We thank the reviewers for efforts and insights, and particularly for revisiting and re-reviewing this manuscript a year later. When we discovered the mistakes in the published paper (acp-21-13729-2021), we prepared and submitted a corrigendum noting the corrections with an extensive set of revised sentences and paragraphs and tables and figures. The Executive Editors wisely thought it best to re-issue a corrected paper that was readable on its own. That is what you have. Based on your responses we will make it clearer still. The title should have the label "—Corrected" added to the end to ensure that the two papers are different. The 2021 paper on the ACP website can note it is withdrawn and point to the corrected one (when published). Based on your suggestions, I will put the explanation of the errors and their impact on the results into a Preface, so that this paper can be read through as simply the up-to-date corrected results. I ask that the Editors allow this to be included without changing the numbering of sections to avoid further confusion.

The new PREFACE is included in the revised paper, but I paste in immediately below so that you can read it without going to the paper. It should be understandable on its own.

> **Preface.** This paper presents a corrected version of the paper published under the same authors and title (sans 'Corrected') as https://doi.org/10.5194/acp-21-13729-2021. While continuing our analysis of the ATom data we found several major mistakes or decision errors. The main conclusions were unchanged except those regarding production of O_3, but most of the numbers and many of the figures changed slightly. A corrigendum to the original 2021 paper was prepared, but the changes were extensive enough so that the ACP editors and the authors decided that a completely new paper should be produced and the 2021 paper withdrawn. The errors that were corrected are described in this preface and discussed at most briefly in the paper. First, we found that measurement errors in PAN and HNO_4 were large (~100 ppt), and when this occurred in the lower troposphere, the rapid thermal decomposition released large amounts of NOx. There is no easy fix for this, and we developed a new protocol (RDS*) for computing reactivities by allowing the species to thermally decompose before use in the model, as described below. This fix greatly reduced O_3 production (P-O3) in the lower troposphere. A second NOx problem involved the propagation of polluted profiles from the Los Angeles basin to gap filling over the tropical eastern Pacific. This correction resulted in the update of the Modeling Data Stream to version 2b. These NOx errors cause noticeable changes in reactivities, especially P-O3. Other decision errors led us to decrease the southern latitude extent of the Atlantic and Pacific transects from 54° S to 53° S to avoid spurious parcels being included. Also, cosine of latitude weighting was applied to data for all figures and tables. The UCI model now includes all higher alkanes and alkenes in the ATom data as C_3H_8 and C_2H_4, respectively. These last three decision errors had detectable but small impacts.
>
> The most worrisome error was the evolution of the UCI CTM model's ATom version from its use in the MDS-0 results shown here to the final calculations with MDS-2 as the UCI2* model in the 2021 paper. The first MDS-0 UCI model was taken directly from the main CTM code line and developed for Prather et al. (2017; 2018) by Xin Zhu (not on the 2021 paper). This model was then further adapted and developed for the 2021 paper and for additional complex sensitivity tests. At this stage (i.e., the UCI2* simulations in the 2021 paper), the results failed several logic tests and were irreproducible. With the decision to withdraw the paper, we returned to the MDS-0 UCI model, and Xin Zhu adapted it to more efficient ATom runs as well as adding several new diagnostics and checks to ascertain the ATom runs were being calculated correctly. As noted in

the paper below, we carefully checked the O_3 budget in terms of rates and tendencies, and these are now consistent in model UCIZ. Further, the sensitivity coefficients ($\partial ln\mathrm{R}/\partial ln\mathrm{X}$ and $\partial^2 ln\mathrm{R}/\partial ln\mathrm{X}\partial ln\mathrm{Y}$) calculated for a subsequent paper are now closer to theoretical expectations for a quasi-linear system. The UCIZ* model results here are our best, revised estimate of the ATom reactivities.

Reviewer #1

This paper is a resubmission of a previous paper under the same name (https://acp.copernicus.org/articles/21/13729/2021/acp-21-13729-2021.pdf), with two corrections made. As I have previously reviewed this paper, I will limit my comments to the corrections.
Changes in the analysis result from two separate corrected errors:
(1) A change is in the modeling data stream (MDS-2) NOx values resulting from new gap-filling protocols. The original version (MDS-2) had high NOx over the East Pacific, and the revised version has lower values due to a different method of gap filling.
This error has been clearly identified, explained, and corrected. The impacts on the analysis are explained well.

Agreed.

(2) Issues with the use of RDS protocol (which generates reactivity datasets based on HNO4 and PAN corrections) in UCI (UCI2*). I found this section more difficult to follow. It is not clear to me what the issues are, other than that either the CTM or the RDS* protocol results in calculations that are irreproducible and inconsistent. I suggest careful rewording of this section with more detail, or eliminating references to the incorrect version. My points of confusion are below:*

Agreed. The new discussion of UCI2* is restricted to the preface and we have pulled the results from the tables as they only confuse. The RDS* protocol is emphasized because we altered the measured abundances of PAN and HNO4 in order to avoid absurd results.

- *What "calculations" is Lines 358 referring to—is it the statistics referred to in lines 342-345, or P/LO3, or everything in Table 2 and S8, or something else?*

Yes, this is confusing (to us also) and we have moved the "corrigenda" issues into the new Preface and now just describe and compare the old MDS-0 with the new MDS-2b.

- *How is the "Atom-specific version of the CTM" discussed on line 361 different than the "Atom-specific UCI CTM" mentioned on line 364? Table 4, bottom right, indicates that reaction rates are changed—can you elaborate?*

Agreed. We have eliminated discussion of the intermediate CTM simulations that were in error. The error was not in the reaction rate coefficients, but rather the setup of the CTM 24-hour run and the extraction of reactivities. This should be much simpler with the new text. Thanks for pushing on this.

- *Is the "updated RDS*" protocol mentioned on line 371 the same as the RDS* protocol in the prior paragraph, or is the RDS* protocol now updated somehow?*

We have revised this discussion. There is only one RDS* protocol as noted 3 comments above.

- *Is it meaningful to compare UCI2 and UCIZ*, since both the MDS and UCI changed?*

We have dropped UCI2 from this paper except in the Preface to explain the need for this corrected paper.

- *It seems Table 4 bottom right still discusses UCI2*, but that the data isn't shown—should it be removed?*

Agreed. This table has been revised to show only the current data sets.

Minor comments

- *Text in Figure 2Corr is too small to be legible.*

Indeed they are, my apologies. The figures should have been enlarged to see the pixels, and they are now so.

Reviewer #2

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 NOx resulted in propagation of high NOx 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 NOx filling and created an updated MDS-2b. This experience points to the importance of having continuous NOx 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 NOx gap filling it is impossible to understand how this version differs from the earlier submission and what motivated a resubmission.

We agree with these comments (and those of RC1). As noted in our overall response we have revised the text with a Preface to note the mistake in the 2021 paper and have revised the main text here to be read cleanly.

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?

Good point, being a co-author on the Voulgarakis 2013 paper, this is embarrassing. The high – reactivity loading of the total budget for CH_4 (especially, as seen in the curtain plots) is due mostly to the temperature effect of the reaction rate coefficient and next to sunlight and water vapor. This is seen in his Figure 8, which shows that 63% of the loss occurs in the tropics below

500 hPa (31.3% of the tropospheric mass from 200 hPa to the surface). If we extend our 53S to 60N transects (.83 of the pole-to-pole mass) then the top 50% hot parcels are 41.6% of the total mass. The fraction of total L-CH4 is 84.5% (averaging Pacific and Atlantic). So our enhancement factor is .845/.416 = 2.03, and Voulgarakis' is .63/.313 = 2.02. Identical as you noted. However as we go to the higher end, we see the effect of the large heterogeneity in L-CH4 as seen in the curtain plots and other statistics, see our table below. Thus, we must correct our text. We split the paragraph in the middle of line 429 and inserted a new paragraph with new material:

"The enhancement factor for the top 50 % L-CH4 parcels is 2.0 (84 % of reactivity in 42 % of mass) given that our 53 ºS – 60 ºN transects cover 83 % of the air mass below 200 hPa and assuming that L-CH4 is negligible poleward of these transects. This enhancement is effectively a large-scale feature because the tropical lower troposphere, being warm and wet with high sun, dominates the budget. It is seen in previous model intercomparisons calculating budgets in large tropospheric blocks like Voulgarakis et al. (2013) with 63 % of L-CH4 in 31 % of the air mass (500 hPa–surface, 30 ºS – 30 ºN). The impact of the extremely hot parcels and the heterogeneity seen in the ATom parcels is evident in the steep slopes above the 90th percentile, yielding enhancement factors of 3 to 4."

ATom-1	Air mass	Of total L-CH4	Enhancement factor
'top 50%'	41.62%	84.5%	2.03
'top 10%'	8.32%	26.0%	3.12
'top 3%'	2.50%	9.5%	3.80
Voulgarikis 2013 Tropics, below 500 hPa	31.3%	63%	2.02

Aside from that, my minor comments are itemized below.

Minor comments

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

Done!

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

Oops. We fixed the referencing to the Figure 2 panels

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

Thanks, that was a mistake and is removed.

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…'

Yes, thanks, this has been reworded and is cleaner.

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?

Correct, but the models in the Atlantic Ocean show a very large drop below 2 km; whereas ATom relative drop is much less. Thus we needed to rephrase this section:

"In comparing ATom results with modeled climatologies, we find a systematic ATom-model difference: models show a large relative drop in O_3 production below 2 km over the tropical oceans but ATom shows an increase (C.Pacific), no change (E.Pacific) or a much lesser drop (Atlantic). We traced this result to the lack of NOx at 20–60 ppt levels in the models below 4 km and believe it provides a clear challenge in modeling ozone over the remote oceans."