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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The nitrogen budget of laboratory-simulated western U.S. wildfires during the FIREX 2016 FireLab study" *by* James M. Roberts et al.

Anonymous Referee #1

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General comments:

The authors present an overview of the N-budget during the FIREX campaign, bringing together datasets from a variety of instruments and techniques (outlines in Table 1) in already peer-reviewed articles of FIREX work. This analysis is certainly a great contribution to the field and specifically to the science of reactive nitrogen budgets from forest fires. The methods used are state-of-the-art, including the instrumentation used for acquiring the data during the campaign in addition to the data analysis, including PMF. I command the authors on putting this data set together and anticipate this contribution, upon revision, to be a go-to paper for the atmospheric organic-N and inorganic-N fate

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Discussion paper

from biomass burning, well fitting within the scope of Atmos. Chem. Phys.

My most significant revision recommendation is related to the writing and to the presentation of the research. It is difficult to read the manuscript as the information is presented in no specific order and without pertinent subsections. Lots of information is relayed, but it is unclear what the scientists set out to do in the first place. My specific feedback is below to improve the manuscrupt.

Scientific feedback:

It is unclear to me exactly what the conclusion of the work is. Is it that N mass balance was achieved in this study (stated in lines 298-299 – which seems to be a particularly important finding in my opinion, even one to be highlighted as the title, or at least feature predominantly in the abstract)? Is that Nr can/cannot be predicted (mentioned on lines 337-338)? Is it that the dominant molecules of Nr were identified? Is it that an estimated Nr/C ratio of 0.37% can be recommended for modeling (lines 450-452)? It seems like the authors touch upon each one of these questions but the support for these conclusions can be better communicated. In my opinion, they are all important questions for the field which this paper nicely addresses, but currently in a rather disordered fashion. My advice is to use subsections addressing each one of the scientific questions listed (and others as the authors see fit) and present the data in a logical way: (1) state the hypothesis and research question; (2) show the appropriate figure and state the results; (3) discuss the implications of the work.

The Comb-N, HT-N and LT-N factors represent different classes of Nr emissions during biomass burning. What was the big picture goal here? Was it to know that for example when NH3 is detected in a plume, that temperatures related to LT-N are taking place at the origin for the fire? In other words, are these identified chemical markers used to estimate the temperature and burning stage of the fire? If so, then the authors should add this goal to their abstract, highlight this result in their conclusion and devote a titled sub-section to this analysis.

Which research question was being addressed by comparing chaparral fuels within this study? Is the information distracting from the other main messages? Is it necessary to include the analysis related to the chaparral fuels in this paper?

Why place an emphasis on Batch 1 and Batch 2 (including Table 2) if the conclusions are that the same factors were obtained irrespective of which "Bacth was used" (explained in lines 367-368)? Is it necessary to describe both of these PMF analysis? I would argue (although happy to be convinced otherwise) that this description is not necessary for the analysis presented in this manuscript. Only one batch could be described (would also affect Table 2) and a simple one-sentence mention that the same factors were obtained with and without inclusion of CO2, CO and Nr.

I'm curious to know whether there is a "time" component expected for the Nr budget. In other words, would the Nr species evolve over time away from a plume of biomass burning? If so, how? There is mention of flame chemistry on lines 348-362 and Figure 8 (where there is a clear behavior of multiple generation production). For example, the brown carbon properties of biomass burning aerosols respond to heterogeneous oxidation (see (Browne et al., 2019)). For example, are the organic-nitrogen compounds evolving from amines to amides to isocyanates (see (Borduas et al., 2016))?

HNO3 is mentioned in the text (lines 87; 94-98; 300-301) but it wasn't clear where its discussion fits within the topic of Nr. Could the authors specify the significance and consider adding a subsection on HNO3?

Lines 281-289: Is the goal of this section to update MCE values with this work, or to compare the MCE values to the literature? Which values are to be used in further modelling for instance?

I'm curious about the atmospheric implications of yak dung (lines 307-310).

Based on the results from FIREX, would the authors continue to recommend acetonitrile (line 311) as an adequate biomass burning tracer?

Lines 322-323: HNCO and HCN are organic compounds (of which the definition is any molecule which contain at least one carbon and one hydrogen) and shouldn't be included in the "inorganic N" category and discussion.

Presentation feedback:

The best way to improve the manuscript is by structuring the text. The results are interesting but are buried in paragraphs with a lack of order. Every paragraph could have subsections identifying the main message of the results.

In general, the structure of a paragraph starts with a topic sentence about what the paragraph will describe. I think being attentive to that structure could really help bring flow to this manuscript. (For example, one could also avoid starting paragraphs by pointing to a Figure (lines 271-272)).

The scientific research questions could be better identified and articulated. For example, the abstract starts with stating the method, without giving context, the research question and the hypothesis. A short revision here could help the reader situate the study.

Lines 84-87 cites a long list of references. I would argue that this list is not so useful unless each study is briefly described and the main message is communicated.

Lines 164-166: would be great to show this data (in the suppl info)

Lines 208-211: could give an example of the calculation

Table 1 is a great summary of the techniques and compounds included in this study. However, I was a little confused about the meaning of the "references" column. The references from where the data from FIREX was published should be identified separately. I think there are currently 5 publications from FIREX which this study correctly references. What is the meaning of the references prior to 2016? Are they instrument references? It would be great to add further details here as well as provide all the acronyms of the instruments in the caption.

Figure 1: The equations aren't mass/atom balanced. For example, HCN → O, OH → CN is a misleading representation of the reaction. One can use curve arrows to show the intermediates with their own products. Or one can add all the products next to the specie. The chemical equations should be mass balances in any case. In addition, it would also be useful to denote all radical species (either with a dot or another way). I would avoid the (s) notation at it is typically reserved for the solid phase. Perhaps H2O + surface could be clearer.

Lines 386-396: an important message! So temperature is a better predictor than MCE? Interesting!

Technical comments:

Lines 37-43: This sentence is 7 lines long and could be broken down into 3-4 sentences.

Line 44: define chaparral fuels

Line 76: use arrow instead of "=>"

Lines 115-123: best to use the present tense in this paragraph rather than future tense (which reads more a like a proposal)

Line 256: Is the "emitted carbon" in the gas phase or the particle phase?

Figure 3: the x-axis in 103 ppmv-sec was a little misleading. Took me a few minutes to understand/see that there was a factor of 1000 between the y-axis and the x-axis. Could be worth highlighting these units differently.

Figure 10: top panel is arguably redundant and could be removed. (same for the equivalent figure in the suppl. Infor)

Table 3: Could be better represented as a graph?

References:

Borduas, N., Abbatt, J. P. D., Murphy, J. G., So, S. and da Silva, G.: Gas-Phase Mechanisms of the Reactions of Reduced Organic Nitrogen Compounds with OH Radicals, Environ. Sci. Technol., 50(21), 11723–11734, doi:10.1021/acs.est.6b03797, 2016.

Browne, E. C., Zhang, X., Franklin, J. P., Ridley, K. J., Kirchstetter, T. W., Wilson, K. R., Cappa, C. D. and Kroll, J. H.: Effect of heterogeneous oxidative aging on light absorption by biomass burning organic aerosol, Aerosol Sci. Technol., 0(0), 1–12, doi:10.1080/02786826.2019.1599321, 2019.

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