We would like to thank the reviewer for their recognition of the novelty of the approach presented here. In the following we separate and number all distinct comments in order of their appearance in the review, highlighting new text added to the manuscript where appropriate.

Specific Comments
1) P2, Line 2: Could the authors state the rationale for choosing to assess performance simply with cosine angles?

Response: We are happy to clarify this, as in response to the other referee. There are indeed other metrics we could have employed to measure distance between mass spectra, however we considered cosine to be the most appropriate. Firstly, because our aim is to replicate the AMS instrument response function, which can be modelled as a linear addition of multiple component mass spectra, we reason that it would make the most sense to use a metric that places linear weight on the peaks' relative intensities. Secondly, while a different metric may place a relatively greater weight on intermediate peaks (thus ensuring a more general agreement over a larger number of peaks), we would have to take care not to also unduly weight the minor peaks, which can be problematic. As such, an element of subjectivity would have been introduced in the choice of algorithm, which in itself would require more testing. It is possible that there is a better closeness metric that could be tested as part of future work and this would be easily testable within the STRAPS framework, however see that as outside the scope of this particular paper.

2) P2, Line 15: could the authors suggest which other analytical techniques could potentially benefit from the method and include appropriate references?

Response: It is possible that other techniques may benefit from this, but this is specifically tested around an instrument that gives ensemble data in response to a liner addition of signatures. Other forms of mass spectrometry, such laser desorption and ionisation and electrospray ionisation, suffer from matrix effects, so the model will need further development for this. It will also be of limited use for 'soft' ionisation techniques where there is little molecular fragmentation (such as chemical ionisation mass spectrometry), as the components will mainly be intact molecular ions (or adducts) that will require no training. However, it could be useful in interrogating poorly-resolved mixtures in gas chromatography mass spectrometry. It may be very powerful when applied to spectroscopic techniques such and nuclear magnetic resonance spectroscopy or Fourier transform infrared spectroscopy.

3) P4, Section 2: It is unclear from this text whether the fingerprint is in fact a single mass spectrum of m/z vs abundance. Can this be section be rephrased slightly with a more explicit statement, please?

Response: Apologies. We have replaced the sentence **'**Each molecule has varying levels of structural features, the combination of which provides each molecule with a 'fingerprint**''** with **'***Each molecule has varying levels of structural features, which can be written in terms of a 'fingerprint'. This fingerprint is a numerical identification of a given structure that can equally be thought of as stoichiometric information*…**'**

4) Further to this, should each column refer to a given m/z, is this enough information, or are other concomitant spectral features required to validate the presence of a certain functional group? If each single column "key" is able to contain sufficient information, can the authors clarify and appropriately state this in the text (further to references e.g. Ulbrich et al.)?

Response: We apologize for any lack of clarity here. The collection of molecules, represented as SMILES strings, is parsed to produce a matrix where each column represents the stoichiometry of a particular key, or feature. This entire matrix is used to fit a predict model for each m/z channel.

We have added the following, similar, text to the end of page 4 to attempt clarification of this procedure: *'To re-iterate, in constructing a model that can predict AMS mass spectra, a library of compounds with measured spectra are used to train a series of regression techniques. This collection of molecules, represented as SMILES strings, is parsed to produce a matrix where each column represents the stoichiometry of a particular key, or feature. This entire matrix is used to fit a predict model for each m/z channel.* '

5) P6, Section 3.1: Can the authors comment on the sensitivity of the technique to the various functional groups listed on line 30, for example? Are there any inherent instrumental sensitivity issues with certain functional groups that might limit the effectiveness of the technique at a top level?

Response: The fact that AMS and even EI in general has issues with certain functional groups (see cited literature, in particular Canagaratna et al., 2015) is well documented. Examples include the overlap of multiple functional groups at m/z=43 and the tendency for multifunctional molecules to generate a large signal at m/z=44. However, providing 'top-down' rules for this would be inherently difficult and it is for this exact reason that we chose to test the technique using pre-existing fingerprinting techniques and objectively determine their comparative performance. With further work, it may be possible to develop an AMS-specific fingerprinting technique based on instrument knowledge and compare this against the conventional fingerprinting techniques, however one must take care not to base the fingerprinting technique too closely on the laboratory data that will subsequently be used for training, as this will introduce an element of confirmation bias and thus may give false confidence in the fitting and subsequent extrapolations.

6) Owing to composition dependence acknowledged by the authors, it would be nice to see additional data c.f. Figures 5 and 6, for other single precursors. Are these data available?

Response: We agree this would be very useful. Firstly, we feel that recommendations for additional data described in section 4 should be pursued before a detailed analysis of additional precursors systems. Secondly, the state of box-models used to study multiple precursors is highly variable and not particularly well documented or with a common data/software repository. The recent study of McVay et al (2015) might improve predictions presented in figure 7-9 due to additional mechanisms such as the formation of HOMs. However, the presentation of other box-model suggests the requirement for tracking each compound in the condensed phase, to be used as input into STRAPS, is not necessarily followed. Given the two commonly used chemical mechanisms, the Master Chemical Mechanism (MCM) and GECKO, carry individual molecular representations as SMILES strings, this would not take much work to improve. It would be a very useful development to have a central repository of box-model output that is visible and easy to access.

7) P7, Lines 30 – 33: Regarding the statement – "This reflects sensitivity to information used in the training process and how similarity between performances should be taken with caution in prescribing which method to take forward", as this represents a limitation, could the authors expand their discussion slightly, i.e. potential magnitude of uncertainty associated with

inaccurate method prescription? Further, could the authors clarify the sensitivity of the technique to user required experience and expertise?

Response: Regarding the first point, we cannot at this stage prescribe a magnitude of uncertainty for any given method without further testing. To re-iterate the recommended data requirements presented and extended in section 4, it would be highly useful to obtain additional laboratory data on systems from a specific series of compounds to enable this quantification. Regarding the second point, we would hope that the use of openly available libraries in the Scikit learn package, and fully documented software repositories, will enable anyone to replicate or extend the work presented here.

8) P8, Lines 18 – 20: When the authors refer to addition of data from mixed systems, are they referring to an ensemble photo-oxidation study, or simply an inert multicomponent mixture? Did the authors consider a test intermediate in complexity, e.g. the obvious intermediate between a single compound mass spectrum and a chamber photo-oxidation experiment would be an analysis of a mixture of 2-3 compounds, without the complex oxidative chemistry. Was this considered?

Response: This is a very good point and, yes, we did consider this. We are specifically referring to a range of mixed systems from inert multicomponent systems to those from additional chamber studies. The inert, or even reactive, multicomponent mixtures would enable us to better validate, and provide more training, to the tools presented here. This would give us increased, or decreased, confidence in the application to chamber systems. It would also enable us to perhaps construct a more generally applicable set of fingerprints to use in the training process.

9) Regarding the AMS data employed (e.g. Figure 7): How were these data treated? Were they experiment averaged, summed, normalised? Despite the reference to Alfarra et al., 2013, it may be useful to briefly state this on introduction of the experimental data in order to provide context.

Response: The data were normalised, as it was the relative peak contributions that were of interest; quantitative agreement on mass concentrations is a separate area of enquiry outside the scope of this work. We have also added a brief reference to the conditions mentioned in the Alfarra et al., 2013 study in section 3.3, page 9: *Figure 7 displays the predicted mass spectra for the GECKO-A model results of Valorso et al. (2011) combined with the experimental data taken from a chamber-based α-pinene SOA formation experiment reported by Alfarra et al. (2013). This spectra represents "aged" aerosol, after 4 hours of experiment, during which the VOC/NOx ratio was ~2.* The same information has been added to the caption of Figure 7.

10) Please check reference formatting throughout, e.g. spaces between text and parentheses and improper use of chronological ordering of multiple citations.

Response: Apologies, these formatting issues have been corrected.

11) P2, Line 21: Please add more indicative primary source references; this paper is rather Specific

Response: Apologies, we have now replaced this reference with the overarching review of Halquist et al. (2009).

12) P2, Line 30: Reference repeated

Response: Apologies, this has been corrected.

13) P3, Line 14: ": : :air and in THE laboratory: : :"

Response: Apologies, this has been corrected.

14) P4, Line 26: "now" rather than "new"

Response: Apologies, this has been corrected.

15) P4, Line 28: "than" rather than 'that"?

Response: Apologies, this has been corrected.

16) P5, Line 10: "than" rather than "that"

Response: Apologies, this has been corrected.

17) P5, Line 30: Full-stop missing after "3.2"

Response: Apologies, this has been corrected.

18) P6, Lines 14 – 16: Rewrite to facilitate ease of reading

Response: We have replaced those lines with the following:' *However we first and foremost wish to demonstrate the efficacy of using pre-defined fingerprints as they are available in the literature, or, within existing open-source software packages. The exact physical processes taking place within instrument are still the subject of considerable debate.* '

19) P8, Lines 9 – 11: Sentence is awkward, I suggest it is rewritten for clarity

Response: We have replaced those lines with the following:' *A recent study of McVay et al. (2016) presented results demonstrating sensitivity of aerosol mass and composition to processes included in a box-model model, including the addition of autoxidation mechanisms. They proposed that autoxidation might resolve some or all of measurement–model discrepancy from chamber simulations, but that this hypothesis could not be confirmed until more explicit mechanisms are established for α-pinene autoxidation(McVay et al., 2016).*'

20) P8, Line 21: "Fingerprints"

Response: Apologies, this has been corrected.

21) P9, Line 1: Repeated word - "value values"

Response: Apologies, this has been corrected.

22) P14, Tables 1 and 2 legends: Right [parenthesis missing

Response: Apologies, this has been corrected.

23) P17, Figures 4: axis labels are too small and potentially unreadable in final print, please increase the text size

Response: Apologies, this has been corrected.

24) P17, Figures 4 legends: Right [parenthesis missing

Response: Apologies, this has been corrected.

25) P19, Figure 5: Axis labels missing

Response: Apologies, this has been corrected.

26) P20, Figure 6: Axis labels missing

Response: Apologies, this has been corrected.