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Atmospheric Chemistry and Physics
Discussions

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 #1

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This paper attempts to understand the seasonal scale processes controlling the concentration of PAN at 5 mountain top sites in the northern mid-latitudes and to then use this as a tool to understand the processes controlling tropospheric O3. It attempts to do this through the framework of the HTAP1 model inter-comparison exercise, a Lagrangian modelling approach and the measurements made at the sites.

The paper is relatively long and often feels a bit winding. It has a large number of authors and this committee approach is obvious in the paper. For all the effort that has gone into the paper my major concern is that it's not obvious what has been learnt and whether it is new and / or interesting? From the abstract the major conclusions

seem to be: there are some differences between the attribution of the Eulerian and the Lagrangian approaches for a single site in Switzerland; VOCs are important for PAN production; ozone and PAN chemistries are qualitatively linked in models. I don't think any of these are particularly novel. I think however, that with some extra work this paper could provide some interesting insights.

These model simulations are old. They were run over a decade ago. How useful is this exercise if the models have now changed substantially? Has anything been learnt over the last decade which should be considered here. I don't think the authors can just ignore this issue.

One concern is that the paper investigates the importance of a number of issues in determining the processing controlling PAN for these mountaintop sites, but it makes little effort to then translate this understanding onto a larger spatial scale. How does PAN from EU VOCs make its way round the world? How strong is the PAN O3 relationship in different models globally? The paper discusses essentially the two sites that it has (one in the US and the multiple ones in almost exactly the same space in the Alps) and then seems to stop abruptly without widening its thinking to a more global or even hemispheric scale. What has been learnt which is more globally or regionally explicable? It would seem like a sensible next step to investigate the wider implications of the perturbation studies.

I'm not sure of the usefulness of the Lagrangian approach in this paper. PAN's lifetime varies significantly with temperature, and whether it being produced or lost depending upon a complex set of chemical reactions. The Langrangian approach may have some usefulness when looking at relatively short-lived, chemically simple tracers but it is not obvious that it has value when looking at PAN. The methodology is not described in any details and issues to do with subgrid processes (convection and boundary layer mixing) are not discussed at all. The authors need to show better its usefulness to PAN and then if they continue to use the measurements they should do something more useful with these calculatins. At the moment they show that the Lagrangian model gives different results than the Eulerian models and then appear to provide a very handwaving route to get to some form of ‘consistency’ between the different modelling framworks. At this point they essentially then proceed ignoring the issues raised by the Lagrangian method. The paper would come to the same conclusions without Langrangian section. I would therefore suggest it was removed.

The authors argue that there needs to be more measurements made from mountain top sites. I’m not convinced. Especially in the Alps there is a raft of data from these sites. It may be that making more mountain top measurements from other locations on other continents would be useful, but the authors provide little evidence for that. Where would they think that these measurements should be made? Himalayas, Urals, White Mountains, Rockies? Presumably from their model simulations they could make some suggests.

Where there are interesting results (notably Figures 6 and 9) the authors don’t really delve into the details. What explains the difference in AVOC emissions between the models? There is almost a Given the authors they should be able to work this out for some of the model? Is it that they are using different emission inventories or making different choices about which species to include? Where are models with more or less chemical detail in this picture? Is there some rational for models that don’t fit on the line? For example, there are two models which have almost the same EU AVOC emissions $\sim$22 Tg C a-1 but have very differing PAN responses. Can we understand these differences in terms of the model chemistry or meteorology? There is lots of interesting things that could be done here but the authors appear have performed a rather perfunctory analysis.

In conclusion I am rather conflicted about what to say. It looks like this paper has taken a long time to produce and a lot of work has gone into this paper probably over the last decade. I am however a bit confused by what has come out of this. I would suggest that the authors think about what the key points are, and then attempt to develop those further. There are plenty of people involved in this publication. It should be possible

to get something out of this. However, at the moment I don’t feel the paper detailed or informative to make it suitable for publication.

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