

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2022-631

Anonymous Referee #2

Referee comment on "Heterogeneity and chemical reactivity of the remote troposphere defined by aircraft measurements – corrected" by Hao Guo et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-631-RC2>, 2022

As a reviewer of the original submission, I find myself unsure how to approach the current version of the article. Has the original submission been withdrawn and should the current version be viewed in isolation, or should the current version be seen as a correction to the original? There is, what I believe to be, mention of the original publication and errors that were found after publication in a couple of places in the current version of the manuscript.

For example, at lines 299 – 304, the authors write:

'After publication and with continued analysis of the unusually reactive East Pacific region, we determined that the method of long-gap filling for NO_x resulted in propagation of high NO_x levels from the over-land profiles into the over-water profiles in the tropics. We separated these two set of profiles used for long-gap NO_x filling and created an updated MDS-2b. This experience points to the importance of having continuous NO_x measurements.'

Then the authors introduce the UCIZ* results, at lines 358 – 362, writing:

'Unfortunately, these new calculations with the revised protocol (UCI2*, shown in the original published version) are not reproducible and inconsistent with the original CTM version used in P2017, P2018, and the MDS-0 calculations. It appears that either the initial conditions, the ATom-specific version of the UCI CTM, or the retrieval of the 24 h average rates is not correct.'

So I am tempted to believe the current version should be viewed as a resubmission of the original manuscript. In that case, the current version should discuss the errors that were found in the original calculations and the reasons for a new submission in a clear, straightforward manner and in a single place within the current manuscript. Without constantly referring to the original submission to separate, for example, the problems with HNO₄ and PAN from the problems with NO_x gap filling it is impossible to understand how this version differs from the earlier submission and what motivated a resubmission.

Even with the revisions, the results seem broadly consistent with the earlier submission and about the only new comment on the findings I have is a question about the method. As summarized in Figure 1 and stated in the abstract (lines 53 – 55) 'We find that 80 %–90 % of the total reactivity lies in the top 50 % of the parcels; and 25 %–35 %, in the top 10 %, supporting previous model-only studies that tropospheric chemistry is driven by a fraction of all the air.' The air parcels being compared are taken over a latitude band from 53S to 60N, which includes a significant sampling of air from mid to high latitudes. Are the conclusions reached here really that different from those seen in modelling studies that find a large fraction of methane is oxidized in a relatively small portion of the atmosphere. For example, as shown in Figure 8 of Voulgarakis et al. (Analysis of present

day and future OH and methane lifetime in the ACCMIP simulations, Atmos. Chem. Phys., 13, 2563-2587, doi:10.5194/acp-13-2563-2013, 2013) over 60% of the global methane oxidation occurs between 30S and 30 N and between the surface and 500 hPa. To what extent does the finding that reactivity is not equally distributed across all parcels reflect heterogeneity at sub-regional scales and how much does it simply reflect the fact that much of the reactivity happens in the tropics where the sun is high and there is lots of water vapour?

Aside from that, my minor comments are itemized below.

Minor comments

Line 63: Missing 'to' in 'can be traced lower NOX levels'.

Line 479: For 'In the central Pacific (row 1)', I think 'row' should be 'column'.

Line 603: There is a stand-alone bit of text that seems out of place - 'P-O3.'

Lines 607 – 608: On first (and second) reading I interpreted 'The extreme eastern Pacific reactivities are seen in the mean values in the legend' to mean that the values shown on the legend of Figure 5 give the means for the eastern Pacific. I think what you mean to say is 'The influence of the extreme eastern Pacific reactivities...'

Lines 636 – 637: Speaking of the missing O3 production in the lowest 2 km, the authors state: 'The occurrence of the same error over the central and eastern Pacific as well as the Atlantic Oceans makes this a robust finding.' Judging from Figure 3, it is not clear to me that the error is present over the Atlantic Ocean?