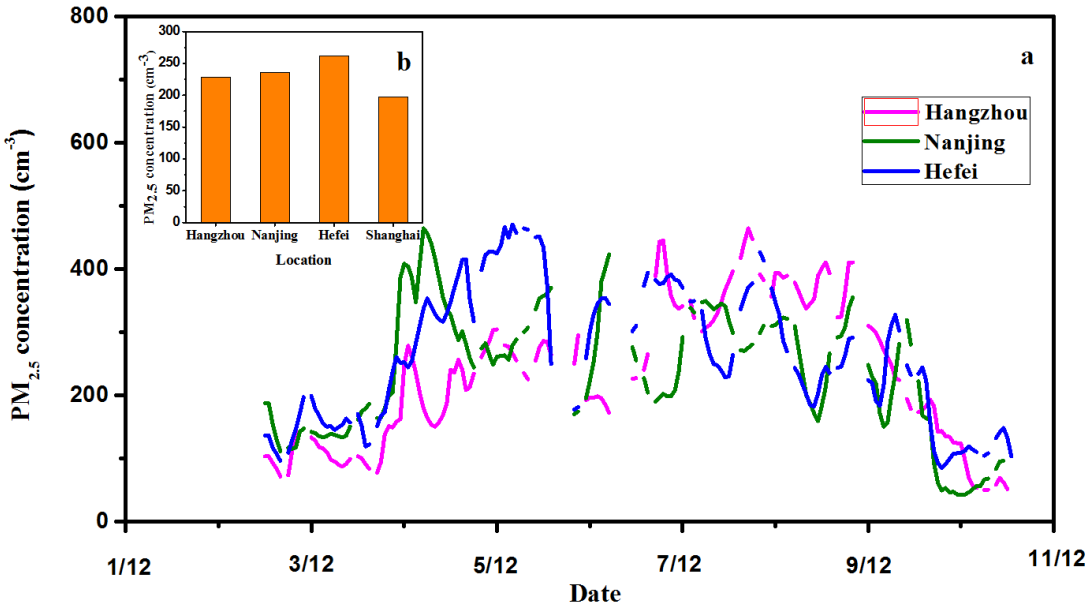


Figure 2. Temporal variations of $PM_{2.5}$ in Hangzhou, Nanjing and Hefei (a) and their mean concentrations (b) from 1 to 10 December.

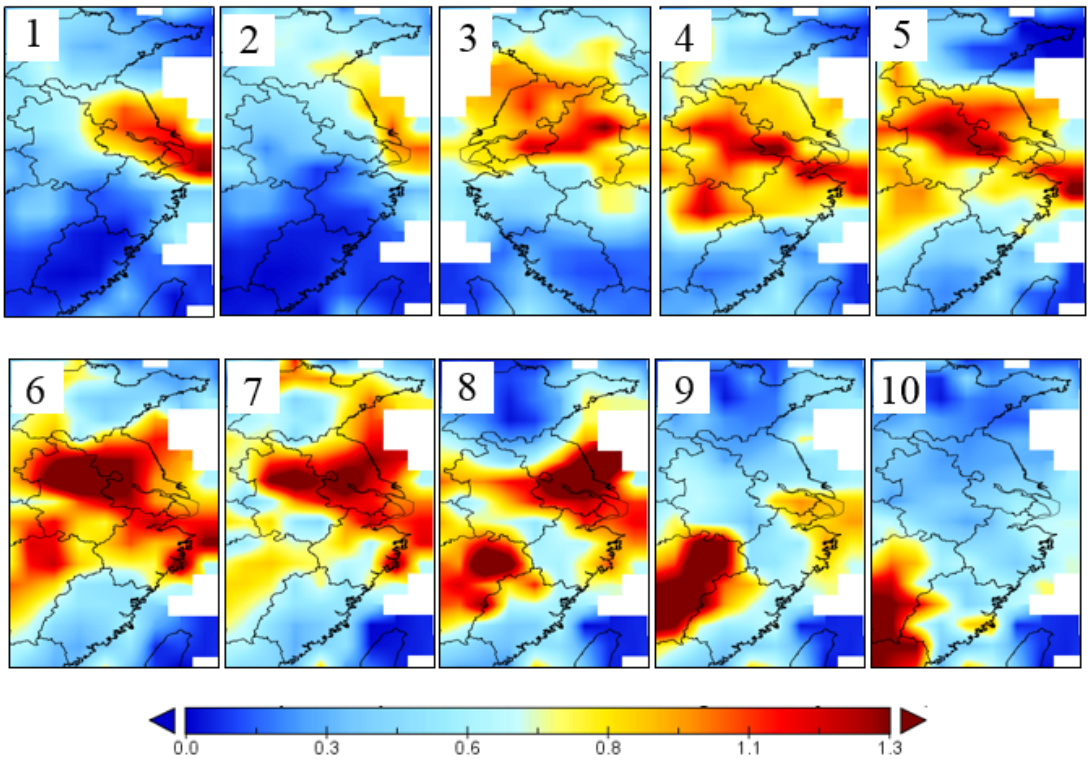


Figure 3. Aerosol optical depth (AOD) at 550 nm from MODIS over the YRD region at 6:00 (UTC) from 1 to 10 December (http://modis.gsfc.nasa.gov/).

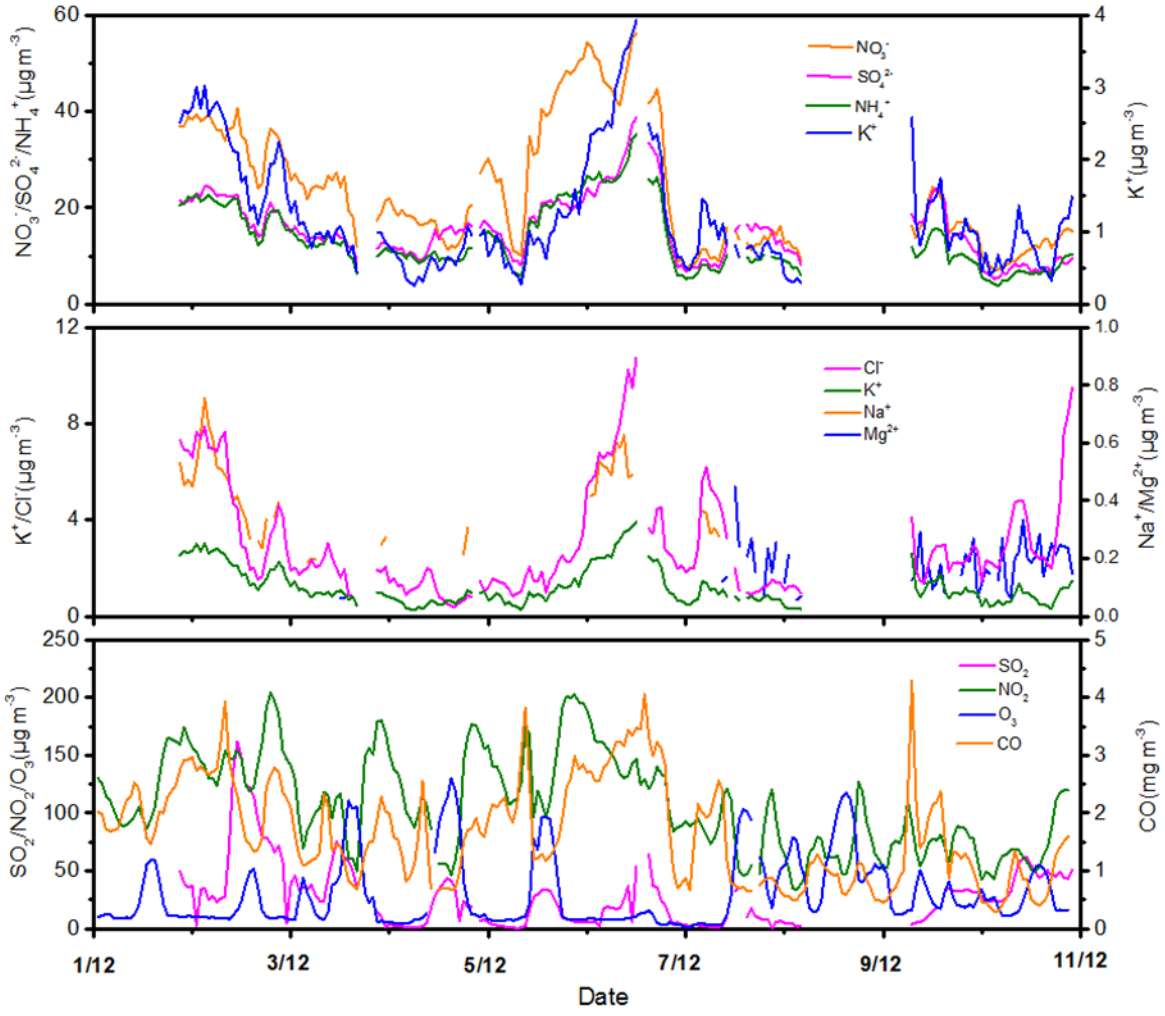


Figure 7: Temporal variations of chemical species in particles from 1 to 10 December.

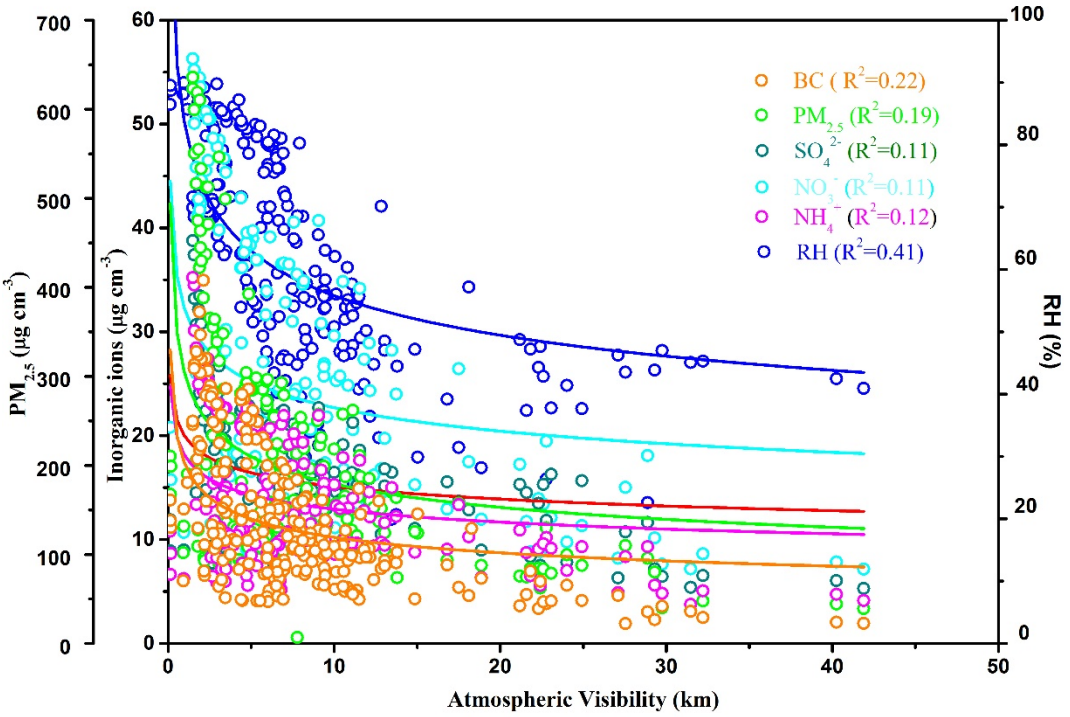


Figure 12: Scatter plots of RH, BC, PM2.5 and inorganic ions in particles versus atmospheric visibility.

Thanks very much!

2016/6/12