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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.

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Responding to Referee #2

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.

We used Purvis et al. (2003) as the reference in Lines 6-8 on Page 13 (6th paragraph in section 4) as follows: “The air mass transport time to the free troposphere can be estimated by combining hydrocarbon lifetimes and trajectories whether the transport was by the WCB on the synoptic scale or mesoscale convection (Purvis et al., 2003).“

Page 17369, Lines 19-24. A number of other papers from ICARTT (JGR ICARTT and INTEX Special Issues) have already been published on the outflow of pollutants (O_3 and CO plus other pollutants). This work should be discussed in the context of these other papers.

We already used Mao et al. (2006) and Millet et al. (2006) from the ICARTT campaign. Now we added a few more papers from ICARTT: “North American outflow was characterized comprehensively for its chemical composition during the ICARTT field campaign. Specifically, the large variations in ratios between the pentyl and C_2-C_4 nitrates over the North Atlantic indicated the impact of different parent hydrocarbons emissions from the U.S. to the North Atlantic by photochemical production during transport from the source regions (Reeves et al., 2007). Canadian and Alaskan forest fires emissions caused elevated CO, PAN, organic compounds and aerosols. Moderately high levels of CO and longer-lived hydrocarbons were found in about 44% sampled data which originated from North America by low and upper level outflow (Lewis et al., 2007).“

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

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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.

We reorganized the Introduction, inserting Methven et al. (2006) on Lines 4-6 on Page 4 (3th paragraph): "Five types of the North America outflow were classified, which were two types of low level transports, fire plumes, and upper and lower level export by fronts (Methven et al., 2007)." Moreover, Methven et al.(2006) and Arnold et al. (2007) were referenced in Line 20, Page 5 (1st paragraph in section 2.1): "This study focused on flight 13 which was conducted on July 28, 2004 with one of the main objectives being sampling of U.S. continental outflow as described in Methven et al. (2006) and Arnold et al. (2007)". Cooper et al. (2006) described high ozone over the eastern U.S. during July 25-28, so we used it as a reference at the final paragraph in section 3. The sentence is that the mixed effects of widespread convection and the WCB over the southeastern U.S. overlapped in their occurrence during July 25-28, 2004, and were also described in Kiley et al. (2006) and Cooper et al. (2006).

We focused on the atmospheric chemistry over the southeastern U.S. boundary layer, and we analyzed the data in this area using DC 8 flights 6, 7, 10, 12, and 19 as mentioned in section 2.1. Thus, we already included the DC8 flight on July 25, 2004 for the SBL area. Moreover, the P3 flight did not observe the chemistry on July 26, 2004, and P3 flight on July 27, 2004 just measured chemistry over the northeastern U.S. Thus, the P3 flight on July 27 was not included in our study.

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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 2×10^6 molecules cm^{-3} for the Lagrangian case involving the DC8 on the 28th, half that used in the current paper.

Arnold et al. (2007) estimated [OH] by the ratio of NMHC mixing ratios and we used the average [OH] measured over the southeastern U.S. If we used the [OH] from Arnold et al. (2007), we got 3 days for region1, 3.9 days for region 2, and 3.3 days for region 3 as the photochemical ages. These are a factor of two greater than our previous estimates. Our photochemical ages were better matched with transport time than with using Arnold OH values.

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

We changed the sentence to “The vertical distribution of mixing ratios of CO, CH_4, CO_2, and COS is displayed for the flight data east of 70°W in Fig. 5. Note that data at altitudes <5 km was obtained near 40°W, not directly underneath of flight legs at altitudes >5 km (Figure 1 and 5)“ in Lines 10 - 12 on Page 11(1st paragraph in section 4).

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

The backward trajectories did not indicate vertical transport by convection. We used

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backward trajectories to examine fast transport over the eastern U.S. and the North Atlantic Ocean. We meant to point out that the fast transport was the reason why the SBL air in regions 1-3 was sampled. To avoid confusion, we revised the sentence to assure the lucidity of that statement. (See line 15-17 on Page 14 (9th paragraph in section 4) in the revised manuscript)

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?

High concentration of ultrafine aerosol implies the occurrence of convection. Wang et al. (2000) observed high concentration of condensation nuclei (>10000 cm^{-3}) in the upper troposphere and suggested that this high concentration of CN was associated with convection by using the NO/NO_y ratio as a chemical clock. They also pointed out that high CN concentration from aircraft emissions in the upper troposphere was not sampled frequently because of faster dilution than the transport of the BL plumes via convection. Thus, we also used Wang et al. (2000) for reference in Line 23 on Page 14-Line 4 on Page 15 (9th paragraph in section 4).

Page 17378, Lines 13-20. Although the CH_3OOH/H_2O_2 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?

Snow et al. (2007) found that the ratios of <1 for H_2O_2/CH_3OOH and <100 ppbv for O_3 indicated convection during the ICARTT study. We originally used CH_3OOH/ H_2O_2 in our paper, the reciprocal of the indicator in Snow et al. (2007). We obtained the H_2O_2/CH_3OOH values of 0.77–2.15 for region 1, 0.25–5.34 for region 2, and 1.16–3.72 for region 3. The mean and median values are as follows; 1.3 for region 1, 2.6 for region 2, and 2.2 for region 3. From these values, regions 1 and 2 seem to be

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impacted by convection. Accordingly, we revised the text to be “Snow et al. (2007) found that ratios of <1 for H_2O_2/CH_3OOH and <100 ppbv for O_3 indicated convection during the ICARTT study. The flight 13 measurements showed lower mean values of H_2O_2/CH_3OOH which were 1.3, 2.6, and 2.2 in regions 1-3, respectively, than a value of 3.1 from the SBL. The ratio ranges were 0.77 – 2.15 for region 1, 0.25 – 5.34 for region 2, and 1.16 – 3.72 for region 3, suggesting that convection impacted regions 1 and 2. These results suggest the likelihood of convection as a mechanism for the re-distribution of trace gases captured in the flight 13 data.“

Page 17380, Lines 3-6. Wouldn‘t coincident high levels of CO and O_3 give a positive slope for the O_3-CO correlation?

We divided the data at 12km altitude into two groups using the CO mixing ratio of 100 ppbv as the criterion. For the data with CO >100 ppbv the slope of the O_3-CO correlation (r^2=0.4) was -0.6, and 1.3 for the data with CO<100 ppbv (r^2=0.27). In the group of CO<100ppbv, CO varied between 73 and 100 ppbv and O_3 between 51 and 108 ppbv. In the group of CO>100ppbv, CO ranged over 100 -139 ppbv and O_3 over 74 - 110 ppbv.

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.

We inserted the sentence in the beginning of Section 5. “As mentioned in section 4, the Outside air included all segments except measurements from regions 1-3. Hence different altitudes and geographical locations were mixed for the Outside data.“

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 CH_4, CO, CO_2 and C_2Cl_4 in the “outside“ air were lower than North Atlantic back-

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ground air as measured at Bermuda and Mace Head. This seems contradictory to me. 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.

As stated in the response to the previous Reviewer‘s comment, the measurements in the Outside group encompassed all the data except the ones from Regions 1-3, which were conducted at different altitudes and geographical locations. This Outside dataset spans vertically the region from the surface to 10.5 km and horizontally the North Atlantic between 40W-70W. Backward trajectories of the Outside indicated various air masses from the U.S. outflow to marine were mixed together in the Outside, but the Outside still shows good correlations between industrial/urban tracers. This suggests with the strong influence of urban and industrial sources even though the mixing ratios of urban and industrial tracers were low. Therefore, the authors believe the statement was not a generalization.

Page 17372, Line 8. Baton Rouge

We corrected it.

Page 17374, Line 28, Unstable

We corrected it.

Page 17380, Line 9. ppbv were

We corrected it.

Page 17380, Line 18. over Asia

We corrected it.

Table 1. UCN is not defined.

We added “(ultra fine aerosol)“.

Figure 1. It would be useful to have a latitude and longitude grid marked on the figure.

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Interactive Comment

What is the underlying image? Increasing the size of the altitude v time plot would be useful.

We added latitude and longitude lines, but we can't increase the size of altitude v time plot because this figure was provided by NASA.

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

We changed images. Note that analyzed sea level pressure at 00UTC July 26 was not available, thus we inserted the analyzed sea level pressure at 12 UTC July 25 because a stationary front existed continuously over the period.

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

We edited text in Fig. 4.

Figure 4 caption. in July

We corrected it in figure 5.

Figure 5. Please give units if the colour scale.

We added "Unit of color bar is hPa." in figure 6.

Figure 6. Text is rather small.

We changed it in Fig. 7.

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