

***Interactive comment on* “Cloud system resolving model study of the roles of deep convection for photo-chemistry in the TOGA COARE/CEPEX region” by M. Salzmann et al.**

Anonymous Referee #1

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The article by Salzmann et al. discusses chemistry results from model simulations representing the December 20-26, 1992 TOGA COARE convection. Reading and comprehending this paper the first and second times is an arduous task because it discusses several major topics of active research. These topics include the production of nitrogen oxides from lightning, the ozone budget in the tropical upper troposphere associated with convection, and the budget of hydrogen oxide reservoirs (H_2O_2 , CH_3OOH , HCHO , $\text{CH}_3\text{C}(\text{O})\text{CH}_3$). While the authors make some significant contributions to each of these topics, the impact of their results is lessened due to the clutter of other discussions in the paper and its supplement.

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There are some major points that need to be brought out in this discussion paper.

Major Points:

1) An important conclusion of this study is "...results of 2-D sensitivity runs are to be interpreted with caution..." because of differences (especially in NO_x) found between 2D and 3D simulations. I think this point should be stressed further. Not only is it important in the realm of lightning- NO_x (because placing a source of lightning in a 3D volume is not equal to placing the source in a 2D "volume"), but if/when super-parameterization* becomes commonly used, its next step will be to incorporate chemical species to represent convective transport, wet deposition, and lightning- NO_x . Thus, efforts to define the limitations of 2D cloud system resolving models must become common knowledge. The authors have the opportunity to do that in this paper.

*super-parameterization refers to using a 2D cloud system resolving model as the convective parameterization in large-scale models (Randall et al., 2003 provides a good summary).

2) One important result that we learned from TOGA COARE is that there is a tri-modal structure in cloud top heights which has an impact on vertical transport (Johnson et al., 1999). It would be interesting to learn how the tri-modal cloud top structure contributes to the resulting vertical profiles of chemical species. Are there significant differences in species profiles if the results are sampled by 1) clear air only, 2) columns with clouds, and 3) columns with different cloud top heights? I realize this is challenging due to the model configuration of 500 m vertical spacing and 2 km horizontal spacing, but nonetheless would be interesting.

Many figures show domain-averaged vertical profiles. I suspect that vertical profiles can have different characteristics depending on how much of the domain is occupied by clouds (cloudy volume versus clear-air volume). In many cases, the clear air vertical profile can smooth out (remove?) the effects of convective transport when a domain

average is calculated.

3) One major concern I have had with this paper is that it is not very well focused. To make matters worse, the paper makes extensive use of supplementary material that must be read to understand the paper. I find this to be an abuse of supplementary material and suggest that the editors accept supplementary material only if it follows the ACP guidelines (data sets, animated visualizations, etc.).

4) Because much of the paper discusses low O₃ mixing ratios in the upper troposphere, I suspect that it is the main motivation for this paper as extremely low O₃ mixing ratios were observed in the tropical upper troposphere during CEPEX (which occurred near the TOGA COARE region during March 1993). I recommend that the authors focus the paper on this topic.

My understanding is that there are four processes that could be responsible for the low O₃ mixing ratios: 1) vertical transport of O₃-poor boundary layer air, 2) horizontal advection from other regions, 3) depletion of O₃ by chemistry in a high NO_x environment that is created by lightning NO_x, or 4) depletion of O₃ via halogen chemistry. This paper also demonstrates the role of the Madden-Julian Oscillation on O₃ profiles as another contribution to low O₃ in the upper troposphere. It seems straight-forward to structure the paper along these lines.

To successfully do this, it must be recognized that the discussion on lightning production of NO_x is important to the O₃ results. This means that the authors will need to select the most relevant results from the lightning NO_x section.

Specific Points:

1. Abstract should be written more concisely by reporting the "headline" results: 2-D vs. 3-D results differ; lightning-NO_x has minor impact on O₃; importance of mid-tropospheric entrainment and undiluted transport of MBL air; influence of ISO on O₃ in the UT.

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2. P. 410, line 26 is an example of supplementary material abuse. Instead, a sentence such as, "Anvils are identified to be regions with $q_{tot} > 0.01$ g/kg that are within 1400 m of the cloud top height above the maximum updraft.", should be used.

3. Justification of the assumptions stated in lines 20-29 of page 410 need to be included. (location of the upper mode - other studies use $z(T=-45C)$; location of the lightning flash to be where w_{max} is; lightning NO production calculated every 56 seconds instead of every time step).

4. Despite an extensive comparison of model results to previous studies, a major weakness of this paper is the lack of direct evaluation with measurements. Thus, at best, the authors can only conclude that the model results are similar in magnitude to those observed at other times in the same region or in nearby regions at other times. The authors did do an exhaustive comparison to previous studies, but it is never as convincing as direct comparisons

Some examples: The model simulates the 19-26 December 1992 period of the Intensive Flux Array region of TOGA COARE. The modeled flash rates are compared to flash rates observed in the same area for 10-11 February 1993 and 11-17 February 1993. The NO and NO_x volume mixing ratios are compared to PEM-West B (February-March 1994) and PEM-West A (September-October 1991).

The evaluation with measurements paragraphs (from flash rates, to NO_x and O₃ mixing ratios) should state that the lack of direct measurements for evaluation is a shortcoming of the results.

5. P. 411, lines 1-8, While the model result calculation looks okay $1021/((248)^2*6) = 0.0028$, the calculation from the Petersen data may be wrong: $800/((600)^2*2) = 0.0011$. Thus, the model results are 2.5 times too big for the flash rate. Further, the Petersen observations are not of the same time period as the modeled storms.

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6. P. 413, Because lightning-NO is placed in the w_{max} column, is the NO_x vertical distribution (on average) dictated by the cloud top height rather than the heights of the Gaussian distribution peaks? Should results be sorted differently - MCS vs non-MCS convection, or via a cloud top sorting method?

7. P. 413, line 25, Is there a justification for α ? And why is it a "small" scaling factor?

8. P. 415, lines 21-23. Here is another example of supplementary material abuse. If the paper is about NO_y species budgets, then that material should be in the main paper. Otherwise, there should not be any references to specific tables in the supplementary material. There should only be a sentence saying "More details on the NO_x , PAN and HNO_3 budgets can be found in the supplementary material." However, because NO_y species budgets are not a data set or animation, it may not meet the supplementary material criteria. Personally, I think it would be nice to have the budget tables included in the supplementary material, but I do not think there needs to be a discussion of the results.

9. Section 3.2. Is the NOLTN simulation a 2D simulation or 3D? This should be clearly stated. Further, in comparisons between 2D and 3D simulation results, it should be noted that besides the volume placement of the NO source, other differences (e.g., strength of updraft resulting in different outflow heights) between 2D and 3D simulations exist.

10. Sections 3.3 and 4.1: The supplementary material describes the photolysis rates to be calculated based on Landgraf and Crutzen (1998) with effects of clouds modifying the clear sky value. How strong is the cloud scattering on the photolysis rates? I assume that dark regions of the clouds would have decelerated photochemistry. Thus, these regions would accumulate NO_2 while depleting O_3 . Is there evidence in the model results for this to occur?

11. Figure 15 and its discussion. How representative is the surface minimum in a 248 x

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248 km² domain compared the minimum values in the UT? Have you tried other values e.g. the 10th percentile value?

12. Section 4.3 is weak because (1) the results are based on fairly coarse model resolution (500 m), and (2) the results are based on 2-d simulation results which do not produce a realistic simulation as noted earlier in the paper.

13. Section 5: It is fairly obvious that when NO increases the partitioning between OH and HO₂ shifts because of the NO + HO₂ reaction. The authors point to the importance of this on the NO_x timescale. I suggest that this effect can be described in one paragraph in section 3.3.

14. P. 421, line 25. Why would u lag O₃ instead of the other way around? If the dominant tendency of O₃ is horizontal advection (section 4.1), should not O₃ lag u? Or, if low O₃ comes from cross-equatorial flow, should it not correlate with v? Does v increase/decrease with changes in the ISO?

15. P. 422, line 5. Here is another example of supplementary material abuse. This supplementary material should be part of the main manuscript so it is easier for someone to comprehend.

16. P. 422, line 9. I find it critical to know when there was a westerly phase or easterly phase of the ISO for both the period simulated here and the CEPEX time period. Is the Wang and Prinn (2000) squall line occurring during the easterly phase or westerly phase of the ISO? And is it in the same phase as the storms simulated in this paper?

17. Conclusions. On line 13, P 423, it states that the 2-D results should be interpreted with caution, while on line 18, P. 424, the 2-D results are used to make a major conclusion about downward transport of ozone as being important. This is inconsistent. I, as suggested, am interpreting the 2-D results with caution, especially based on comparison of Figures 12b and 14b which show for z = 11-14 km positive vertical advection

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tendency in the 3D simulation and negative vertical advection tendency for the 2D simulation. I do not find downward transport of ozone to be a major result (based on the flaws listed in point 12) and recommend removing this conclusion.

18. Conclusions. P. 424, lines 5-7. These lines should be written with more tact. For example: "The causes of extremely low O₃ mixing ratios in the upper troposphere were examined through a series of sensitivity studies. We found that low O₃ can be a result of lightning-produced NO when high NO production rates and flash rates are used, similar to that found by Wang and Prinn (2000). However, a recent review of NO_x production by lightning (Schumann and Huntrieser, 2007) indicates that the NO production rates used in this sensitivity simulation is 10-100 times greater than most values reported in the literature. We think this source rate is unrealistic. Other factors contributing to low O₃ include undiluted transport of O₃-poor boundary layer air via convection, but this is not enough to give extremely low O₃ mixing ratios in the UT. Instead we think the ISO is partly responsible for the extremely low O₃ mixing ratios ..."

Technical Details:

1. Page 411, line 16 should be $Z = 2.76$
2. P. 413, The paragraph structure on this page is poorly written. Please reconstruct to the basics: thesis sentence, detailed support of the thesis sentence, conclusion sentence.
3. P. 413, line 27 Change "On the other hand" to "Consequently".
4. Figures:
 - a. Despite the applauded efforts of the authors to improve the quality of the figures, I still found myself magnifying the figures on the screen to 300% to read them clearly. I recognize that some of this is due to the ACPD formatting of the figures, but when it comes to publication I recommend ensuring easy viewing of the figures.

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b. For the contour and 3-d plots, I assume that the interpretation is to get an idea of where the high and low values are and not to determine the value at a particular location. If it is the latter, then a more distinct separation of color is needed.

References:

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Randall, D., Khairoutdinov, M., Arakawa, A., Grabowski, W. W., 2003: Breaking the cloud-parameterization deadlock. *Bull. Amer. Meteor. Soc.*, **84**, 1547-1564.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 403, 2008.

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