Interactive comment on “Impact of Biomass Burning emission on total peroxy nitrates: fire plume identification during the BORTAS campaign” by Eleonora Aruffo et al.

Anonymous Referee #2

Received and published: 5 May 2016

This paper uses TD-LIF measurements of total peroxy nitrates (PNs) during the BORTAS campaign to identify biomass burning plumes. Two methods – a threshold defined by the 99th percentile of total PN concentration on the background flight B625, and one based on the six sigma deviation from the mean of background periods in each flight – are used to identify biomass burning plumes and are compared to other methods that have been used in the literature. A couple high CO, high CH3CN, but low total PN periods in flight B623 are further analyzed using HYSPLIT back-trajectories, which show that these periods generally include air masses near the surface (pressure greater than 750 hPa) and are not consistent with the fire locations during the period. The paper then uses a neural network approach to try to simulate the HCN and total PN concentrations in two flights, and shows that adding the pressure significantly improves the model fit.

Major Comments:

The use of the TD-LIF total PN measurement to identify biomass burning plumes is unique, and could be useful for campaigns and locations where other species are not available. However, I have some serious concerns about this paper.

First, PNs are not long-lived tracer species, nor are they unique to biomass burning. PNs are formed in the atmosphere due to the chemistry of reactive organic species and NOx, which can come from not only by biomass burning but from a variety of anthropogenic pollution sources and the interaction of anthropogenic NOx with biogenic VOCs. So it is unclear if this method would work to identify biomass burning plumes outside of the BORTAS dataset, where it is more likely that the measured PNs are due to biomass burning than to anthropogenic pollution.

Second, while pressure might be a useful discriminant of different air masses in this study, it is unclear why this would be generally true. In this dataset, the different pressures line up with different origins as demonstrated by the HYSPLIT back-trajectories, but that argues more for using back-trajectory models to identify different air masses than the use of pressure itself. In addition, if the high-pressure air masses in flight B623 are not from biomass burning, why is CH3CN elevated during these periods in Figure 1, and why is CH3CN almost perfectly correlated with CO? What is the source of this CH3CN if not biomass burning? Isn’t it more likely that near the surface, the PNs thermally decompose, but the air mass is still influenced by biomass burning as indicated by the CH3CN?

Third, the methods in this paper are generally unclear. The detection limits, known biases, etc. for the measurements made in BORTAS are not included here. The method used to identify “background” portions of each flight for the 6-sigma method is not described. The meteorology used to drive the HYSPLIT back-trajectories is not well described.
described. The neural network model is not described.

Fourth, the comparison with other plume identification criteria is off, as it uses techniques even when the required data is not available. For example, in Table 3 Alvarado et al. is reported to result in 1.3% of flight B619b being identified as from biomass burning, but notes that only CO was available. That's inconsistent with the method used in that paper, which required correlation of CO with HCN or CH3CN, so no value should be reported for those conditions. Similar stricter constraints should be used in Table 3 for other studies.

Fifth, the paper is not really about an atmospheric measurement technique. TD-LIF is used, but not described, here, and using the total PN measurements to identify smoke plumes is not by itself a measurement technique. Furthermore, there is little science in the paper other than the comparison of this new method with previous methods that have been used to identify smoke plumes, but in those previous studies identifying the smoke plumes was the first part of a larger scientific study rather than an end in itself.

So based on these I do not feel I can support publication of this paper in AMT. However, major revisions might make it acceptable, so I detail other more minor concerns and typos that could also be fixed in a revision below.

Minor Comments:
P1, L26: “statistical and percentile methods” is not clear – please be more specific about your techniques here.
P3, L14-15: Please describe how these background periods were identified, and make clear if you used the same approach to identify background periods for your PN-based technique.
P4, L22-25: This description of TD-LIF is way too brief given the whole paper is written around it. How selective is it in separating PNs, ANs, and HNO3? What is the accuracy, precision, and detection limit? What are the known biases or problems?
P5, L14-15: The accuracy and detection limit for these instruments should be included in Table 1, not just referenced.
P6, L8: Furfural reacts too fast to be considered a “tracer” – you mean these species are known to be emitted by biomass burning.
P7, Table 2: Make clear that the 6 sigma refers to 6 sigma beyond the mean of the background concentration, not 6 sigma from the mean of the whole dataset.
P7, L12: Again, how do you identify background to use the 6 sigma method?
P14, L18: Please describe the meteorology you used to drive HYSPLIT – that is extremely important to the results.
P16, L8: This is the photochemical age, to be clear, and is not necessarily the same as the chronological age. Please make that clear.
P17, L3: I'm not sure why you don't plot flight B623 here, with CO versus CH3CN. I think that would show an extremely strong correlation.
P17, L7-10: We need a lot more details on the modeling to assess what you have done here.
P18, L15: A scatter plot of the measured versus simulated total PN mixing ratio for each flight would be useful.
P18, L20-L23: Add equations explaining these metrics – the text explanation is not that clear.
P19, L4: Wouldn't systematic errors show up in the FB as well, so this suggests only an increase in random errors?
P20, L4-6 and P21, L4-6: These captions are all messed up, and I'm surprised it wasn't caught earlier before publication in AMTD. They need to be fixed.

Typos:
P1, L31: Total PNs and HCN were not used as input parameters – this should be NO and CH3CN.

P2, L29-30: Remove “the” before “40%” and “20%”, and make it “particle phase” not “particles phase”

P2, L32: “Analysis of the chemistry”

P3, L5: Instead of levels, use the more specific “volume mixing ratios”

P4, L9: Cut “of”

P5, Table 1: Subscript “RONO2”

P7, L2: I’d suggest “used” instead of “introduced as tracers”

P13, Figure 2: Use different shapes in addition to different colors, as some readers may be red-green color blind. Also, the use of a green dot in the center of the red circle and vice versa is confusing.

P15, L5: FLAMBE is an emission dataset that uses satellite data – you might want to refer to the satellite that actually identified the fires instead.

P16, L4-5: This phrasing is very hard to understand.

P18, L1: “impacted”, not “interested”

P19, L6-8: Lowercase “model”, add period after “indices”