Atmospheric Chemistry and Physics Discussions

Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2021-1030-RC2, 2022

Comment on acp-2021-1030

Anonymous Referee #2

Referee comment 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., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1030-RC2, 2022

This study performed chemical speciation for $PM_{2.5}$ and PM_{10} samples collected in Hong Kong during 2020/01-2021/02. The results showed that the annual average concentration of PM_{coarse} (PM_{10}-$PM_{2.5}$ mass) accounted for ~50% of PM_{10}. Unlike $PM_{2.5}$, only ~75% of PM_{coarse} mass was explained by identified chemical components. The authors supposed that the unidentified part was dominated by geological components and aerosol liquid water. Moreover, several tools were utilized to apportion PM_{coarse} to specific sources and areas, particularly for the unidentified fraction. In general, this manuscript is well organized and written. But two major issues should be addressed before the consideration for publication.

1. In this work, the thermodynamic equilibrium model (ISORROPIA II) was adopted to estimate aerosol liquid water (ALW) in PM_{coarse}. After mass closure and PMF analysis, the authors concluded that the unidentified PM_{coarse} (4.1 μg m^{-3}, ~25%) was substantially contributed by ALW (1.2 μg m^{-3}).

Have the authors performed mass closure for $PM_{2.5}$ or PM_{fine}? Because the fine particles are more enriched with water soluble components (e.g., secondary inorganic ions), ALW should contribute more fractions to $PM_{2.5}$. According to section 3.1.1 (lines 158-160), it seems that $PM_{2.5}$ is mainly composed of NH_4^+, NO_3^-, SO_4^{2-}, OC, and EC (~80%).

If ALW contributes a significant fraction of PM_{coarse} based on filter sampling, there's no reason that it contributes less to PM_{fine}.

In fact, ALW is not stable on filters, and is subject to loss during long-term sampling and transportation.

So, the contribution of ALW to unidentified PM_{coarse} might not be estimated appropriately with the current study design.

2. When input PM_{coarse} mass for PMF analysis, it was presumed that the unidentified PM_{coarse} fraction have the same sources as identified components.

In this work, four factors linked with soil dust, copper-rich dust, fresh sea salt, and aged sea salt were identified using measured species data. Since understanding the sources and formation pathways of PM largely depends on how well they are identified, the sources of un-speciated coarse PM are unknown and might not be the same as measured species. If the unknown fraction of coarse PM was apportioned to the four identified factors, some factors contributions would be over-estimated. Because PMF may over-attributed PM_{coarse} to certain factors as it fits measured species (Shrivastava et al., 2007). This will occur if makers for unknown PM_{coarse} are not included in the PMF model (Shrivastava et al., 2007).

Therefore, the source apportionment method for unidentified PM_{coarse} mass is not appropriate. The authors should focus on sources of identified PM_{coarse} components.

References

Shrivastava, M. K., Subramanian, R., Rogge, W. F., and Robinson, A. L.: Sources of organic aerosol: Positive matrix factorization of molecular marker data and comparison of results from different source apportionment models, Atmospheric Environment, 41, 9353-9369, 10.1016/j.atmosenv.2007.09.016, 2007.