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Interactive comment on "Regional and intercontinental pollution signatures on modeled and measured PAN at northern mid-latitude mountain sites" *by* Arlene M. Fiore et al.

Anonymous Referee #2

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Fiore et al. very nicely highlights the importance of understanding and simulating PAN distributions to understand tropospheric ozone distributions. However, I was disappointed that there were not more specific results on the causes of model-measurement discrepancies. This is a very clearly written paper, though rather long relative to the new results presented. Previous work is referenced well. The figures clearly illustrate the points being made. One aspect of the paper that seems new to me is the use of the long-term mountaintop measurements for model evaluations and this is a nice presentation of their value.

I understand the interest and motivation to make use of the HTAP model simulations,

however, it seems to me there are a lot of limitations in using these simulations to understand PAN. The HTAP1 simulations did not use consistent emissions inventories across models, so it is very difficult to distinguish model chemistry and transport differences from purely emissions differences (in NOx and VOCs). The HTAP2 simulations, performed with more modern models, specified the emissions inventories to use for all simulations, and therefore might yield more conclusive results. However, in my experience, the simulation of PAN seems to be highly dependent on the BL dynamics of the model, and fine-scale chemistry, so it is difficult to see how much can be learned from monthly mean outputs, even with many models. It is my opinion that much more could be learned by the factors controlling PAN distributions using a single model with high time resolution output and comparison to the numerous aircraft measurements, as well as focused ground-based campaigns, that are available.

Previous studies have clearly illustrated that the chemical mechanism of a model has a big impact on PAN - not only the Emmerson and Evans studied referenced many times in this paper, but also Knote et al., Atmos. Environ., 2015. Previous work has also shown large multi-model differences, even when using the same emissions, in 3D models (e.g., Arnold et al., 2015; Emmons et al., 2015). So these points in this paper are not new.

Another concern I have is with the procedure for determining source attribution through emissions perturbations, which has been accepted by HTAP as standard procedure. The non-linearity of the chemistry in PAN and ozone formation will affect even relatively small perturbations such as 20% used here (see Butler et al., GMD discussions, 2018, and references therein). It seems to me that the large scatter shown in Figures 7 and 8 might largely be due to the non-linear chemistry in PAN formation on top of the differences in emissions and chemical mechanisms. Also, in Figs.7&8, what is the significance of the dashed line at 1.0 for the PAN ratio? Doesn't the r value correspond to a 1:1 line between y and x axes?

I am not entirely sure what to recommend for this paper. In its present state, it does

not seem to me to have enough new results to justify publication. Just as it is not really informative to evaluate ozone simulations without evaluating the precursors, perhaps more could be learned about the performance of the models if there were simultaneous evaluations of NOx and PAN-precursor VOCs to indicate why some models disagree so greatly with observations. I think the paper would also be strengthened by condensing the paper to focus on the really new results, and with less space used on the confirmation of previous findings.

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