Atmos. Chem. Phys. Discuss., 13, C9515–C9518, 2013
www.atmos-chem-phys-discuss.net/13/C9515/2013/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric peroxyacetyl nitrate (PAN): a global budget and source attribution" *by* E. V. Fischer et al.

G.A. Folberth (Referee)

gerd.folberth@metoffice.gov.uk

Received and published: 29 November 2013

GENERAL COMMENTS:

The manuscript discusses the global budget of PAN, an important reservoir for NOx in the troposphere and a chemical species central to tropospheric ozone production. An extensive source attribution for PAN is also included. The authors apply a typical modelling approach using a well established and tested 3D global chemistry-transport model (GEOS-Chem). In addition to deriving the PAN budget and identifying and quantifying the major sources of PAN the work includes further innovation in two ways: 1) by improving the GEOS-Chem representation of the chemistry of PAN and its precursors synthesizing recent advancement into the model and 2) by creating a new compre-

hensive database of PAN observational data for model evaluation. The global PAN distribution in the GEOS-Chem model is evaluated against this newly created dataset.

While maybe not breaking ground on all fronts, in my opinion this paper represents a solid and most useful piece of science. It is well within the scope of ACP, overall well written and presented and is positively worth publishing.

I believe there really isn't much to be criticised about this manuscript. Abstract and introduction are useful, existing literature is comprehensively cited and the results are presented and discussed clearly and logically. Tables and figures are prepared and used well, are meaningful and are helpful in discussing the science. The conclusions are concise and down to the point. The language is clear and the thread is (mostly) consistent.

I do not always agree with the choice of figures selected for the main text and the supplement (see specific comments) but I appreciate that it is sometimes difficult to bring down the large amount of visual data to the size of a meaningful manuscript. At times the discussion strays away from the specific result being discussed extending onto closely (and not so closely) related results but always remains topical and factual. It is debateable whether this is only a question of writing style or may be considered a more serious weakness; in any case, I do not believe it diminishes the manuscript in any significant way.

SPECIFIC COMMENTS

p. 26,851; l. 5-7: The paper states that the "finer horizontal resolution [in the GEOS-Chem sensitivity experiment] produces 10-20% more PAN". Unfortunately, this finding is not explored any further. It would be interesting to know to what process the increased PAN production is owed (better resolution of (pyro-)convection, more accurate distribution of fire emissions, etc.).

p. 26,854; l. 15 ff.: Should figures from the supplement be made an integral part of the

main text? In other words, why not move the figure from the supplement to the main text when it is discussed there, anyway. Either include the figure in the main text or remove the discussion from the manuscript. The reader should not feel it necessary to turn to the supplement in order to follow the main line of discussion.

p. 26,855; l. 5: It would be helpful, I think, to point out that the austral-Spring features discussed in Figure 2 correspond to the SON seasonal mean plots.

p. 26,856; l. 13: Again, if it is an important result/figure then it should be included in the main text rather than putting it in the supplement.

p. 26,856; l. 8 ff.: Usually, by applying an annihilation perturbation one risks forcing the model into a non-linear response (as is, indeed, argued a few lines below in connection with the isoprene sensitivity scenario). I have two related questions to this: 1) why has the annihilation perturbation scenario been chosen for most of the sensitivity experiments (e.g., to increase the signal-to-noise ratio perhaps) and 2) has there been made an attempt to analyse whether the response in these annihilation perturbation experiments are non-linear (e.g., by repeating one of these experiments with a 20% perturbation and then up-scaling the response to 100%; a comparison between this and the annihilation experiment should reveal non-linearities).

p. 26,860; l. 20: With the new chemistry mechanism in GEOS-Chem, which includes an OH-recycling mechanism, it would be interesting to quantify the impact of OH-recycling on PAN formation. This should be easily done by one further sensitivity experiment.

p. 26,883; Table 1: I do not quite understand why the terpne lifetime is longer than that of isoprene; I would have thought that this generally was the other way round. Is this because of the lumped nature of the species?

p. 26,889; Figure 5: I really like this figure and the way it presents the connection between PAN and its precursor species.

SMALL STUFF AND TYPOS:

p. 26,846; l. 12: “both” –> “all”

p. 26,850; l. 3: ...directly to PAN and HNO3, respectively. (comma after HNO3).

p. 26,857; l. 18: remove one “that” from sentence.

I have checked the references against the citations in the text and found a few inconsistencies worthwhile checking:

“Beine et al., 2000” is in the references but does not seem to be cited in the text.

“Bottenheim et al., 1994” is in the references but does not seem to be cited in the text.

“Lurmann et al., 1986” is in the references but does not seem to be cited in the text.

“Val Martin et al., 2008” is in the references but does not seem to be cited in the text.

“Worthy et al., 1994” is in the references but does not seem to be cited in the text.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 26841, 2013.