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Comment on acp-2020-533

Anonymous Referee #2

Referee comment on "Physical and chemical properties of urban aerosols in São Paulo, Brazil: links between composition and size distribution of submicron particles" by Djacinto Monteiro dos Santos et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-533-RC2, 2021

This manuscript reported the composition of submicron aerosol measured by an aerosol chemical speciation monitor (ACSM) in Sao Paulo, Brazil. PMF analysis is performed for the sources apportionment of organic aerosol. One novel aspect of this manuscript is to apply multivariate linear regression (MLR) analysis to estimate the size-dependent PM composition. However, several concerns regarding this analysis are listed below and should be carefully addressed. Given that the majority of the manuscript focuses on reporting the measurements of PM composition and size distributions, I recommend this manuscript to be classifies as measurement report. Overall, I recommend publication after major revisions.

Comments

- One major concern regarding the MLR analysis is that the mass balance seems violated. In Table 5, the sum of beta coefficients over one column should be no larger than 1, in order for the coefficients to be physically meaningful. In addition, the MLR model results of each PM constituent are additive, which leads to total PM1 concentration = 15.5 * nucleation model volume + 0.793 * Aiken model volume + 0.826 * Accumulation model volume. This suggests that more particles in the nucleation model are required by MLR than actual measurement. Another way to check the mass closure is to convert the reconstructed mass loading in each mode to volume using particle density and then compare the converted volume to the measured volume in that mode.

- Another concern is that particle sizes measured by ACSM (70-900 nm aerodynamic diameter) and by SMPS (10-450 nm electrical mobility diameter) do not fully overlap. As large particles account for a dominant fraction of PM mass, the narrower range of

SMPS measurement may cause substantial uncertainty in the MLR analysis.

- More information about the procedure to fit the particle size distribution should be provided. How to decide the number of lognormal modes in fitting the measured particle size distribution? Is the center of each mode allowed to vary between different particle size distributions? If so, what is the variation of the center diameter?
- The discussions in section 3.3 is scientifically correct, but cannot be inferred based on the observations in this study. In this section, the particle size distributions are compared between low and high PM concentration. As the PM concentration is driven by large particles, it is not surprising that the number concentration in accumulation mode shows the largest difference in this comparison, followed by aiken mode. Looking at Lines 290-292 in conclusion section, it is correct that "the accumulation mode shows a larger increase from low PM1 conditions to high PM1 conditions". In this same line, it is also correct that "when aerosol surface is enough to favor the condensation of vapors onto pre-existing particles, inhibiting nucleation, and resulting in particle growth". However, the linkage between these two correct statements are missing in this study. In other words, what is observed in this study is due to how the data are segregated, and it cannot be used to infer any particle growth mechanism.
- Results from the MLR analysis should be elaborated and further explored. For example, why is eBC dominantly in the accumulation mode, but almost zero in the Aiken mode? Does this observation suggest the eBC at the measurement site is heavily coated from regional transport? If so, does it contradict with vehicle emission as the major source in this study? Why is a large fraction of inorganic ions in the Aiken model, but not the case for organics?
- Stepwise selection should be applied in the MLR to explore the explanatory power of each term. In table 5, please list the p value of each term, which will help to examine the importance of each predictor.
- Line 134. Is the SO2 0.61 ppm or ppb?