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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Rapid convective outflow from the U.S. to the upper troposphere over the North Atlantic during the NASA INTEX-NA airborne campaign: flight 13 case study" *by* S. Y. Kim et al.

Anonymous Referee #2

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General Comments

This paper presents a case study of polluted air from the US boundary layer that has been transported up to the upper troposphere (UT) over the western Atlantic. Using chemical tracers and trajectories it provides evidence for the source region being the southeastern region of the US. However, I do not believe that the authors make a good case from the measured data for the transport to be via rapid convection.

The ratios of the C3H8/C2H6 indicate a photochemical age of the sampled air to be

1-2 days which the authors say is reasonably similar to the transport times deduced from the backward trajectories. Since the kinematic trajectories are calculated using the vertical velocities from the GFS analyses, convection is likely to be represented to some extent, but I think it is questionable as to how well small scale convective systems will be resolved. Even, if the trajectories give transport times similar to that indicated by the tracers, it does not indicate if convection was an important transport process or whether the rate of transport can be explained by large scale advective transport along the warm conveyor belt associated with the cold front. If the trajectories are to be used as evidence of convective uplift, then a more thorough description of the trajectories is required, in particular the resolving of convection, and ideally some diagnostics to show the impact of convection on the vertical velocities used.

A previous study of rapid uplift in a cold front over Central Europe (Purvis et al, JGR 108 (D7): Art. No. 4224 APR 15 2003) used changing ratios of hydrocarbons with different atmospheric lifetimes to determine a timescale for transport in a similar way to this study. However, Purvis et al presented hydrocarbon lifetime measurements in certain regions which suggested photochemical ages of 1-2 hours compared to 3-D trajectory transport times of around 2-3 days and thus it was concluded that convective transport embedded within the cold front rather than larger scale advection along the warm conveyor surface dominated the transport of ozone precursors into the free troposphere. This paper should be referenced.

Although Kim et al provide meteorological charts and satellite images to suggest that convection was probably active, I don't believe that they provide clear evidence from the flight data that the transport is so rapid that it must have been via convection (see also below). In my opinion the authors do not provide sufficient evidence to support the main conclusion and title of the paper.

Specific Comments

Page 17369, Lines 19-24. A number of other papers from ICARTT (JGR ICARTT and

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INTEX Special Issues) have already been published on the outflow of pollutants (O3 and CO plus other pollutants). This work should be discussed in the context of these other papers.

Page 17370, Line 22. Methven et al (JGR 111 (D23): Art. No. D23S62 DEC 2 2006) has already illustrated that the some of the polluted air sampled on flight 13 of the DC8 on the 28th July was actually sampled several times as part of a Lagrangian experiment. This same air mass was sampled on the 25th July, also by the DC8, on the 26th July by the NOAA P3 and then later downwind by other research aircraft based in the Azores and Europe. Arnold et al (JGR 112 (D10): Art. No. D10S40 2007) inferred OH concentrations and air mass dilution rates from successive observations of nonmethane hydrocarbons including those made on the DC8 (flight 13). Elevated ozone in the upper troposphere during this event was also examined as a case study in Cooper et al (JGR 111 (D24): Art. No. D24S05 DEC 12 2006). These works should be referenced. The Methven et al paper illustrates that the P3 was sampling over the eastern US on the 26th and the DC8 was sampling over the southeastern US on the 25th. Given that these flights were over the suggested source regions of the polluted air sampled on the 28th by the DC8 it would be good to look at data from these flights. The hydrocarbon tracer data may indicate rapid uplift from the BL into the warm conveyor belt on time scales of a couple of hours which would provide evidence of convective transport.

Page 17373, Lines 2-5. For the calculation of the photochemical tracer age, a value for the OH concentration was taken from that measured on the DC8 in the SBL. What time of day were these measurements made? Given that the calculated chemical ages were of the order of 1-2 days, this would include night-time. A 24 hour average OH concentration would then seem appropriate. Arnold et al (2007) calculated values of 24-hr average OH of around 2x106 molecules cm-3 for the Lagrangian case involving the DC8 on the 28th, half that used in the current paper.

Page 17375, Lines 12-13. Surely the data plotted in Figure 4 are not from the entire

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flight, but just the data east of 70W (page 17371, line 8). I think it is also worth pointing out that the data collected below 5km is all collected around 40W and thus do not represent the lower troposphere across the longitude range of the flight, i.e. not below regions 1-3. This explains some of the diversity in the trajectories for the air "outside"; these regions.

Page 17378, Lines 3-5. Do the backward trajectories really indicate that the air masses were influenced by SBL air transported vertically through convection?

Page 17378, Lines 5-13. Since the ultrafine aerosol correlations do not parallel the results of Twothy et al, how does the analysis imply the impact of convective outflow?

Page 17378, Lines 13-20. Although the CH3OOH/H2O2 ratios observed in regions 1-3 are greater than observed in the SBL, the ratios are not $>$1, which the authors state would be a diagnostic indicator of wet convection. Therefore does this really strongly suggest the likelihood of convection as the primary mechanism for the re-distribution of the trace gases? Wouldn't the ratio also increase if the air had been transported through frontal cloud?

Page 17380, Lines 3-6. Wouldn't coincident high levels of CO and O3 give a positive slope for the O3-CO correlation?

Section 5. I think it is good that the outside air is divided into sub-groups especially since the air is sampled over a wide altitude range. It also would be good to comment on whether the sub-groups do reflect air sampled at different altitudes.

Section 5 and 6. I am confused as to how the correlation analysis (section 6) on the "outside" air leads to the conclusion that the entire tropospheric column over the North Atlantic during the time period surrounding flight 13 was impacted by North American anthropogenic emissions when in section 5 it is noted that the average concentrations of CH4, CO, CO2 and C2Cl4 in the "outside" air were lower than North Atlantic background air as measured at Bermuda and Mace Head. This seems contradictory to me.

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Please also avoid such generalisations as "entire tropospheric column over the North Atlantic" when the vertical profile data come from a small region around 40N 40W.

Technical Corrections

Page 17372, Line 8. Baton Rouge

Page 17374, Line 28, Unstable

Page 17380, Line 9. ppbv were

Page 17380, Line 18. over Asia

Table 1. UCN is not defined.

Figure 1. It would be useful to have a latitude and longitude grid marked on the figure. What is the underlying image? Increasing the size of the altitude v time plot would be useful.

Figure 2. The figures are very small and difficult to read.

Figure 3. Please state the location of the skew T and log P data. Test is very small.

Figure 4 caption. in July

Figure 5. Please give units if the colour scale.

Figure 6. Text is rather small.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 17367, 2007.