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Atmospheric Chemistry and Physics Discussions

Interactive comment on "HO_x measurements in the summertime upper troposphere over Europe: a comparison of observations to a box model and a 3-D model" *by* E. Regelin et al.

Anonymous Referee #2

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This paper presents a valuable dataset for the HO_x measurements in the summertime upper troposphere over Europe. The authors then compared the HO_x measurements to a box model and a 3-D model. While the box model was able to reproduce measured OH and HO_2 in upper troposphere, the 3-D model shows less agreement with observations. The authors attribute such discrepancy to the underestimate of H_2O_2 in the 3-D model. I have a few major concerns on this paper:

1. The temperature dependence of HO_2 calibration. As I mentioned in the previous comment, no temperature dependence of HO_2 calibration was ever reported in previous aircraft HO_x measurements by this technique, particularly for such strong tempera-

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ture dependence (change by more than a factor of 2 from 260 K to 290 K). According to the authors, the Penn State instrument (ATHOS) needs to be corrected by 12% for such temperature change. But this is nowhere near the temperature dependence shown in this instrument. As a result, the measured HO_2 in upper troposphere is higher than all previous studies, including INTEX-A, PEMT-B, and TRACE-P. Although this instrument was installed in a wing pod (compared to the Penn State instrument which is set up within the freight compartment of the DC8), the temperature in detection axis doesn't seem affected. It seems to me that major conclusions in this paper rely heavily on the temperature dependence of HO_2 calibration. But this requires further investigation.

If we assume the air velocity is on average 20 m/s, the time for traveling from the first axis to the second axis is only 16(cm)/20(m/s) = 8 ms. If the authors attribute the temperature dependence of HO_2 calibration factor to wall loss, which means half the signal is lost within a temperature decrease of 30 K, this wall loss rate would be extremely large. Is radial diffusion rapid enough to provide such fast wall loss? I also expect the radical plume would be a parabola shape with the peak in the center, and the radical measurement is confined to center of the flow (see Figure 2 in Donahue et al. (1996)). So even if the wall loss is extremely fast, HO_2 concentration in the center may not be proportionally affected. I think the authors should examine this temperature dependence of HO_2 calibration more carefully.

2. In-flight calibration. Martinez et al. (2010) showed the in-flight calibration. Was in-flight calibration also conducted in this study? If so, is there any useful information to back up the temperature dependence of OH and HO_2 calibration factor?

3. Model comparison. I don't think the results from 3-D simulations provide much insightful information. While the authors were trying to prove that underestimate of HO_2 is due to the underestimate of H_2O_2, there could be other possibilities. For example, such underestimate could be due to the underestimate of HCHO, which is another major source of HO_x in upper troposphere (Jaegle et al., 2000). Another possibility is J value. According to Table 2, J values in global model also appear to be biased low

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by 30% or more. It seems to me that these possibilities could be easily examined by doing a HO_x budget analysis as done in Jaegle et al. (2000), which is more convincing than the scattering plots in Figure 9-11.

4. Convection transport. In section 5.2.3, why OH and HO_2 are underestimated on southern flight tracks but not on northern flight tracks? Is there any reason behind this? I think the authors should provide some in-depth discussion on this.

Minor comments:

1. In section 4.1, could the authors give some brief introduction on the MESSy? It is not clear how this interface works.

2. Page 30633, "Observed HO_2 mixing ratios exceed the INTEX-(N)A, PEM-(T)B and TRACE-P observations." I think authors should be more quantitative on this.

3. Page 30636, "HO_2 is almost always underestimated even if the H_2O_2 is well reproduced by the model, as seen in Fig. 9d." This tells me that underestimate of H_2O_2 may not be the only reason for underestimate of HO_2.

4. Page 30637, "Figure 10b indicates in the same area a correlation between the degree of OH underestimation and the underestimation of NO. Thus, a missing HO_2 conversion rate is likely responsible for the OH underestimation." I don't understand this sentence.

5. Page 30640, "The model tends to underestimate not only H_2O_2 but also NO mixing ratios in convectively transported air masses." I couldn't find any description on this from the section of "influence of convective transport" (Section 5.2.3). How did you get this conclusion?

6. Is Fig. 10a a zoom-in version of Fig. 9c? But they look very different. This definitely should be clarified.

Reference

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