

## ***Interactive comment on “Near-infrared laser desorption/ionization aerosol mass spectrometry for measuring organic aerosol at atmospherically relevant aerosol mass loadings” by S. Geddes et al.***

### **Anonymous Referee #2**

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The manuscript entitled, "Near-infrared desorption/ionization aerosol mass spectrometry for measuring organic aerosol at atmospherically relevant aerosol mass loadings", by S. Geddes et al., details a new instrument for measuring the chemical composition of complex organic aerosols. The authors describe an aerosol mass spectrometer that has sufficient sensitivity and molecular specificity (i.e. little fragmentation) to make ambient measurements. This instrument is an important advance in the development of soft ionization methods for aerosol analysis.

Overall, the manuscript is well written and is well within the scope of Atmospheric

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Measurement Techniques. The title, abstract, and figures (with the exception of Fig. 4, see below) are sufficient for a reader to follow the experimental methods, analysis and generally support the conclusions reached by the authors. The manuscript is the first report from this group on this new type of aerosol mass spectrometer and as such the manuscript generally provides most (see specific comments detailed below) of the experimental details necessary for the wider community to evaluate its potential applicability for measuring ambient organic aerosol.

### Specific Comments

(1) Figure 2: The signal response vs. particulate mass sampled should be a linear function. While the data points generally fall upon the dashed line, the points measured at higher mass loadings (40–100 pg) show significant deviation. One would less scatter at higher loadings due to better signal-to-noise ratios. In fact the data look like they might be better fit to a non-linear function. The authors need to address why this might be so. Measurements at higher mass loadings, while not atmospherically relevant, do provide critical tests of the method to determine over what range this instrument operates linearly. For example, do the authors see saturations effects at high loadings? Are there space charge limitations that ultimately determine the linearity of the signal with loading? At higher loadings how do the authors know that all of the aerosol is completely desorbed and ionized from the AI probe? Given the quality of the data in Fig. 2 a more thorough discussion and perhaps measurements at higher mass loadings are needed to illustrate more clearly both the capabilities and limitations of their instrument.

(2) It difficult for a reader to draw any firm conclusions from Fig. 4 regarding the time evolution of the chemical constituents of the alpha pinene SOA. This is in part due to the figure format itself. The authors should recast this data as a two dimensional plot (Ion Intensities of various peaks vs. Coa) so the reader can more closely compare individual ion species with the total aerosol loading (i.e. Figure 3a) for example. The authors then need provide a more detailed discussion of the time behavior of the

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various aerosol components. Do most of the ion signals simply grow with Coa or do various components evolve in different ways as the aerosol is formed? Do the authors see changes in partitioning as a function of loading?

(3) The mass spectra (figure 3) of the alpha pinene SOA contains only a handful of peaks (5 or so assigned) between  $m/z$  100-300, which suggests that the SOA formed is rather simple chemically. While the authors do a good job of putting the peaks they do assign in the proper literature context, this mass spectrum is significantly less congested (with fewer high molecular weight features) than others reported in the literature using a variety of ionization sources (e.g. VUV, APCI, etc.). This deserves more discussion than what is currently included in the manuscript.

(4) The authors need to estimate what fraction of the total SOA they are detecting so the reader can get a sense of how quantitative the technique is for measuring SOA. I would like to see a plot of the total ion signal as a function of SOA loading or reaction time. Does the total ion signal follow the total aerosol loading? A plot of total ion signal vs. aerosol loading could be compared to Figure 2 to better elucidate whether the instrument response is similar to that of a pure particles (Oleic acid).

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