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Comment on acp-2020-1266

Anonymous Referee #2

Referee comment on "Opinion: Papers that shaped tropospheric chemistry" by Paul S. Monks et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1266-RC2>, 2021

Review of the paper by Monks et al entitled "Opinion: Papers that shaped Tropospheric Chemistry".

This wonderful paper represents a very important contribution because it shows the evolution of an important research field until year 2010. It will provide to the new generations of students and scientists an excellent historical introduction that will allow them to realize how the concepts have evolved and how science progresses. It is particularly interesting to understand that progress in atmospheric chemistry resulted from the complementarity between laboratory experiments, field measurements, data analysis and modeling.

I like the paper very much. I will highlight its strengths but I will also make some suggestions to make the paper somewhat more inclusive. I find the paper a bit unbalanced towards process studies and laboratory work (which should be highlighted as they are) at the expense of field work and modeling.

Strength of the Paper

The paper covers a lot of material and reports a lot of important results that shaped the field of atmospheric chemistry. A lot of bibliographical material is provided. Several graphs and tables are very interesting including, for example, the documents by sources and year and by country. The two tables with the top 10 cited papers are useful.

The grouping of 19 topics presented in Section 1.2 is excellent.

The discussion about how the papers were selected and the limitations to this approach is very good. It shows that the methodology does not provide an absolute standard and that work not included in this list can also be excellent.

The paper is well written and easy to read. I am sure that people starting their career will like to read the document.

Suggestions.

As I said above, I find the paper to be a bit unbalanced towards fundamental processes at the expense of more global and regional investigations. This could be easily addressed by adding some references to a few important observational and modeling accomplishments. Sometimes, big advances related, for example, to field campaigns are not presented as a single paper, but are dispersed in several papers, and so none of them has a highest citation rate. For example, a major accomplishment before 2010 has been the organization of several airborne campaigns. Not only the ER-2 measurements of the ClO/O₃ reactions have been key for the ozone hole question, but also the numerous airborne field campaigns in the 1980's and 1990's (many NASA field campaigns) that have provided insight on global tropospheric chemistry, and in particular on questions related to the oxidation capacity of the atmosphere.

A lot has been learned from field campaigns (such as PEM, Trace P, ACE, and several others) and so perhaps, the authors could mention large projects that have provided fundamental understanding of the functioning of the atmosphere at the global and regional scales. Other topics that illustrate major advances:

Field campaigns to assess NO_x produced by lightning and the importance of thunderstorms

Production of methane by rice paddies, particularly in Asia

Monitoring of methane and carbon monoxide by international networks

Systematic observations of ozone and CO from commercial aircraft

Perhaps a bit more should be said about space observations. Much progress has been done before year 2010. The measurement of CO from the Space Shuttle gave a first global view of the distribution of this species and highlighted the role of biomass burning, particularly in the tropics. Beside GOME, and SCIAMACHY, one should perhaps refer to other important satellites and instruments such as OMI, MOPITT, MODIS, TES, etc. since they provided again a global view that was very useful to understand global budgets and the role of transport.

Finally, the Section on modeling is a bit weak. Models have been key for the interpreting observations, analyzing the role of chemical processes and projecting future changes in the atmospheric composition. The first relatively detailed 3-D model of the troposphere (although a bit intermediate) were developed in Mainz (MONGUNTIA), in Brussels (IMAGES) and at Livermore (GRANTOUR). They were followed by major efforts to develop really comprehensive chemical transport models (GeosChem at Harvard, MOZART at NCAR, Uni. Of Oslo model, and MESSY in Mainz, CHASER in Japan, etc.

For stratospheric work, which is also mentioned in the paper, 2-D models such as the model of Garcia and Solomon have been a key tool to implement the fundamental concepts of the stratospheric residual circulation (and related meridional transport) resulting from planetary wave breaking presented by Holton and McIntyre, and to assess how the residual circulation could explain the meridional transport of ozone and other tracers in the middle atmosphere. Several 3-D stratospheric models were also developed for the stratosphere. One of the first ones was developed at MIT by Cunnold, Aleya and Prinn and at GFDL by Mahlman. Some more advanced models include WACCM developed at NCAR

Nothing is said about the first attempts to develop inverse modeling techniques and chemical data assimilation methodologies, which were initiated in the late 1990s.

I do not want to provide a "list of what is missing" since the goal of the paper is to highlight bthe most cited papers, but I am making some general points that perhaps would be welcome, and I am not asking to the authors to necessarily including all of them in the text. These are just constructive suggestions.

Some minor remarks on the text

Around line 109: How many responses were provided in response your solicitation of the scientific community?

Line 320; Can you add a reference and perhaps an estimate of the uncertainty about the

-1.1 Wm⁻² ?

Figure 4 looks dark to me and low quality (it may be my printer).

Lines 364 to 367: Difficult to understand if you are not specialized (amorphous solid state). Beside the fact that it challenged the traditional views, what are the consequences of this discovery?

Line 539: Second or third? I thought that methane was second (and close to ozone).

Figure 10: the figure does not read well. Complicated to understand with such a short caption. Also, on the y-scale, it seems strange to express an integrated flux in ppb. I guess that it is integrated over time, but for how long.

Line 742: I am not sure that research is trying to count the numbers of VOCs present in the atmosphere.

Line 949: It is appropriate to cite Hampson, but a reference to his report should be added.