

Aruffo et al. reports the use of total peroxy nitrates (Σ PNs) measured by thermal-dissociation laser-induced fluorescence along with other trace gases typically associated with biomass burning (CO, HCN, and CH_3CN) to identify biomass burning plumes during the BORTAS campaign. The authors compare their biomass burning thresholds they determined from their Σ PNs measurements with thresholds published in prior studies. Finally, they utilize an artificial recurrent neural network (ANN) to indicate their measurements correctly identify biomass burning plumes.

While the use of Σ PNs or the different speciated acyl peroxy nitrates (e.g., PAN) to identify biomass is not novel (e.g., Alvarado et al., 2010; Fischer et al., 2011; Tereszchuk et al., 2013), the introduction of the thresholds needed for the Σ PNs measurements to indicate biomass burning plumes is an important aspect of this paper to assist the community in better classifying different plumes. Similarly, the use of ANN to identify air mass classification and chemical history is beneficial for the community.

However, for this paper to be published in AMT, further discussion, description, and limitations of their techniques need to be addressed—see general and specific comments. Currently, many of the sections fall short in providing the community the details necessary to apply these techniques to the community's own research in identification of air mass history to better understand the emissions and chemistry that impacted the air mass.

General Comments

- 1) A large fraction of the paper addresses prior methods to identify biomass burning plumes, and the authors compare their method (using Σ PNs) with the prior methods. However, the authors do not address which method, or methods, is best in identifying biomass burning plumes. For example, in Table 3, some methods that agree well with the Σ PNs method also produce potentially false positive identification of biomass burning plumes during other research flights. What are the limitations of these various methods? Which of these methods (or combination of methods) are the most robust in identifying biomass burning plumes in a variety of conditions (for different ages of plume, for different altitudes of plume, for different intensities of fire, and et cetera)?
- 2) A discussion of the limitations of the Σ PNs would be extremely beneficial. The authors noted that the results are most robust for pressures less than 750 hPa; however, that leaves the reader to wonder how much of that is due to the thermal decomposition lifetime of Σ PNs and the chemical age of the plumes intercepted during the BORTAS campaign. A figure that shows approximate chemical age (or other form of ages) in the main text or part of supplement, especially versus pressure (or temperature), would improve the discussions throughout the paper. Also, how much does the thermal lifetime of Σ PNs impact your interpretation of Figure 3?

For example, in Figure 1, at low altitudes (pressure less than 800 hPa), the measurements of CO and CH_3CN show high correlation with each other whereas Σ PNs shows no correlation. At these high pressures, the lifetime of Σ PNs is less than 1 hour. Is this due to aged biomass burning plume that have stayed in the boundary layer where Σ PNs have

thermally decomposed? Or is this due to aged biomass burning plumes that have been transported into the planetary boundary layer, causing the Σ PNs to decompose?

Also, a large fraction of biomass burning plumes stay in the planetary boundary layer (e.g., Gonzi et al., 2015), where the Σ PNs lifetime is extremely short (less than 1 hour). With that in mind, at what point does the use of Σ PNs as a marker of biomass burning plume not work? How much of the air higher than 2000 m a.s.l versus lower than 2000 m a.s.l. was impacted by biomass burning (Page 15, line 12)?

Finally, there are other types of air masses that maybe classified as biomass burning plumes while using Σ PNs measurements whereas these air masses originate from a different source or combination of sources. For example, Apel et al. (2015) observed and modeled production of PAN downwind of an air mass influenced by both biomass burning and lightning NO_x emissions. Similarly, Alvarado et al. (2010) noted that many of the biomass burning plumes observed during ARCTAS were influenced by thunderstorms. How well can your thresholds disentangle these influences?

- 3) What happens if 99th percentile above the background of each flight is used instead of using the value from one flight? As another way to consider it, what would happen if the 6σ above background for B625 is used? Just wondering why you did not use these two methods the same way (either for all research flights or for the background research flight)?
- 4) In Figure 4, you show the back trajectories for the air masses sampled. However, between 120° and 90°W, it is hard to tell which trajectories are most important for your analysis since the high and low pressure back trajectories overlap. Also, it appears that there are numerous biomass burning events in that region between 120° and 90°W. How did these biomass burning events impact this study?
- 5) The authors barely describe the ANN and the results. Without a more in-depth description of how the authors initialized and ran the model, the section provides minimal benefit to the community.

Also, what is the benefit of using ANN if a researcher has access to measurements? Are there other parameters that could be used to improve the performance of ANN (e.g., chemical or transport age of plume)? How well does the chemistry, biomass burning intensity, and/or meteorology need to be known to calculate meaningful results? The authors noted that ANN is ideal since it is capable of simulating non-linear relationships, which causes me to wonder this. It was unclear why the addition of pressure led to instances of causing further disagreement between modeled results and measurements.

Specific comments

- 1) In general, please double check your grammar. For example, Page 7, line 18 – 19, you are missing a noun in front of are summarized. What is summarized? There are other instances where grammar needs to be corrected throughout the paper.
- 2) Be consistent when the names and chemical formulas are introduced. Page 3, line 19, acetonitrile is the first time it is defined, however, the chemical formula is used in numerous lines prior to that (Page 3, line 8; Page 3, line 12, et cetera).
- 3) Page 4, line 19 – 20: was HNO_3 measured by your instrument as well?
- 4) Page 4, line 23 – 25: Please better explain how the ΣPNs and ΣANs are determined.
- 5) Page 6, line 6: Convert seconds to either UTC hours and/or local time. Same with Figure 1 on Page 6 and Page 14, line 4 – 5.
- 6) Page 7, line 18: I wondered what mixing ratio ΣPNs corresponded to for the 6σ threshold.
- 7) Page 8, Table 3: Please define what the grey areas are in the table in the table caption. Also, what are the units for $\Sigma\text{PN} > 0.418$? Approximate values for the 6σ would be nice. Finally, please be consistent about the Lewis et al. definition. There are instances where the definition is flipped from < 200 to > 200 .
- 8) Page 14, line 18: Please capitalize Lagrangian.
- 9) Page 15, Figure 4: Do you have any measurements of the intensity and emission heights for all these biomass burning events?
- 10) Page 16, line 11 – 13: How does deposition (dry or wet) impact the analysis of NO_y chemical ages for air sampled in boundary layer?
- 11) Page 22, line 17 – 18: Line reads that air masses located at lower pressures are clean of biomass burning and originated from northern Canada. However, I thought that Figure 4 indicated that the air masses at lower pressures were impacted by biomass burning and originated from northwestern United States/southwestern Canada. Please clarify.

References

Alvarado, M. J., Logan, J. A., Mao, J., Apel, E., Riemer, D., Blake, D., Cohen, R. C., Min, K.-E., Perring, A. E., Browne, E. C., Wooldridge, P. J., Diskin, G. S., Sachse, G. W., Fuelberg, H., Sessions, W. R., Harrigan, D. L., Huey, G., Liao, J., Case-Hanks, A., Jimenez, J. L., Cubison, M. J., Vay, S. A., Weinheimer, A. J., Knapp, D. J., Montzka, D. D., Flocke, F. M., Pollack, I. B., Wennberg, P. O., Kurten, A., Crouse, J., Clair, J. M. St., Wisthaler, A., Mikoviny, T., Yantosca, R. M., Carouge, C. C. and Le Sager, P.: Nitrogen oxides and PAN in plumes from boreal fires during ARCTAS-B and their impact on ozone: an integrated analysis of aircraft and satellite observations, *Atmos. Chem. Phys.*, 10, 9739–9760, doi:10.5194/acp-10-9739-2010, 2010.

Apel, E. C., Hornbrook, R. S., Hills, A. J., Blake, N. J., Barth, M. C., Weinheimer, A., Cantrell, C., Rutledge, S. A., Basarab, B., Crawford, J., Diskin, G., Homeyer, C. R., Campos, T., Flocke, F., Fried, A., Blake, D. R., Brune, W., Pollack, I., Peischl, J., Ryerson, T., Wennberg, P. O., Crouse, J. D., Wisthaler, A., Mikoviny, T., Huey, G., Heikes, B., O’Sullivan, D. and Riemer, D. D.: Upper tropospheric ozone production from lightning NO_x-impacted convection: Smoke ingestion case study from the DC3 campaign, *J. Geophys. Res. Atmos.*, 120, 2505–2523, doi:10.1002/2014JD022121, 2015.

Fischer, E. V., Jaffe, D. A. and Weatherhead, E. C.: Free tropospheric peroxyacetyl nitrate (PAN) and ozone at Mount Bachelor: potential causes of variability and timescale for trend detection, *Atmos. Chem. Phys.*, 11, 5641–5654, doi:10.5194/acp-11-5641-2011, 2011.

Gonzi, S., Palmer, P. I., Paugam, R., Wooster, M. and Deeter, M. N.: Quantifying pyroconvective injection heights using observations of fire energy: sensitivity of spaceborne observations of carbon monoxide, *Atmos. Chem. Phys.*, 15, 4339–4355, doi:10.5194/acp-15-4339-2015, 2015.

Tereszchuk, K. A., González Abad, G., Clerbaux, C., Hadji-Lazaro, J., Hurtmans, D., Coheur, P.-F. and Bernath, P. F.: ACE-FTS observations of pyrogenic trace species in boreal biomass burning plumes during BORTAS, *Atmos. Chem. Phys.*, 13, 4529–4541, doi:10.5194/acp-13-4529-2013, 2013.