

*Response to Review Comments by Anonymous Referee #1 on “Measurement report: Characterization and source apportionment of coarse particulate matter in Hong Kong: Insights into the constituents of unidentified mass and source origins in a coastal city in southern China” by Yee Ka Wong et al.*

**General Comments by Anonymous Referee #1:**

The given manuscript discusses the importance of  $PM_{\text{coarse}}$  in formulating policies due to its growing relative contribution to  $PM_{10}$  loading in urban atmospheres. The paper uses the PM composition data derived from the measurements conducted in Hong Kong. The use of positive matrix factorization resulted in identifying four  $PM_{\text{coarse}}$  sources, including soil dust, Cu-rich dust, fresh sea salt, and aged sea salt mixed +secondary inorganic aerosols. Results also showed that these four sources can explain unidentified fraction of  $PM_{\text{coarse}}$ . Overall, this work presents a simple approach to understand  $PM_{\text{coarse}}$  composition/sources which can be applied to other locations with similar monitoring needs.

**Response to General Comments:**

We thank the reviewer for the comments and agreeing with the significance of this work. Our response to the comments is given in the following. The response text is marked in blue. References cited in this response document are placed at the end.

This work is nicely constructed, and my comments are listed below to consider:

- Please add reference to the sentence “For example, nitrate in the coarse mode is formed by the reaction between nitric acid ( $HNO_3$ ) from oxidation of  $NO_x$  and pre-existing alkaline aerosols (e.g., sea salt and dust).”

**Response:** We cited the study of Bian et al. (2014) here (Line 47–48), which used field measurements to show that the availability of sea salt and dust particles is one of the major factors affecting the formation of coarse nitrate.

- Please provide more details on the uncertainty matrix. I can’t find any information on the uncertainty of species those concentration is above the detection limit.

**Response:** The derivation of the precisions of measurements is given in the last paragraph of Sect. 2.2. These precision values were used as the PMF uncertainty matrix. We added the following statement in the corresponding section of the main text for clarification:

Line 140–142: “The measurement precisions for each species in each sample described in Sect. 2.2 were used as the uncertainty inputs for the PMF modeling.”

- Why was Deming regression applied?

**Response:** Deming regression was applied because it considers the measurement uncertainties of both variables to be compared in the regression. This avoids biased fitting caused by only considering measurement error in the y variable as in ordinary least square regression. We added this explanation in the main text as follow:

Line 152–153: “This technique is applied to consider the measurement uncertainties of both variables to be compared in the regression.”

- Line 155: What is the basis for using a ratio of 2 to calculate organics, author should provide clarification.

**Response:** Here we take the general understanding that coarse mode organics is more related to biological particles such as pollens, spores and vegetative detritus. These particles are enriched in more oxygenated compounds such as polyols and carboxylic acids. The ratio of 2 is a reasonable estimate adopted from the study

of Edgerton et al. (2009), in which the organic matter-to-OC ratio for coarse mode organics was reported. We included this justification in our main text as follow:

Line 161–163: “The coarse organics were estimated by multiplying the measured OC with a factor of 2, assuming the organics are mainly associated with biological particles, which are enriched in oxygenated compounds such as polyols and carboxylic acids (Edgerton et al., 2009).”

- Line 160: Add reference for using 1.6\*OC.

**Response:** We cited the study of Turpin and Lim (2001), which recommended the use of a ratio of 1.6 for typical urban aerosols.

- Add reference-“Coarse mode nitrate mainly forms by the uptake of HNO<sub>3</sub> by pre-existing alkaline particles forming NaNO<sub>3</sub> in reaction with sea salt and Ca(NO<sub>3</sub>)<sub>2</sub> with soil dust.”

**Response:** We cited the study of Bian et al. (2014) to strengthen the statement.

- Line 220: Add reference for Si estimation.

**Response:** The reference has been provided in the last column of Table 1.

- I am not convinced with the factor 1, it is a mix of two sources. Apart from crustal elements, there is also a significant contribution of Pb, V, Mn and Zn which suggests this source is not properly resolved.

**Response:** We thank the reviewer for his/her views on the PMF analysis. We have considered the comments and revised the source identification section in Sect. 3.2. In the revised version of the manuscript, the back-trajectory analysis part is moved to the source identification section to aid the source interpretation.

We also expanded the source identification part with additional discussion on the possible source categories for the dust factors. By considering the evidence from relevant studies, we renamed the first dust factor as soil dust/ industrial and coal combustion, and the second factor as construction dust/copper-rich emissions.

- The discussion about the seasonal contribution/variation of PMF factors should be enhanced. Currently, the given discussion is not sufficient to understand their origin. In addition, it would be great if author can also provide some insight on sources based on the previous receptor modeling results. Are the present results aligned with the previous observations?

**Response:** We have revised the discussion on the seasonal variation in source composition, please be referred to Sect. 3.4.1.

We also evaluated our source apportionment results with the useful information taken from a previous local PM<sub>10</sub> study by Yuan et al. (2013). The study analyzed the speciation data obtained from the local PM<sub>10</sub> network over an 11 year long period. In particular relevancy, the authors observed that the dust contributions were similar across different monitoring stations in Hong Kong, indicative of the regional nature of this source. The source origin of fugitive dust identified in our study aligns with that proposed by Yuan et al.

- Is the contribution of Cu-rich dust and construction dust by Zhou et al., comparable? What is the similarity between these two sites? Are the air masses originates from construction active area?

**Response:** The Cu-rich dust factor is to certain extent similar to the construction dust by Zhou et al. Both factors show higher abundance of Ca than Si. Also, both factors are depleted in Al, Si, and K. However, no coarse mode Cu was reported in the PMF factor profiles by Zhou et al., and hence it remains uncertain to what extent the construction dust factor is similar to ours.

The discussion on this factor has been expanded in the revised manuscript (Sect. 3.2.2). Based on the updated analysis, this factor is renamed as construction dust/copper-rich emissions.

Both Foshan (Zhou et al.'s study area) and Hong Kong are located within the Greater Bay Area (GBA). The Foshan city is one of the most important industrial hubs in the GBA, whereas Hong Kong is a commercial city with much less intense industrial activities. It is well documented that the air quality in Hong Kong is heavily influenced by emissions in the GBA, especially under meteorological conditions favorable to the transport of air pollutants. Therefore, the Foshan city might represent one of source areas responsible for the degraded air quality in Hong Kong imposed by regional transport of air pollutants.

As to the source areas that the continental air masses typically travelled before reaching Hong Kong, it is believed the air masses might pass through the various economic and industrial hubs in GBA or even the larger southern China region, where emissions from multiple sources are carried and mixed. Hence it is difficult to determine whether the air masses originate specifically from construction active areas.

- In the section 3.4, author mentioned that the aerosol samples were not corrected for sampling artifact of nitrate. Did author try to apply the correction and observed any change in the nitrate measurement?

**Response:** We did not try to correct for the sampling artifact of nitrate. In principle, the artifact effect could be evaluated by performing co-sampling of aerosols with a denuder installed upstream of the filter to remove gas phase nitric acid. The extent of the nitrate sampling artifact is expected to be temperature-dependent and aerosol chemical composition-dependent, therefore varies from day-to-day. This variable nature makes its correction difficult. The effect of this type of artifact on coarse nitrate measurement warrants further investigation.

## References

- Bian, Q. J., Huang, X. H. H., and Yu, J. Z.: One-year observations of size distribution characteristics of major aerosol constituents at a coastal receptor site in Hong Kong – Part I: Inorganic ions and oxalate, *Atmos. Chem. Phys.*, 14, 9013–9027, <https://doi.org/10.5194/acp-14-9013-2014>, 2014.
- Edgerton, E. S., Casuccio, G. S., Saylor, R. D., Lersch, T. L., Hartsell, B. E., Jansen, J. J., and Hansen, D. A.: Measurements of OC and EC in coarse particulate matter in the Southeastern United States, *J. Air Waste Manage.*, 59, 78–90, <https://doi.org/10.3155/1047-3289.59.1.78>, 2009.
- Turpin, B. J. and Lim, H. J.: Species contributions to PM<sub>2.5</sub> mass concentrations: Revisiting common assumptions for estimating organic mass, *Aerosol Sci. Technol.*, 35, 602–610, <https://doi.org/10.1080/02786820119445>, 2001.
- Yuan, Z. B., Yadav, V., Turner, J. R., Louie, P. K. K., and Lau, A. K. H.: Long-term trends of ambient particulate matter emission source contributions and the accountability of control strategies in Hong Kong over 1998–2008, *Atmos. Environ.*, 76, 21–31, <http://dx.doi.org/10.1016/j.atmosenv.2012.09.026>, 2013.