Dear ACP Editor

After carefully read the comments from you and the two reviewers, we have revised the manuscript. Our response to the comments are itemized as follows

Anonymous Referee #1

General comments

Many papers have characterized atmospheric aerosol chemical compositions, however, very few of them have used multiple techniques like authors did in the current work. In this paper, authors have determined diacids, metals, EC, OC, and isotopic compositions of TN and TC in the PM10 samples from inland China, and further calculated the concentrations of POC, SOC, ON, and IN, etc. Based on the compositions of those secondary aerosols (i.e., diacdis, nitrate, sulfate), primary aerosols (i.e., metal elements and EC) and isotopic compositions, authors further dis-cussed the relations of those primary and secondary species, and have showed some insights into the source and secondary aerosol formation process. The paper is organized well, and the results and conclusions are of novelty. The data and related discussions are very helpful for readers to improve the understanding of the characteristics of Asian aerosols including compositions, sources, and formation processes. Therefore, I recommend an acceptance of this paper for publication in ACP. I found some mistakes in the current format, which should be corrected before publication.

Response

We thank the reviewer's comments, and made the revisions. See the details below.

Detailed comments

1. Page 6900, line 10-20, the discussions on pH are good. It is very useful for comparison of relative acidities samples and identification of the aerosol sources, although the pH values do not reflect the real acidity of the particles. The results presented here strongly demonstrated the importance of dust emissions in inland China. But I think it would be better for understanding the aerosol properties if authors measure the real acidity using some method like titration.

   Response

   We agree with the reviewer that measurement of the real acidity of the samples is very helpful for better understanding the aerosol properties. On one hand, after determination of pH using a titration method, the sample would be no longer useful for other measurements. On the other hand, the samples were assigned for a comprehensive analysis of lipids, diacids, inorganic ions, EC, OC, isotopic compositions and elements. There were no additional samples. Thus we cannot measure the real acidity of the samples by a titration technique in the current study. Instead, we determined the pH of the extracts using a pH meter, and then the extracts were used for ion measurement.

2. Page 6902, line 5, I would like to add one more sentence, i.e., inorganic nitrogen compounds (i.e, nitrate and ammonium) are the major nitrogen-containing matters in inland China, whereas organic nitrogen species are relatively minor.

Response
Suggestion taken. See page 8, line 181-183.

3. Page 6909, Table 1, the title, composition should be compositions. Through Table 1 to Table 4, the letter “n” denoting the sample numbers should be in regular format not the italic.
   Response
   We made the corrections.

4. Page, Table 2, glyoxylix is wrong, should be glyoxylic. The concentration numbers for Al, Ca and Fe in the 4th column should be presented with no space.
   Response
   We made the corrections.

5. Page 6917, Fig.4, the caption should be changed as Concentration ratios of C3 and C4 to C2.
   Response
   We made the corrections.

6. Page 6918, the figure caption, reference should also be Wang 2010, same thing for Fig. 7 caption in page 6920.
   Response
   We made the corrections.

Anonymous Referee #2

General comments

This paper is presenting a nice set of interesting data. However, the discussion is rather short for each part, and some of the conclusions are not supported by the data (cf rem for page 6903). I suggest some important revisions before publication in ACP.

Response

We thank the reviewer’s comments, which are very helpful for improving our paper quality. We made the related revisions according to the comments and also added more discussion. Detailed modifications are shown below.

Specific comments

1. Page 6898 line 10: “field blank”
   Response
   Suggestion taken.

2. Page 6899 line 4: more details are needed for the description of the protocol for water extraction that strongly influences the pH measurements.
   Response: Suggestion taken.
   We added the detailed procedures in the text. See the revised paper, page 5, line 103-106.

3. Page 6901 line 10: I seriously doubt that larger OC/EC ratios in suburban areas during both seasons can result from biogenic emissions. Does your organic measurement support such a hypothesis?

   Response

   There are much more plants in the suburban area than in the urban area. Our previous studies have showed that the airborne particulate biomarker like C_{31}-alkane at the suburban site is more abundant than at the urban sites with the carbon preference index (CPI, odd/even) of total *n*-alkanes in the suburban samples around 10% higher than in the urban samples (Wang et al., 2009; Xie et al., 2009). On the other hand, biomass burning for house heating and cooking is still common around the suburban site, which releases more OC compared to fossil fuel combustions. Therefore, OC/EC ratios are higher in the suburban area than in the urban area. Such a higher ratio caused by biomass burning was also observed for India aerosols (Ram and Sarin, 2010). The related discussion was revised with above points. See page 7, line 155-160

4. Page 6902 lines 14-17: how do you explain such a large difference of glyoxylic acid? Does this mean that glyoxylic acid is resulting from coal combustion?

   Response:

   Coal burning emission is an important source of benzene, toluene, ethene, and acetylene, which can be photochemically oxidized into glyoxylic and pyruvic acids (Huang et al., 2005; Warneck, 2000). Thus the higher concentration of glyoxylic acid in the samples may be ascribed to coal combustion in the region. The discussion was added into the text. See page 9, line 198-201.

5. Page 6902 lines 18-20 and figure 4: Does figure 4 include only data for the urban area? Are the ratios the same for the suburban area? Does it say something about the oxidation and sources?

   Response

   Yes, it includes urban and suburban data. We made a note in the figure caption. See page 17, line 435-437. The positive linear correlation between the ratios of C_3/C_2 and C_4/C_2 suggests that C_3 and C_4 may be the major precursors of C_2, both can be further oxidized into the smallest dicarboxylic acid (Wang and Kawamura, 2006). This statement was added. See page 9, line 202-204.

6. Page 6902, section on organic acids: there are no discussions on the other measurements presented in Table 2.

   Response

   Suggestion taken. We added one sentence into the text for discussion of other diacids. See page 8, line 192-195.

7. Page 6903 lines 15-16 (and also in conclusion): this is most probably plain wrong. Some part in the correlation is most probably due to covariations related to meteorological variations. Most important, coal burning is a large source of Ca and Fe.

   Response

We disagree with the opinion on covariations related to meteorological variations. If covariations related to meteorological variations resulted in the positively correlation of POC with others, then linear correlationship of primary components such as POC, Cr, PAHs, Fe and Pb should be observed. In fact, however, there was no such a relation. For example, Pb and Fe did not correlate each other (See Figure 1 below). Thus, we believe that covariations related to meteorological variations are not the case in the current study. Instead, a strong correlation of POC with crustal species such as Fe and Ca^{2+} ($r^2 > 0.8$ in winter and $r^2 > 0.6$ in spring), and intermediate linear correlation of POC with PAHs and fossil fuel derived *n*-alkanes ($r^2 < 0.6$ in winter and $r^2 < 0.4$ in spring) demonstrated that POC in the PM_{10} largely resulted from soil dust and coal burning emission.

However, we agree with the reviewer's view that coal burning is important. Thus we revised the discussions. See page 10, line 225-233.

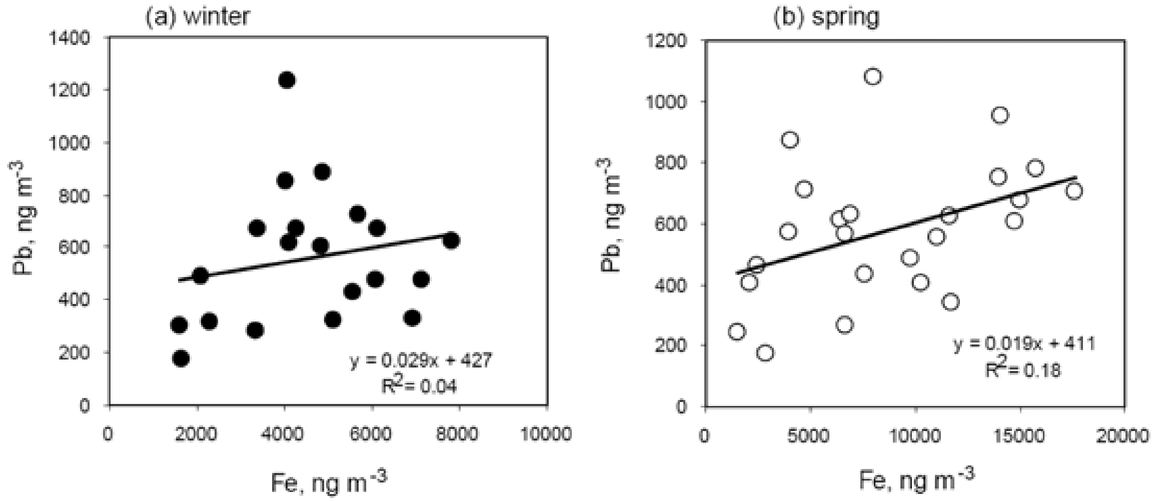

Fig. 1. Correlation of Pb and Fe in the PM10 samples during winter nad spring in Baoji city

8. Page 6903 lines 23-24: a major source of what?

Response

Here means coal burning emission is a major source of Pb and PAHs. We modified the sentence to make the statement clear. See page 10, line 234-236.

9. Page 6904, lines 1-17: there are some redundancies in this paragraph. One hypothesis behind your explanation is that the sources are constant in each season (hence the δ13C at the emission are constant)

Response

Suggestion taken. We re-wrote the paragraph to make the discussion more concisely. See page10, line 236-248. Our assumption that the sources are constant in each season is reasonable, because the diognostic ratios of organic (PAHs and n-alkanes) and inorganic tracers (Fe, Al, Ca andCa^{2+}) consistently pointed out that fugitive dust and coal burning emission are the major sources of the airborne particles. Hence the δ13C at the emission are constant.

10. Page 6904 lines 18-27: again, the sources of N may be of extreme importance for the isotopic composition.

 Response

 We agree with the reviewer that the sources are very important for the isotopic compositions. However, given that the sources in each season are constant, the changes in isotopic compositions of TN are largely caused by the condensation/adsorption of NH_4^+ and NO_3^- onto previously existing particles. See the related discussions in page 11, line 249-247.

Reference:

Huang, X.F., Hu, M., He, L.Y. and Tang, X.Y.: Chemical characterization of water-soluble organic acids in PM2.5 in Beijing, China, Atmospheric Environment, 39, 2819-2827, 2005.

Ram, K. and Sarin, M.M.: Spatio-temporal variability in atmospheric abundances of EC, OC and WSOC over Northern India, Journal of Aerosol Science. Special Issue for the 9th International Conference on Carbonaceous Particles in the Atmosphere, 41, 88-98, 2010.

Wang, G., Kawamura, K., Xie, M., Hu, S., Gao, S., Cao, J., An, Z. and Wang, Z.: Size-distributions of n-alkanes, PAHs and hopanes and their sources in the urban, mountain and marine atmospheres over East Asia, Atmospheric Chemistry and Physics, 9, 8869-8882, 2009.

Wang, H. and Kawamura, K.: Stable carbon isotopic composition of low-molecular-weight dicarboxylic acids and ketoacids in remote marine aerosols, Journal of Geophysical Research-Atmospheres, 111, D07304, doi;10.1029/2005JD006466, 2006.

Warneck, P.: Chemistry of the Natural Atmosphere., pp. 270-275. Academic Press, San Diego.2000.

Xie, M., Wang, G., Hu, S., Han, Q., Xu, Y. and Gao, Z.: Aliphatic and polycyclic aromatic hydrocarbons in atmospheric $PM1_0$ particles in Baoji, China: Implications for coal burning, Atmospheric Research, 93, 840-848, 2009.