

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 NO_x when compared against observations. This paper employs a new approach aimed to isolate model chemistry to find why NO_x 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 NO_x:HNO₃ partitioning is essentially all NO_x, (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 dose 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 NO_x 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 NO_x:HNO₃ ratio in ways ignored in the present analysis.

p 20132: "only NO_x 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 NO_x:HNO₃ is high, NO_x 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.