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Interactive comment on "Characterization of organic nitrogen in aerosols at a forest site in the southern Appalachian Mountains" *by* Xi Chen et al.

Anonymous Referee #1

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Review of Chen et al I am reviewing this paper as someone with quite a lot of experience of measuring bulk organic nitrogen but with much less expertise in organic matter characterisation. Overall I think this is a useful paper that demonstrates the potential importance of the organic nitrogen in this region and also provides some useful characterisation of the some of the organic matter in the aerosol. The provision of data on organic carbon and nitrogen together is for me particularly useful. The wide range of data does allow some inter-component relationships to be used to suggest something about organic C and N cycling, although all the correlations may not prove a causal link. The sampling and analysis is state-of-the-art for the compounds analysed and provides a high quality and useful data set. I am happy to see it published but I would

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suggest a few changes before publication.

Specific comments I do wonder if the title is really appropriate given how little of the organic nitrogen is characterised. Line 69 There is now a global model of atmospheric organic nitrogen cycling that should perhaps be referenced – Kanakidou et al 2012 Global Biogeochemical Cycles doi 10.1029/2011GB004277. Line 72 To my mind the work of Altieri cited here and their more recent paper (Altieri et al 2012 ACP 12 355703571) represent the best effort to characterise the atmospheric organic nitrogen and yet neither here or later in the paper is this work discussed. It is relevant because it identifies reduced nitrogen as a dominant component of the atmospheric organic nitrogen, yet the authors here are characterising oxidised nitrogen based organic matter. The rationale for their choice of compounds is not really explained in the section line 112-125 where I might expect it to be. Line 140 The site map needs to be in the main text not the supplementary material. Line 145-148 Given their importance from the results at this site, the authors might want to comment on ammonia sources. Line 151 – how many samples in total? I guess about 60 but it does help to know when looking at the statistical work. Line 151-3 Gonzalez-Benitez discussed the issue of semi-volatile organic nitrogen and it may be useful to at least note this, although it is very hard for most of us to sample for this. Line 221 I think “less than” should be “better than” if I understand the point Section 2.4 Please explain what the PMF is being used to investigate. The section here is a detailed description of the mathematical manipulations but it does not explain anything about the process to the non specialist. Line 317-8 How does how ozone consumption lead to a seasonal maximum? Line 337-340 For a wider audience I would suggest it is worth noting this %organic N is consistent with other data from the world beyond the USA. Line 342-344 The claimed seasonal cycle looks very small to me from the graphs. Line 349-352. The correlations are presented for each season, and that is OK although with only about 20 samples and so many variables I wonder about the statistical validity of the approach. I would therefore suggest that the equivalent correlation for the whole data set should also be presented. The observation of the correlation of WSON and WSOC is interesting and there is rather limited

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such data valuable. I also note a much stronger correlation of WSON and NH4 than NO3. This is consistent with other data (see Cape et al 2011 cited) and points along with the Altieri work above, to a key role for reduced nitrogen in WSON formation. Line 359 "source contributions" of what? presumably WSON and C Line 374-7 We have all had problems such as described here, but is it really useful to include the samples collected when local burning impacted the sampler? This is particularly relevant because throughout much of the paper the authors show they can only characterise a few percent of the WSON. Then suddenly on line 508 they say they can characterise 28% which would be very impressive but I think this is for these local burning episodes and so by including this high percentage the authors may mislead readers into thinking as a community we are beginning to be able to characterise quite a lot of the WSON. This is also relevant to line 587 and the abstract. As the authors note in line 552 they and the rest of us have yet to be able to characterise very much of this material Line 434-5 Given how small a percentage of WSON appears to be made up of N containing organosulphate compounds, I'm not sure its correct to make the statement "which reflected... to WSON" here. Line 440 group of ORGANIC compounds Line 447 is 6-9% (which is what I think your report) really "a substantial proportion"? Line 446-453 Here and elsewhere I think the authors need to be careful about interpreting correlations as showing causal links. Line 562-565 I think the authors conclusions are valid for the material they have characterised, but that does not necessarily mean that all of the organic aerosol has been similarly aged. Line 581-3 I do not understand what the sentence starting "PMF analysis" means. I am not really sure that figure 5 and 6 add much to manuscript

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