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Interactive comment on "Size-resolved source apportionment of ambient particles by positive matrix factorization" *by* J. S. Han et al.

J. S. Han et al.

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Authors' response to comments made by reviewers on "Size-resolved source apportionment of ambient particles by positive matrix factorization" by J. S. Han et al.

Response to Anonymous Referee #2

General comments:

This paper describes the application of positive matrix factorization to chemical speciation analyses of ambient particles collected over Gosan, Korea. The overall organization of the paper is good. The mathematical techniques applied are well described and the results are interesting. However, the conclusions can be strengthened with a better description of the sampling techniques and the meteorological conditions observed

during the sampling period.

: The detailed description of the sampling techniques was discussed in previous works such as Han et al (2004).

(Reference) Han, J. S., Moon, K. J., Ahn, J. Y., Hong, Y. D., Kim, Y. J., Ryu, S. Y., Cliff, S. S., and Cahill, T. A.: Characteristics of ion components and trace elements of fine particles at Gosan, Korea in spring time from 2001 to 2002, Environ. Monit. Assess., 92, 73-93, 2004.

For the specific cases observed in Figure 6, episodic analysis using meteorological information has been added to this paper using backward trajectory model (HYSPLIT 4) with a new Figure 7 as shown in response to referee #1.

(a) continental aerosol, (b) soil dust:

The difference between continental aerosol and soil dust was definitely observed in the backward trajectory analysis using HYSPLIT4 (Draxler, 2004). Figure 7a and 7b respectively shows the back trajectories in 3 hr intervals observed when the intensities of continental aerosol and soil dust source were independently higher than other periods as shown in Figure 6. The trajectories reveal that continental aerosol was transported from further regions including northeastern China than soil dust.

(c) biomass/biofuel burning:

Figure 7c shows that the backward trajectories from 9 to 11 May and at 16 May 2002 when the intensity of biomass/biofuel burning source was relatively high. The trajectories passed by not only the cultivated regions in central China but also the forests and grassland located in northeast China and North Korea. Therefore, it is inferred that this source includes field combustion of agricultural residues as well as biofuel combustion.

(d) ferrous metal source (coarse), (e) ferrous metal source (fine):

As shown in Figure 7d and 7e, the difference of trajectories when they respectively have

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high intensities also supports the separation of two ferrous metal related sources. The source in coarse size range was estimated to be transported from southern industrial regions in South Korea while fine aerosol source regions related to the steel industry could be mainly located in major industrial areas in northeastern China.

(f) volcanic emission:

Finally the volcanic emission source, containing large amount of Al, Si, K, Ca, and Fe, was resolved in the fine size range (0.56˜0.75um) when the trajectories passed around Kyushu Island in which there are several active volcanoes as shown in Figure 7f.

Specific comments:

1. The title would be more informative if it included the geographical location of the DRUM sampler, eg: Size-resolved source apportionment of ambient particles in southern Korea

: Title has been changed to “Size-resolved source apportionment of ambient particles collected at Gosan background site in Korea by positive matrix factorization”

2. p. 5227: 1st Paragraph - What was the RH of the inlet to the DRUM sampler? What was the flow rate? What was the height of the sampler? What were the prevalent meteorological conditions during the time of sampling? What were the likely origins of the air masses?

: The detailed description of the sampling techniques was discussed in previous works such as Han et al (2004). And Episodic analysis using meteorological information has also added to this paper using backward trajectory model (HYSPLIT 4) as is stated above and in response to referee #1.

3. p. 5230: 3rd Paragraph - It does not ‘confirm’ the successful application of PMF. However,it does ‘suggest’ or ‘support’ it.

: “confirms” has been replaced to “suggests”.

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4. p. 5230: 4th Paragraph - While secondary sources were not explicitly resolved, sulfur was. In fact a large portion of sulfur that was resolved was likely in the form of sulfate. How might this have impacted the results?

: Although a large portion of sulfur is generally resolved in the form of sulfate, all secondary aerosols were not composed of sulfate. Secondary aerosol is mostly composed of water-soluble ion components including sulfate, nitrate, ammonium, and carbonaceous materials. PMF analysis separates each source in the form of source profile representing the composition of source. Therefore, the result of PMF analysis only using trace elements (including sulfur) can't completely resolve the secondary aerosol.

5. p.5231: 1st Paragraph - You only fourteen of the fifteen resolved sources. You should explicitly state that the ferrous metal source was resolved into coarse and fine mode here.

: Ferrous metal sources resolved in coarse and fine size range were not an exactly same source. These sources show definitely different source profile and intensity as described in this paper. Generally, ferrous metal related sources can be emitted by various manufacturing processes including electric furnace dust, medium steel furnace, special steel furnace, stainless steel furnace, electric steel furnace, and so on. In addition, two sources are distributed in different size ranges. Therefore, it is inferred that they can be originated from different ferrous metal related sources and different source regions. On account of these facts, the ferrous metal sources in coarse and fine size ranges have to be separately counted as a different aerosol source. Two different ferrous metal related sources have been separately discussed in the revised manuscripts.

(Reference) Philip K. Hopke (1985) Receptor Modeling in Environmental Chemistry, John Wiley & Sons, New York.

6. p. 5231: 2nd Paragraph - It is unclear to me how you compared the elements resolved in this paper to those of previous studies. Did you integrate the mass of each

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element over all sizes from your results? If this is the case, the sentence in the same paragraph 'and the known profiles from previous studies are simply averaged source compositions regardless of aerosol size range' is not clear to me.

: The profiles of fifteen sources were determined by not integrating the mass of each element over all sizes but taking an average of the profiles resolved as a same source in different size ranges. On the other hand, the known profiles in the previous works were mostly obtained from the analysis of TSP aerosol samples collected at emission sources. Therefore, they represent averaged source compositions over all size ranges.

7. p. 5233:1st Paragraph - Could the observation that ferrous metal-related source resolved into two 'independent' sources also be explained meteorologically? Do the atmospheric conditions support a rain-out of coarse mode aerosols when the source contribution is weak compared to the fine mode?

: The analysis of backward trajectory efficiently shows that these two ferrous related sources were originated from different source regions. That fact supports that two ferrous metal related sources are independent meteorologically.

8. p. 5223: 3rd Paragraph - Briefly describe the Beta Ray Absorption method.

: The brief description on the Beta-ray absorption method has been included in the revised manuscript as shown in following.

"The total PM ($<$12um) mass collected by a DRUM sampler was calculated from the PM10 mass concentration obtained at Gosan ambient air quality monitoring site (33° 15'N, 126° 12'E) operated by the Ministry of Environment Korea. PM10 concentration was measured with an interval of 1-hour by the beta-ray absorption method, which has a detection limit of 0.1ug/m3."

9. p. 5223: 3rd Paragraph - Where did the weighing factor of 1.02 assumption come from?

: Weighting factor of 1.02 was based on the previous result that the PM10 mass occu-

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pies about 98% on the average of PM12 mass at Gosan (Han et al., 2005).

(Reference) Han, J. S., Moon, K. J., Lee, S. J., Kim, J. E., and Kim, Y. J.: Size distribution characteristics of particulate mass and ion components at Gosan, Korea from 2002 to 2003, Journal of Korean Society for Atmospheric Environment, 21 (E1), 23-35, 2005.

10. p. 5234: 2nd Paragraph - Could water explain some of the difference in resolved and unresolved mass? What are the uncertainties associated with your chemical resolution techniques?

: The difference in resolved and unresolved mass could not be explained by water contents in aerosol because the data measured in rainy days were basically eliminated from the analysis. This fact has been also described in the revised manuscripts. The uncertainty associated with PMF analysis can be estimated by the correlation between measured and predicted PM mass. In this study, the correlation coefficient is 0.82, meaning that the total uncertainty of the resolved source contributions in eight size ranges is about 18%. In the previous works, ion components such as sulfate, nitrate, and ammonium and organic carbon occupied about 30% of TSP and 40~70% of PM2.5 collected at Gosan back ground site. These results well support that the unresolved mass contribution (46.6+-18%) in this study can be described by the contribution of secondary aerosols mainly composed by ion components and organic carbon.

(Reference) Lee, J. H., Kim, Y. P., Moon, K. C., Kim, H. K., Lee, C. B.: Fine particle measurements at two background sites in Korea between 1996 and 1997, Atmos. Envirn., 35, 635-643, 2001.

Kim, Y. P., Lee, J. H., Baik, N. J., Kim, J. Y., Shim, S. G., Kang, C. H.: Summertime characteristics of aerosol composition at Cheju Island, Korea, Atmospheric Environment, 32 (22), 3905-3915.

Han, J. S., Moon, K. J., Ahn, J. Y., Hong, Y. D., Kim, Y. J., Ryu, S. Y., Cliff, S. S., Cahill,

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T. A.: Characteristics of ion components and trace elements of fine particles at Gosan, Korea in spring time from 2001 to 2002, Environ. Monitor. Assess., 92, 73-93.

Lee, J. H.: A study on the long-range transport of air pollutants in northeastern Asia, Doctor dissertation, Kunkuk University, 1999.

11. p. 5242: It would be useful to include the size cuts on each stage. How do you explain source number five's presence on stage five and seven, but not on stage six? Make sure to state the C = coarse and F = fine.

: Table 2 has been changed with the information on size cut of each stage.

"stage 1: 5.0um˜Inlet, stage 2: 2.5˜5.0um, stage 3: 1.15˜2.5um, stage 4: 0.75˜1.15um, stage 5: 0.56˜0.75um, stage 6: 0.34˜0.56um, stage 7: 0.26˜0.34um, stage 8: 0.07˜0.24um"

In the case of ferrous metal related sources, the source profiles and intensities of them were definitely different in the coarse and fine size ranges indicating that these sources are independent. On the other hand, source number five, municipal incineration source, showed considerably similar source profiles and intensities although it is observed in different size ranges such as stages 1, 5, and 7. The discrete size distribution of incineration source could be caused by the difference in the source region.

12. Figure 1: A key is needed for the symbols.

: Figure 1 has been changed to a figure without any symbols.

13. Figure 6: It would strengthen your conclusions if the temporal variations were compared to back trajectory analyses. This may be especially plausible for the volcanic emissions and biomass burning peaks.

: Episodic analysis using meteorological information will be added to this paper using backward trajectory model (HYSPLIT 4) as shown in response to referee #1.

Technical Corrections:

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1. p. 5224: 1st Paragraph - the dust aerosols emitted has less to do with the high population density, and is more related to geography and farming practices.

: “high population density” has been replaced by “high density of industrial activities” as shown in following.

"Northeast Asia is known to emit a large amount of Asian dust particles as well as anthropogenic pollutants, due to its high density of industrial activities and increasingly high rate of energy consumption."

2. p. 5225: 2nd Paragraph - Spell out particulate matter the first time you. p. 5226: 2nd Paragraph - Spell out DRUM the first time you use it.

: (p5225-6) PM and DRUM are spelled out first time it used in the revised manuscript.

3. p. 5234: 3rd Paragraph - ‘Fig. 8’ and ‘Figure 8b’ the 8 should be 9. This also applies to the same paragraph on the following page.

: Figure numbers have been corrected.

4. p. 5238: Paatero (1997) is miscited.

: Miscited reference was corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 5223, 2005.