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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of simulated photochemical partitioning of oxidized nitrogen in the upper troposphere" *by* B. H. Henderson et al.

Anonymous Referee #2

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General comments:

Models of the upper troposphere generally have too little NOx when compared against observations. This paper employs a new approach aimed to isolate model chemistry to find why NOx is under-predicted, and this approach is applied to the INTEX-NA data set. It is based on these assumptions: (1) the time since convection is known via back-trajectories, (2) the initial NOx:HNO3 partitioning is essentially all NOx, (3) uptake of HNO3 on particles can be neglected, (4) mixing with background air can be represented by a single value of a single parameter of x per cent per day, and (5) that a healthy does of statistics (beyond this reviewer) can be applied and that all can be

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summarized and evaluated according to the mean time since convection – that is, a mean over all parcels sampled and modeled.

This is an interesting approach and generally worth publishing. However, the weight of all these assumptions coupled with the reliance on complicated (for this reviewer) statistics leaves this reviewer not fully convinced of the main result, that is, that it is simply the NOx chemistry that is too fast and is therefore responsible for the too small NO2. Nonetheless, assumptions are clearly spelled out, so each reader can decide for her/himself. On the other hand, some things are dismissed rather casually, without quantification (see below – effect of particle uptake, effect of sample bias, treatment of mixing [is given sensitivity check by varying the parameter, but is this adequate?]). Also, I can't accept the claim made many times that downward transport is via convective downdrafts (exclusively?). I hope these points will be addressed in a revision.

Specific comments:

p 20129: "until the air parcel is removed from the upper troposphere by convective downdrafts." This is not a complete and accurate portrayal of the dynamics. A lot of downward transport occurs through large-scale descent, not part of convective events.

p 20130: "... and mixed with background upper tropospheric air until downdrafts associated with subsequent deep convection remove air parcels from the upper troposphere." Same point as above. I don't think this is true. Other scale downward motions play a huge role.

p 20130: "Particle chemistry is most likely of limited importance in our study..." This claim is not supported in any quantitative manner. I am concerned that uptake of HNO3 on particles could significantly effect the NOx:HNO3 ratio in ways ignored in the present analysis.

p 20132: "only NOx and HNO3 have age-dependent mixing ratios." To what extent does this result simply derive from the way in which the data are divided up? Using

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a NO:HNO3 ratio helps to bring this about. Is too strong to say it guarantees it, but it sure pushes things in this direction. When NOx:HNO3 is high, NOx tends to be high and HNO3 tends to be low.

p 20134-5: "The time between convective lofting and subsequent subsidence, hereafter air parcel lifetime ..." Isn't lifetime really just the time since convective lofting, even if subsidence has not occurred? Please clarify.

p 20135: "a distribution of time since convection defined by the frequency of frontal systems." How can this define the distribution? It certainly contributes, but other factors help define it, too. A parcel can go up in one frontal system. It needn't come down in the next. Large-scale subsidence processes also contribute.

p 20135: It is acknowledged that the aircraft sampling was biased toward young parcels. How does the doubling described compensate for this? This seems to be pulled out of thin air, but perhaps there is a justification.

p 20138: "bias-corrected" <– this is a potentially misleading phrase. Is it really correct? A doubling has been applied that is aimed to act in the direction of correcting, but does that really make it correct? Why a doubling and not some other factor?

p 20146, typo: "an test"

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 20125, 2010.