Dear Editor,

After carefully reading the reviewers' comments, we have revised our manuscript. All the responses are itemized below.

Anything about our paper, please feel free to contact me at wanggh@ieecas.cn

Sincerely Yours truly,

Gehui Wang

13/09/2014.

Anonymous Referee #1

General comments:

In the current work a semi-continuous observation on aerosol chemistry in Xi'an, a mega-city near Loess Plateau, was performed by characterizing the hourly collected TSP samples during a dust storm event. Moreover, size distributions of inorganic ions, WSOC and WSON in the dust storm period were also investigated. Hourly changes in aerosol compositions including chemical forms of nitric and sulfuric salts were explored. Kinetics of nitrate and sulfate formation during the dust storm event and the post-event were discussed. Finally, the authors proposed a mechanism for the nitrate formation on the dust surface. The results of this paper are quite interesting. Their findings on chemical forms of nitrate and sulfate in the dust particles and size distribution patterns of nitrate, sulfate and ammonium for first time revealed the infant state of atmospheric ageing process of East Asian dust in the area near the dust source regions, which is very helpful for researchers to improve their understanding on the full image of physicochemical evolution of Asian dust from the desert to the continental outflow region. The work was well designed. The organization of the paper is in good format, and related discussion is reasonable. Therefore, I believe this paper should be accepted by the journal after a minor revision. Following is the detailed comments.
Response: We thank the reviewer for the comments, and have carefully revised our paper. See the details below.

Detailed comments.
Comments:
1) Page 17442, line 3, of East Asian dust ageing process is better than "… of dust ageing process…".
Response: Suggestion taken. Please see the revised manuscript page 3, line 65.

Comments:
2) Page 17444, lines1-5, can author give the specific numbers of concentrations of sulfate and nitrate in 1997 and 2012, could author give a brief explanation why sulfate has sharply decreased in the city?
Response: Suggestion taken. We have added the concentration numbers into the text with a brief explanation. See page 5, line 117-119.

Comments:
3) Page 17444, line 4, gas not gases; lines 7, "…10-30% of the dust mass…", give the reference; line 8-9, should be HNO_3(g), which is more accurate.
Response: Suggestion taken. See page 5, line 123-124. We found the original statement "…10-30% of the dust mass…" , which we cited, is not correct. Calcite is the fourth abundant mineral in Chinese clay, but the content of calcite in east Asia dust is the most variable (McNaughton et al., 2009). From Liu et al (1985), we found that the content in East Asian dust should be 3.6-21%. Thus, we revised this sentence (see page 5, line 123).

Comments:
4) Page 17444, line 20-22, It's better to change as "We first investigated…. Then we identified...", which is consistent with the following statements.
Response: Suggestion taken. See page 5, line 134-136.

Comments:
5) Page 17444, line 25, delete the "dust".
Response: Suggestion taken. See page 6, line 139.

Comments:
6) Page 17445, what is the brand of the size-segregated sampler, I think this information is important.
Response: Suggestion taken. We have added the information into the text. See page 6, line 150.

Comments:
7) Page 17447, line 8-11, this sentence is a little bit of confusing to me. Please re-write.
Response: Suggestion taken. We re-wrote the sentence. See page 8, line 195-198.

Comments:
8) Page 17449, line 5, should be originated not originates; line 11, anthropogenic sources not species. Page 17451, line 4, NaCl, not $NaCl^-$.
Response: Suggestion taken. See page 10, lines 242, 248-249, and page 12, line 293.

Comments:
9) Page 17452, line 7, what is the meaning of the slope 0.28, if it means the 1:1 molar ratio of NH_4^+ to NO_3^-, i.e., the value of 0.28=18/64, please clarify.
Response: Yes. The slop of 0.28 is corresponding to the 1:1 molar ratio of NH_4^+/NO_3^-, because here we used the mass concentration. To avoid any potential confusing problem, we modified the statement as "nitrate and ammonium mass concentrations not only displayed the same size distribution patterns during the whole sampling period but also strongly linearly correlated each other during the dust storm and transition periods with a slope of 0.28 that is equal to the 1:1 molar ratio of NH_4^+ to NO_3^-….". See page 13, line 325-330.

Comments:
10) Figure 2b, the maximum of EC in the dust storm period is around 30μg m^{-3}, but in Table 1, the range is 0.0-3.2μg m^{-3}, is this number wrong?
Response: We corrected this mistake. It should be 0.0-32. See page 22, line 727 (Table 1).

Anonymous Referee #2

General comments

This study focused on aerosol chemistry in inland China and discussed the possible chemical mechanisms of nitrate and sulfate. The authors designed the research during one Asian dust storm. The hourly dust samples were collected to understand the chemical changes from early stage of dust storm to end. Through comparisons chemical compositions of different dust samples at different stages of dust storm and size resolved aerosol sampling, the authors obtained the chemical forms and formation mechanisms of nitrate and sulfate. The study is good presentation for the research results. However, the paper needs to be one minor revision.

Response: We thank the reviewer's comments, and have revised the paper. See the details below.

Comments:

1) 17441 L2 deleted in the current work./ Total suspended particulate (TSP) samples were

Response: Suggestion taken. See page 2, line 37.

Comments:

2) L17, biomass burning emitted Cl^-. The author should provide any evidence.

Response: Suggestion taken. There are many literatures showing that aerosols derived from biomass burning are enriched with Na^+, K^+ and Cl^-, which largely stay in fine particles(Andreae et al., 1998; Shen et al., 2008). In addition, K^+ in fine particles is often taken as a key tracer of biomass burning (Agarwal et al., 2010; Shen et al., 2008). We have added some references into the text to prove that biomass burning emission is an important source of fine particulate Cl^- in Xi'an city during our observation period. Related discussions are modified. See page 12-13, line 304-315.

Comments:

3) L12-21, the author need to consider why NH_4^+ and NO_3^- were mixed with mineral dust particles in coarse mode instead of the externally mixed with dust particles although the good linear from NH_4^+ and NO_3^-.

Response: As we have stated in the manuscript, Cl^-, SO_4^{2-} and NO_3^- are the major anions and Na^+, Ca^{2+} and NH_4^+ are the major cations in the TSP samples, among which Cl^- and SO_4^{2-} strongly linearly correlated with Na^+ and Ca^{2+} rather than NH_4^+. Instead, NH_4^+ only well correlated with NO_3^- and no correlation was found between NO_3^- and other cations. Moreover, linear fit regression showed that NH_4^+ and NO_3^- linearly correlated each other with a molar ratio of 1:1. The above results clearly indicate that NH_4^+ and NO_3^- were mixed with mineral dust particles as the chemical form of NH_4NO_3. Therefore, during the observation periods both ions displayed an identical temporal variation pattern and presented same size distribution patterns in the dust and non-dust periods. See the related discussions in page 10, line 254-257, page 13, line 325-330, and page 14-15, line 358-365.

Comments:

4) L23 $CaSO_4$ particles are minor hygroscopic materials. The Na_2SO_4, $CaSO_4$ and NaCl have high deliquescence relative humidity but during the dust period the relative humidity is quite low. My question is any evidence to prove these particles absorb water to form aqueous phase.

Response: We agree with the reviewer that $CaSO_4$ is minor hygroscopic, and Na_2SO_4 and NaCl have high deliquescence relative humidity points, which are 84.2±0.4% (RH) and 75.3±0.1% at 25^{o}C,

respectively (Tang, 1980; Tang, and Munkelwitz, 1993). Under these high RH conditions Na_2SO_4 and NaCl particles can deliquescence and become water droplets. However, this is not the case in the current work. Actually, in this paper we have talked about the formation of liquid phase on the dust surface, which can occur under very low RH conditions. For examples, a liquid phase formed on dust particle surface during Asian dust storm event has been reported by Li and Shao (2009). They used transmission electron microscopy (TEM) to observe mineral particles collected during a dust storm event and found visible coatings on the dust particle surface, which contain Na, Ca, O and N. During this dust storm event RH was 20-45%. Laboratory study by Goodman et al (2000) showed that even under RH<20% conditions a liquid phase can quickly form on the calcite surface via water vapor uptake of $Ca(NO_3)_2$ that is produced by heterogeneous reaction of $HNO_3(g)$ with $CaCO_3$. Andreae and Rosenfeld (2008) also pointed out that dust is insoluble but wettable. In the current work, we do not have any direct evidence to prove that these particles absorb water to form aqueous phase. However, we have measured the hygroscopicity of the ambient aerosols during the whole campaign by determining the hygroscopic growth factor of the water-soluble fraction of the TSP samples (Huang and Wang, 2014). The results showed that κ value of the water-soluble fraction of dust particles ranged from 0.20−0.38 (0.30±0.04) (Huang and Wang, 2014), indicating a wettable nature of the dust particles (Andreae and Rosenfeld, 2008).The above field observation and laboratory measurements demonstrated that a liquid phase can be formed on the dust surface under a low RH condition, which can be ascribed to the hygroscopic salts (e.g., NaCl and Na_2SO_4) in dust particles. We have added the above related discussions into the text. See page 15, line 368-372.

We agree with the reviewer that $CaSO_4$ is minor hygroscopic, thus we deleted $CaSO_4$ from the related discussions and modified our statement. See page 2, line 56.

Comments:

5) 17444. L27, hygroscopic salts include NaCl, Na_2SO4 not $CaSO_4$? I am worried about the $CaSO_4$ here.

Response: We removed $CaSO_4$ from the sentence and modified the statement. See page 6, line 141-142.

Comments:

6) 17448 L6, $PM_{2.5}$ mass concentration

Response: Suggestion taken. See page 9, line 218.

Comments:

7) 17449 L17-19, sulfate concentration, ammonium concentration

Response: Suggestion taken. See page 10, lines 254 and 255.

Comments:

8) 17451 L4 $NaCl^-$ should be NaCl

Response: We have corrected this mistake. See page 12, line 293.

Comments:

9) 17452, Na_2SO_4 and $CaSO_4$. Again my question is L23

Response: In the surface soil of north China like Inner Mongolia and Qinghai Province, Na_2SO_4 and $CaSO_4$ exits as the hydrated salts, i.e., mirabilite($Na_2SO_4 \bullet 10H_2O$) and gypsum ($CaSO_4 \bullet 2H_2O$), both co-exist together and are water-soluble (Zheng, 1991). These salts, along with NaCl, can absorb

water vapor, forming a liquid phase on the dust surface. Please see our response above.

Comments:
10) 17453, L25-26, need reference or evidence.
Response: For the evidence to prove that a liquid phase can be formed on the dust particle surface. See our response to the 2nd reviewer's above comment point (4) and page 15, line 368-372.

Reference:

Agarwal, S., S. G. Aggarwal, K. Okuzawa, and K. Kawamura: Size distributions of dicarboxylic acids, ketoacids, α-dicarbonyls, sugars, WSOC, OC, EC and inorganic ions in atmospheric particles over Northern Japan: implication for long-range transport of Siberian biomass burning and East Asian polluted aerosols, Atmos. Chem. Phys., 10(13), 5839-5858, 2010.

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Zheng, X.: Salt lakes on the Inner Mongolian plateau of China, Chinese Geographic Science, 1, 83-94, 1991.