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# ***Interactive comment on “Year-round observations of water-soluble ionic species and trace metals in Sapporo aerosols: implication for significant contributions from terrestrial biological sources in Northeast Asia” by C. M. Pavuluri et al.***

## **Anonymous Referee #2**

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General comments: Recently, awareness of the PM pollution has arisen in East Asia region due to the several reasons such as the rapid industrialization in East Asian countries, aridification of inland area, and massive forest fires in Siberia. As mentioned by the authors, long term monitoring will be required for the better understanding of the atmospheric behavior of PM and its potential sources. The authors concluded that soil dust, biogenic emissions, and biomass burning are more important sources for Sapporo TSP than anthropogenic sources and sea-salt emissions. Unfortunately, I could not share the evidences supporting this conclusion from current manuscript.

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Discussion Paper



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I believe that soil and biogenic factors are important contributors of PM generation. From the results of factor analysis, however, I could not confirm whether the contributions of soil and/or biogenic sources certainly exceed the anthropogenic contributions. More quantitative discussion will be strongly required. The authors tried to identify PM emission sources from the viewpoint of similarity in temporal variation of respective species. Qualitatively, this approach seems to be reasonable for primarily generated species. However, the temporal variation of secondarily generated species is generally dominated by complicated chemical reactions in the atmosphere in addition to the amount of precursors. Thus, it seems too simplistic to identify sulfate as biogenic origin from the similarity in temporal variations of SO<sub>4</sub><sup>2-</sup> concentration and BVOC emission. Moreover, the dataset is too small to discuss the correlation of the seasonal trends of respective species (Fig. 4). The factor loading matrix (Table 2) clearly indicates that several different sources are mixed together in respective factors. For example, factor 1 consists of fine mode species (i.e., SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup>) and coarse mode species (i.e., Ca, Al, Ti, and Fe). Even though the factor loadings of coarse mode species are higher than those of fine mode species, it is not always true that the contributions of coarse mode species are more important than fine mode species. This is because the atmospheric concentrations of SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> are much higher than those of coarse mode species. Thus, the anthropogenic contributions may not be negligible. The poor factorization is probably due to the lack of particle size separation and coarse sampling time resolution. From this factorization, it is difficult to estimate respective source contribution quantitatively. Consequently, I conclude that the current manuscript is unsuitable for publication.

Specific comments: P6598, L20-21: More information is needed. Although the authors cited several papers supporting this implication in later section, but in my opinion, Zn, Cu, and Ni are not always treated as tracers of biogenic sources. They are also well known as anthropogenic origin (e.g., steel industry, waste incineration, tire wear, and brake dust) in many other papers.

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P6601, L17-19: Do you mean acidic PM has been observed in Sapporo? How was the pH of extracts?

P6606, L7-8: Pearson's correlation coefficient is applicable to the normally-distributed dataset. I wonder if mannitol is really normal distribution. Good correlation might be obtained from few high concentration data of mannitol.

P6606, L28-P660, L2: This statement is very interesting. Trace elements such as V, Cr, Mn, Fe, Ni, Cu, Zn, and As have been well known as anthropogenic tracers. If these elements are originating from biological sources, can you specify their potential origins in detail? Do you have any ideas on chemical forms of these elements containing in their potential biological sources?

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 6589, 2013.

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