Response to Reviewers

Manuscript: amt-2019-106
Manuscript title: Classification of iron oxide aerosols with a single particle soot photometer using supervised machine learning

The discussion below includes the comments from the reviewer (bold) and my responses to the specific comments (red). Modifications to the manuscript text are given in italics, and line numbers refer to the original document.

Response to Reviewer #1:

The author demonstrates for the first time the usefulness of supervised machine learning algorithms for post-processing of the waveform data acquired by the single-particle soot photometer, with particular attention to the classification of iron oxide aerosols. First, the author provides a detailed review of the previous works and clarifies the is- sues to be solved/mitigated in this work. Second, the author defines the (physical and mathematical) features embedded in the signal waveforms and explain the machine learning algorithm applied to them. Finally, the author shows the suggested algorithm can reduce the chance of misclassification of the iron oxide aerosols than the conventional simpler algorithm. Along with the presentation of the results, the author also fully explains the limitation of the applicability due to a particular selection of laboratory samples used to train the algorithm. The manuscript is very logically written, and all the figures are easy to understand. Considering the superior quality of discussion and presentation, I can recommend the publication of this work. However, I request minor revisions to improve the readability and influence to a broader audience (including other SP2 users).

I would like to thank the reviewer for their positive comments and very useful suggestions to improve the clarity and flow of the manuscript. I have addressed their specific comments below.

Minor comments: Most of the contents in sections 3.3-3.5 look like an overview of "the established theory" of machine learning. If so, the author could shorten these sections (or moved to the supplementary information).

Thank you for this comment. I have chosen to move the discussion about decision tree classifiers to an Appendix for the interested reader, but have retained the details about training and optimizing the random forest in the main text for readers who may be coming from an interdisciplinary background.

p.2, line 8. nitrogen - > nitrate?

Thank for pointing out this typo; this line has been updated.

p.8, line 24. real part -> imaginary part?

Thank you for pointing this out. I have updated the text.

p.18, line 9-10. "retaining the 11 most important features for the 6-class case and the 9 most important for the 3-class case" Please refer Table 3 in this sentence. Otherwise, readers could not follow which features are used here.

Thanks for pointing this out. I have updated these lines to include the reference to Table 3. I have also added additional clarification to Section 4.1 on the ranking of feature importance.

p. 18. L. 9-10. We remove the least important features (retaining the 11 most important features for the 6-class case and the 9 most important for the 3-class case; see Table 3, columns 3 and 5 for the subset of features for each case)

p. 18, L. 2-3. The relative importance of different features (given in Table 3) is estimated from the fraction of samples in the data set for which the decision pathway is impacted by that feature (Pedregosa et al. 2011).

p.24, line 6-7. "This method improves upon the performance of previous classification methods using only 3 or 4 features derived from the single particle signals" Please clarify which features the author mention here.

These lines have been updated to clarify the features I was referring to were the incandescent peak height, the color ratio, the core scattering, and the post-incandescent scattering amplitudes.

p.25. line 4-5. "we recommend acquiring samples for training data sets with the same instrument, optical configuration, and operating conditions as the data sets to be processed." To my opinion, it is better to mention this point as "important requirement" rather than "recommendation".

Thanks for pointing this out. I've adjusted the language of this paragraph to clarify that using an instrument with the same configuration is absolutely necessary to apply the supervised learning algorithm with confidence. I've also added additional details to the supplemental information demonstrating the dependence of the color temperature ratio on the alignment of the detectors in the instrument (Figure S3).

p.25 L.4-5 In order to use supervised learning algorithms to classify aerosols with an SP2 during aircraft and field observations, it is very important to acquire the samples for training data sets with the same instrument, optical configuration, and operating conditions as the data sets to be processed. Several of the features (in particular the color ratio) demonstrated strong dependence on detector alignment or may be affected by the specific laser power settings (See Supplementary Materials Figure S3 for additional details).

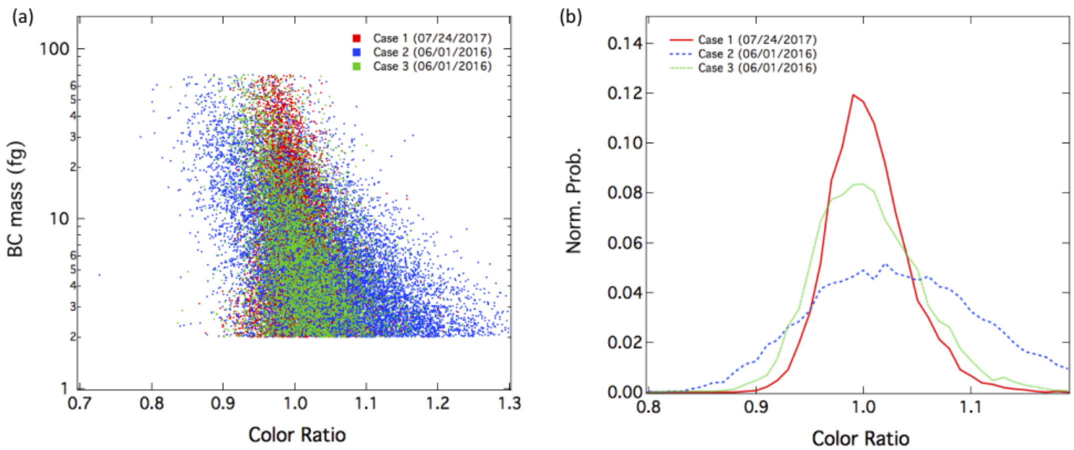

Figure S3. Influence of detector alignment on the incandescent blue amplitude (mass) to color temperature ratio relationship for fullerene soot samples (a) Mass vs. color ratio for fullerene soot sampled by the NOAA SP2 on three different occasions, with three independent alignments for the blue and red PMT's. The color ratio in each case was normalized to 1.0 for fullerene soot with a mass of 10 fg. Greater variability in color ratio was observed when the detectors are not well-aligned (as in case 2) (b) Normalized histograms of the color ratios for fullerene soot for particles with masses between 2 and 70 fg for the three different optical alignments demonstrate differences in the width of the distributions.