

Wang et al. studied approaches for the automatic classification of SPMS data by machine learning. The authors created a dataset containing 24,000 particles and used supervised algorithms to tackle the classification problem. Considering the importance of automatic classification of large amounts of SPMS data and the potential use of this automatic approach in future real-time monitoring, I would support the final publication after addressing the comments below.

Specific Comments

Section 2: The previous work should be highly summarized rather than listed one by one. I recommend that the summary of related work be concluded in the introduction. Some parts in Section 2, e.g., the description of different algorithms, should be elaborated in the methodology. The authors should better organize the structure of the manuscript.

Lines 71–73: Please rewrite the sentence. The numbers are presented misleadingly.

Line 175: Please add more details about the SPMS measurement and analysis, e.g., sensitivity, calibration, uncertainty, software, etc. SPMS also gives the particle size information. Could the authors provide more results about the particle size measurement? Will the particle size affect the automatic classification results?

Line 188: How do authors divide the data into two parts for labeling and verification? Are there any criteria, or are they just random? If the data are derived from the same sampling site, which means these particles probably have similar composition, is it reasonable to divide the data into different parts and use the “blind data” for verification?

Line 350: The authors mentioned the signal of K^+ and the signal of $C_3H_3^+$ at the same m/z position, which brought some uncertainty to the prediction. Is it possible to distinguish these ions at the same m/z position in SPMS? Could the authors estimate the uncertainties of applying the method used in this study to analyzing the SPMS data from other sites with different aerosol compositions?

Section 4.2: Since the optimized models with the five algorithms all performed well, which algorithm would the authors recommend in the future work? Now the prediction accuracy of supervised algorithms exceeded 97%. Will the accuracy still be perfect when the approaches are used for analyzing other datasets? How would the aerosol sources impact the prediction results? The authors should add more discussion on the uncertainty of the method, and the feasibility of application in other areas.