Response to Reviewer #2

In this well-structured and very detailed paper, four sparse algorithms (SPLSa, SPLSb, EN and EN-PLS) were considered and evaluated for selecting relevant mid-infrared absorption bands in the calibration model building process. Using FTIR spectra obtained in transmission mode, two types of sparse calibration models were constructed for predicting/interpreting: (1) abundances of four organic functional groups (alcohol COH, carboxylic COH, alkane CH and carbonyl CO) and (2) TOR OC and EC concentrations in ambient aerosol samples. The paper has also presented a thorough analysis of the constructed models. I find the paper suitable for publication in AMT and only have very few minor comments[.]

We thank the reviewer for the positive comments.

Page 3, line 75 onwards: Samples were taken from seven sites in the IMPROVE network. Did the investigators set out any criteria for selecting these sites for the study? It might be a good idea to provide a brief description of the sites to gauge their representativeness of the network. Throughout the paper, nothing was mentioned about these sites except that they were comprised of rural and urban sites (from Figures 3 and 4).

These sites comprise the entire IMPROVE 2011 spectra set available (Ruthenburg et al., 2014), and we have developed and extensively evaluated a set of base case models for prediction of FG and TOR OC and EC Ruthenburg et al. (2014); Takahama and Dillner (2015); Dillner and Takahama (2015a,b). The number of monitoring sites for which FT-IR is available has since expanded (e.g., Reggente et al., 2016), but for this work we have selected the original, well-studied set of samples to specifically investigate the impact of sparse algorithms on each type of calibration model.

We have added to the Introduction section (1):

"These past studies evaluate various performance metrics achieved by statistical calibration models using the full set of wavenumbers, and we evaluate the effect of variable selection on model performance and interpretation."

The chemical composition as parameterized by OM/OC has been included in the Methods section (2.1.1):

"The OM/OC ratio estimated in ambient samples span a range of 1.46 and 2.01 between the 10th and 90th percentiles, with a median ratio of 1.69 (Ruthenburg et al., 2014)."

We are currently extending our application of sparse algorithms for further understanding $PM_{2.5}$ in a wide range of environments. With regards to representativeness, we hope to address this topic in future studies.

Page 4, line 100: The 250 laboratory standards used were mixtures of seven compound types. What are these compounds? If this has been described elsewhere, citing the relevant literature would suffice.

The compounds are: 1-docosanol, D-glucose, fructose, levoglucosan, malonic acid, adipic acid, suberic acid, arachidyl dodecanoate, 12-tricosanone. We apologize for the error but

there are nine compounds instead of seven, and has been corrected. These compounds have been documented by Ruthenburg et al. (2014) and the citation has been inserted in to the Methods section (2.1.1).

Page 40, Figure 3 legend: What do the investigators mean by anomalous clusters? PM: PM_{10} or $PM_{2.5}$?

We thank the reviewer for pointing out these omissions. In the caption of Figure 3, we have added this statement: ""Anomalous" samples are those identified by Ruthenburg et al. (2014) (38 samples or 5% of the total set) that share similar spectral profiles and large disagreement with TOR in estimated OC. The cause for the disagreement is at present time unknown."

We have noted that these are $PM_{2.5}$ samples in the Introduction section (1): "We revisit calibration models for four FGs developed using laboratory standards (Ruthenburg et al., 2014; Takahama and Dillner, 2015), and TOR OC and EC calibration models developed with ambient $PM_{2.5}$ samples collected in 2011 at seven sites within the Interagency Monitoring of PROtected Visual Environment (IMPROVE; Malm et al., 1994; Hand et al., 2012) monitoring network (Dillner and Takahama, 2015a,b)."

and in Methods section (2.1.1): "For this work, we use 794 pairs of ambient $PM_{2.5}$ samples collected in the IMPROVE monitoring network[...]"

Grammatical/typographical corrections: Page 38, Figure 1 caption: Should this be Appendix C instead of Section C? Please go over the manuscript again and proofread it.

We thank the reviewer for catching this and other typographical errors. This is Appendix C instead of Section C and the correction has been made. Additionally, minor errors found upon final proofreading have been corrected and are highlighted in the manuscript accompanying this response.

References

Dillner, A. M. and Takahama, S.: Predicting ambient aerosol thermal-optical reflectance (TOR) measurements from infrared spectra: organic carbon, *Atmospheric Measurement Techniques*, 8, 1097–1109, doi:10.5194/amt-8-1097-2015, 2015a.

Dillner, A. M. and Takahama, S.: Predicting ambient aerosol thermal-optical reflectance measurements from infrared spectra: elemental carbon, *Atmospheric Measurement Techniques*, 8, 4013–4023, doi:10.5194/amt-8-4013-2015, 2015b.

Hand, J. L., Schichtel, B. A., Pitchford, M., Malm, W. C., and Frank, N. H.: Seasonal composition of remote and urban fine particulate matter in the United States, *Journal of Geophysical Research: Atmospheres*, 117, doi:10.1029/2011JD017122, 2012.

Malm, W. C., Sisler, J. F., Huffman, D., Eldred, R. A., and Cahill, T. A.: Spatial and seasonal trends in particle concentration and optical extinction in the United States, *Journal of Geophysical Research: Atmospheres*, 99, 1347–1370, doi:10.1029/93JD02916, 1994.

Reggente, M., Dillner, A. M., and Takahama, S.: Predicting ambient aerosol thermal-optical reflectance (TOR) measurements from infrared spectra: extending the predictions to different years and different sites, *Atmospheric Measurement Techniques*, 9, 441–454, doi:10.5194/amt-9-441-2016, 2016.

Ruthenburg, T. C., Perlin, P. C., Liu, V., McDade, C. E., and Dillner, A. M.: Determination of organic matter and organic matter to organic carbon ratios by infrared spectroscopy with application to selected sites in the IMPROVE network, *Atmospheric Environment*, 86, 47–57, doi:10.1016/j.atmosenv.2013.12.034, 2014.

Takahama, S. and Dillner, A. M.: Model selection for partial least squares calibration and implications for analysis of atmospheric organic aerosol samples with mid-infrared spectroscopy, *Journal of Chemometrics*, 29, 659–668, doi:10.1002/cem.2761, 2015.