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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of aged aerosols in size-resolved Asian dust storm particles transported from Beijing, China to Incheon, Korea using low-*Z* particle EPMA" *by* H. Geng et al.

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We greatly thank Anonymous Referees #2 for his/her valuable and supportive comments. Following the comments and suggestions, we will modify our manuscript as much as possible. Our replies to the reviewers' comments are provided as follows.

* Comment 1. The authors give one conclusion that CaCO3 can react with HNO3 and produce the Ca(NO3)2. Reaction of the acidic gases and basic particles make sense. However, the author gave Mg-containing aluminosilicates can produce soluble

Mg(NO3)2. It is surprising result however more evidence should be provided. The authors didn't made laboratory experiment and found any literature then gave the conclusion. That's not convincing result. The related discussion can occur in the main text, but if the author make it as one important conclusion, the current elemental information were weak. Otherwise, the author need to give more direct evidences (27982).

Response: Indeed, many studies have proved that in the atmosphere CaCO3 can react with HNO3 and produce the Ca(NO3)2. In this manuscript and our previously published articles, both the unreacted and reacted CaCO3 particles were identified using the low-Z particle EPMA. Also, a number of Mg-containing aluminosilicates were found. In the present study, the calculated atomic concentration ratios showed an obvious increase in [Mg]/[Al] and [Mg]/[Si] in the Mg-containing aluminosilicate particles for samples S2 and S3 compared to sample S1. We did not mention that soluble Mg(NO3)2 must have been produced. In the future, we will make laboratory experiment to investigate the reactions of Mg-containing aluminosilicate particles with air pollutants under different humidity and to observe whether Mg(NO3)2 would be produced. Herein, what we analyzed is chemical compositions of the real aerosol particles collected in the atmosphere rather than standard particles in a lab. Whether soluble Mg(NO3)2 could be produced in a lab is another story. Recently, we found some literature which supports our observations, i.e., "the ratio of Mg/Al have been used as an indicator in determining the contribution of local and non-local sources of airborne particulate pollution (Ma, Q., Liu, Y., Liu, C., Ma, J., He, H.: A case study of Asian dust storm particles: Chemical composition, reactivity to SO2 and hygroscopic properties, Journal of Environmental Sciences, 24(1), 62–71, 2012) and that some atmospheric mineral particles were coated by (Ca, Mg)(NO3)2 during haze and Asian dust episodes (Li, W. J. and Shao, L. Y.: Observation of nitrate coatings on atmospheric mineral dust particles, Atmos. Chem. Phys., 9(6), 1863-1871, 2009). These literatures will be cited in our revised manuscript.

* Comment 2. The authors should consistently use the same term or formation, such as

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NOx or nitrogen oxides. Response: All the “nitrogen oxides” in the text will be replaced by “NOx”.

* Comment 3. 27974, 23-25, why is this sentence here? It is not related this study. Response: This sentence (as well as references therein) will be deleted.

* Comment 4. 27978, 18, between-among, deleted “or reactions”. Response: Thanks for the keen comment. In the sentence “The particle size will change during mixing or reactions between the different types of particles” (page 27978, line 18), “between” will be changed to “among” and “or reactions” will be deleted in the revised manuscript.

* Comment 5. 27978, 21, English grammar have-had? Response: Yes, “have” should be changed to “had”.

* Comment 6. 27979, 15, elemental carbon (EC) is not suitable here. EC, soot, and BC are different terms based different study methods. EC was normally used by thermo method, soot was used to describe one particle containing black carbon and some organics, and BC was used by optical absorption method. Obviously, soot is the best term if the author identify single particle from their morphology.

Response: We agree on the comment that EC, soot, and BC are different terms based on different study methods, as said by Buseck, et al. (Are black carbon and soot the same?, Atmos. Chem. Phys. Discuss., 12(9), 24821-24846, 2012). However, soot, easily identified from their special morphology using EPMA, is just one type of carbon-rich particles. Char or coal dust, tar ball, and etc., though having similar X-ray spectra to soot (much higher C peaks than O peaks), are different from soot in morphology. Therefore, none of EC, soot, and BC is the best term. In the revised manuscript, we will use the term “Carbon-rich” as this type of particles in which atomic concentration of C is larger than at least 3 times of O concentration. This type of particles was described in our previous papers such as “Ro, C.-U., Hwang, H., Kim, H., Chun, Y., Van Grieken, R.: Single-particle characterization of four Asian dust samples collected in Korea, using low-Z particle electron probe X-ray microanalysis, Environmental Science

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& Technology, 39, 1409-1419, 2005".

* Comment 7. 27980, 9, between-among Response: "between" will be changed to "among".

* Comment 8. 27980, 19, why is Ca2+ not Ca? Response: "Ca2+-" will be changed to "Ca-".

* Comment 9. 27984, 19-21, why is mean of this sentence here? The author discussed the coagulation of SSA and mineral dust but not chemical reactions.

Response: Thanks for the good suggestion. The sentence "This indicates that ADS particles experienced chemical reactions during their long-range transport over the sea. At least many of them were mixed with SSAs by collision or coagulation or by in-cloud processes (Ma, 2010)" (P27984, line 19-22) will be removed.

* Comment 10. 27985, 4-6, VOCs can condense into organics during the transport. The reason could be explained why the organics was higher in Incheon than Beijing.

Response: Thanks for the good advice. Following it, we will add the sentence "In addition, volatile organic compounds (VOCs), which can condense into organics during transport, also contribute to the increase of OC particles in Incheon (Atkinson, 2000; Wang and Zhao, 2008)". (Atkinson, R.: Atmospheric chemistry of VOCs and NOx, Atmospheric Environment, 34, 2063-2101, 2000.; Wang, P. and Zhao, W.: Assessment of ambient volatile organic compounds (VOCs) near major roads in urban Nanjing, China, Atmospheric Research, 89, 289–297, 2008.)

* Comment 11. 27985, 22-25, Iron could be from different sources. I believe that iron during dust storm should be from the natural sources. Response: The sentence (in line 22-25) will be changed to "They can be from natural sources and human activities (Flament et al., 2008)".

* Comment 12. 27986, 8, Delete "This is surprising". Response: It will be deleted.

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* Comment 13. 27986, 17-25, I suggest that the author make short because most are not from this study. Response: Following the suggestion, we will shorten the sentences (line 16-25) as “Notably, there are many (C, N, O, S)-rich particles in samples S2 and S3 (Fig. 8), whereas none are observed in sample S1, suggesting that the (C, N, O, S)-rich particles were formed favorably in Incheon. Besides (NH4)2SO4 and water-soluble organic matter, NH4NO3 is likely included in the (C, N, O, S)-rich particles as NH4NO3 is water-soluble and the measured N levels are sometimes much larger than S levels (Fig. 11)”.

* Comment 14. 27987, 6-10, Sources of biomass burning near Beijing or Incheon. The conclusion could be right, but the result is from this study. The citation most focus on biomass burning in other period. I suggest that the author should delete this part because the whole paper focus mineral dust not biomass burning.

Response: The suggestion is good. In order to focus on description of mineral dust, we will simplify the paragraph (line 6-10) as “K-containing particles, mostly from biomass burning (Wang et al., 2007; Liu et al., 2000, 2005), are encountered only on stage 6. They have much less abundance in sample S1 than in samples S2 and S3 (2.4% vs. 15.4% and 15.4 %) (Fig. 8).”

* Comment 15. 27988, 14-16, Should the authors add any reference here? Response: The reference (Turšič, et al., 2003) will be added at the end of line 14-16. (Turšič, J., Berner, A., Veber, M., Bizjak, M., Podkrajšek, B., and Grgić, I.: Sulfate formation on synthetic deposits under haze conditions, Atmospheric Environment, 37(25), 3509-3516, 2003.)

* Comment 16. 27988, 19-24, Is it right? Why does the author give one absolute conclusion. Yes, I see the table 6. The atomic concentration from the SEM should have a big error on N.

Response: Although the atomic concentration might have a big error on N from the SEM-EDX, we think it made few influence on our conclusion. Many studies have

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demonstrated that there was intense replacement of CO3- or Cl- with NO3- in the nitrate-containing reacted CaCO3 or sea salt particles. Respecting the comment, we will change the sentence in line 24-25 to “The atomic concentration ratio of Cl/N and Cl/Na in the nitrate-containing reacted SSAs is in 0–0.21 and 0-0.25, respectively (Table 6), indicative of strong Cl depletion”.

* Comment 17. 27989, 1-4, Please reword this sentence. Response: This sentence will be reworded as “The abundant OC particles on stage 6 in sample S1 were thought to have contributed to the formation of (C, N, O, S)-rich particles by being dissolved in airborne water droplets when they transported over the sea”.

* Comment 18. 27989, 11-13, I don’t understand this sentence. Response: The sentence will be changed to “In Beijing, many NaCl-containing particles likely resulting from dried salt lakes were found; and in Incheon many sea spray aerosols were encountered. By EPMA, it was difficult to distinguish NaCl-containing particles from sea spray aerosols because they have similar shapes in SEI and similar Na and Cl signals in their X-ray spectra. Correspondingly, the sea spray aerosols detected in Incheon might include a few NaCl-containing particles from Beijing.

* Comment for Table 3. 5. EC include soot and tar ball? Some study show that tar ball is brown carbon. 6. OC, particle types: liquid droplet or irregular, solid particles. The description can also be used for 7. This is not particle type. 8. Why is not KNO3?

Response: In Tables 3 and Figures 7 and 8, EC will be changed to “Carbon-rich” which include soot, tar ball, and char or coal dust. Please see the response to comment 6. There are many types of OC particles in ambient aerosols. Because EPMA is unable to detect H in the organic matter, it is difficult to distinguish the types of OC particles only based on atomic concentrations of C and O. So, we classified them mainly according to their secondary electron images (SEIs). There were two distinct types of OC particles based on their SEIs. One is type 1 for dark, liquid droplets; and another is type 2 which appears bright, irregular, and solid shape. As far as (C, N, O, S)-rich particles

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are concerned, they are (NH4)2SO4 or NH4HSO4 mixed with water soluble organic carbon (WSOC). Possibly, NH4NO3 is mixed with them. Following the comment, we will change the table's header: "Group" (the first column) will be changed to "Particle types" and the original "Particle types" (the second column) will be changed to "Characteristics and possible compositions". In all the K-containing particles, we didn't find N peaks in their X-ray spectra. Thus, we think there were few KNO3-containing particles in the samples.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 27971, 2013.