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*Interactive comment on* "Global Atmospheric Chemistry – Which Air Matters" *by* Michael J. Prather et al.

**Anonymous Referee #2**

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General comments The manuscript presents an interesting new approach for the comparison of observed atmospheric in-situ data alongside an aircraft flight track and results from chemistry-climate models. Such a comparison has the intrinsic problem that the observational data are related to specific conditions of time and geographical location which cannot be reproduced adequately within a global model due to their coarse grid and difficulties to represent real transport, emission, and deposition processes. This is related to the question of representativeness of the observations and the full variability of atmospheric constituents. The manuscript tries to add a new approach to overcome these problems by including more of the climatology of the model data and by shifting the comparison of single constituents towards the comparison of relationships between constituents including an additional weighting by processes like methane oxidation and ozone production. My feeling here is that both approaches (to-

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gether or each of them separately) are very promising but the final concept how to do the comparison between observations and models is missing. Since the title, abstract, and introduction of the manuscript builds up a certain expectation about the presentation of a new and better tool for this kind of comparison I did feel quite disappointed at the end. The manuscript explains how to generate relationships between constituents and the additional weighting processes, both for the observations and for the model data, but the comparisons are then done by visual inspection and their subjective interpretation. Since I myself at some occasions would come to different interpretations looking at the presented figures of relationships between constituents, both weighted or unweighted, it is clear that this new approach is not facilitating the developement of objective arguments, at least at the moment.

Detailed comments: As I see it, the new approach in the manuscript contains three different parts. The first one shows how to compare between 3-D model results and observed data. The traditional way would be to compare observed data obtained at a specific time and location to model results from matching grid boxes for the same time of day and year. The new way is to compare model data from a broader region and time so that the exact match to the location and time of the observation is no intrinsic requirement any longer. This broadens the view because the model might be able to reproduce observed phenomena not at the exact coordinates but at a slightly different time or location. The key point here is: are the model climatology of a species and the unbiased climatology of observations similar or not. The second part demonstrates how to calculate important chemical reaction pathways with respect to methane loss and ozone loss/production (called reactivities) for a list of observed key species without the observation of reaction partners like HOx radicals. This is done by preparing a spin-up time of the involved CCM models, initializing the observed key species in the appropriate grid boxes of the model, switching-off model processes like transport, emission, deposition, and then propagation the single boxes in a 0-D fashion 24h into the future. The reactivities are then calculated as 24 hour averages of the processes methane oxidation, ozone production and destruction, and attributing these results to

the observed data. These reactivities are also calculated for the model data itself just by switching–off transport, emission, and deposition without initializing the key species to observed values. The manuscript tries to provide evidence that the switching-off of transport, emission, and deposition does not alter the reactivities too much by comparing full and switched-off model runs. But since the reactivities are 24h averages, the concentrations of the species are the same at the beginning of this 24h period shifting away only slowly afterwards, and since all species are in a kind of balance to each other due to the spin-up period, the real information content of this exercise might be less than anticipated. The exercise by initializing the key species to different values than the models own values is not done. Since such new values of the key species might not be in balance with the other species calculated during the model in the spin-up period, there might be discrepancies when the system tries to adjust to a new balance within the following 24h. The third part of the approach is to compare relationships between key species, either weighted or unweighted by the reactivities, rather than the key species themselves. I find this kind of approach very interesting since the underlying atmospheric processes introduce complex couplings between different species. Looking at a single species might give a wrong impression because good agreement between model and observation can be caused by compensating errors. But how to compare these relationships prepared from model data and from observations? There is no objective tool proposed to do that. The interpretation of a match or mismatch looking at relationships in the observations or in the model results still remains to be a subjective choice of the authors or readers of a manuscript. This gap is mentioned in the paper itself at line 645. Overall, I find that the proposed approach might have a lot of advantages, but its real capability is not proven or even is not illustrated. At the moment, the authors only can show results of climatologies of data calculated by six different CCMs. I think that one of them, model D, is quite different to the other five, for example looking at the probability distribution of HCHO in figure 5, the large values of J-O3(1D ) (together with model B) in figure 4, the large values of P-O3 at 20N-60N in figure 3, the large values of L-O3 at 20N-60N in figure 3, or the small values of L-

CH4 at 20S-20N in figure 3. If one would take the results of this model D as kind of observed dataset along the fight track shown in figure 9 and would follow the proposed approach, can one expect to reveal the difference of model D compared to the other five? This kind of question is already started at line 547 going to line 561. But besides mentioning some of the differences of model D there is no clue to explain its different behavior. If such an explanation is not possible for model D, can we really expect to learn something once observed data from ATom are available?

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