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Contributions of regional transport and local sources to ozone exceedances in Houston and Dallas: Comparison of results from a photochemical grid model to aircraft and surface measurements

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[1] During the 2000 Texas Air Quality Study (TexAQS) and 2006 Texas Air Quality Study (TexAQS II) field experiments, aircraft measured ozone concentrations upwind, across, and downwind of the Houston and Dallas urban areas. Background ozone transported into Houston contributed, on average, approximately 50% and 66% of the total ozone on 8-h ozone exceedance days investigated by aircraft flights during TexAQS and TexAQS II, respectively. Analysis of a flight over Dallas on one exceedance day showed that transported ozone constituted 72% of the total ozone concentration. The aircraft measurements show that these two major metropolitan areas can be brought close to exceeding the 1997 8-h National Ambient Air Quality Standard for ozone of 0.08 ppm solely by the ozone contribution of regional transport before additional contribution from local sources. Large local contributions were also observed, particularly in Houston. Transport contributions to Dallas area ozone were quantified using the Comprehensive Air Quality Model with Extensions (CAMx) photochemical grid model and source apportionment methods. Model-predicted ozone concentrations were compared to ozone measurements from the aircraft and the surface monitoring network, and showed agreement on the importance of regional transport and local ozone formation. These results emphasize the benefits of regional control strategies, and suggest that local controls alone may not be sufficient to ensure attainment of the 8-h ozone standard in Houston and Dallas.

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1. Introduction

[2] Measured ozone concentrations are the result of ozone formation and loss due to local sources of precursor emissions, transport of ozone and precursors from nearby or distant regions, and complex, nonlinear interactions between local and transported ozone and precursors. It is important to separately quantify the relative contributions of local sources of emissions and regional transport in order to design effective ozone control strategies. The change in 1997 from a 1-h, 0.12 ppm ozone U. S. National Ambient Air Quality Standard (NAAQS) to an 8-h, 0.08 ppm ozone standard enhanced the importance of the contribution from regional ozone in determining the ozone attainment status of U.S. metropolitan areas [*NARSTO*, 2000]. In March, 2008 the Environmental Protection Agency (EPA) announced that it is revising the primary National Ambient Air Quality Standard for ozone from 0.08 ppm to 0.075 ppm. The results are discussed here relative to the 0.08 ppm standard that was in place at the time of the measurements. The implications of our findings for future exceedances of the 0.075 ppm standard are discussed briefly in the final section.

[3] The relative contributions of regional and local sources of ozone have been quantified through direct measurements in several U.S. urban areas outside of Texas. *Kleinman et al.* [2000] reported findings from flights upwind and downwind of the New York City urban area, and determined the local ozone contribution from measurements made during flight segments upwind of the City and within the urban plume. They found background ozone concentrations upwind of the urban plume ranging from 35 ppbv to 80 ppbv on the 4 flights they selected for analysis. Data from 23 flights upwind and downwind of Phoenix were analyzed by *Nunnermacker et al.* [2004], who determined that the background ozone contribution had a median of 51 ppbv and a maximum value of 84 ppbv over the course of the experiment. *Nunnermacker et al.* [1998] reported on the Southern Oxidants Study in which multiple aircraft measured the Nashville urban plume on 2 days

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during the summer of 1995. On these 2 days, the aircraft measured background concentrations of approximately 56–80 ppbv. *Fast et al.* [2002] made aircraft and ozonesonde measurements of elevated layers of ozone above Philadelphia during the early morning and then quantified the relative contributions from local precursor emissions and regional transport using a chemical transport model. They found that during high-ozone episodes most of the surface ozone was produced from local precursor emissions, but that, depending on the day, as much as 30–40% of the total surface ozone could be attributed to transported ozone. In summary, there is agreement among all of the measurement campaigns noted above that regional transport can make an important contribution to the total ozone measured in these urban areas.

[4] The Houston and Dallas metropolitan areas are each home to approximately 5 million people and exceed the NAAQS (i.e., are designated as nonattainment areas for 8-h ozone). Houston is a port city with extensive petrochemical production and refining facilities. Emissions of highly reactive volatile organic compounds from these facilities have been shown to result in ozone production rates and concentrations that are higher than those found in urban areas with a more typical mix of anthropogenic emissions [*Kleinman et al.*, 2002; *Ryerson et al.*, 2003]. Houston's meteorology is also favorable for ozone production, with a sea breeze circulation that can confine pollutants to the urban area, thereby contributing to high ozone levels [*Banta et al.*, 2005]. The Dallas area ozone problem is more typical of large U.S. cities, with volatile organic compound (VOC) and nitrogen oxide (NO_x) emissions dominated by mobile sources [*Olaguer et al.*, 2006]. Analysis of surface monitor data has shown that transport of ozone into the Houston and Dallas areas can contribute to high ozone in both of these cities [*Nielsen-Gammon et al.*, 2005].

[5] Here, we combine measurements from aircraft flights and from the surface monitoring networks to estimate the contribution of the regional transport of ozone relative to local ozone formation in the Houston and Dallas urban areas, and compare these measurement-based estimates to model results. On the relatively few days when the aircraft data are available, the contribution of the regional transport of ozone is estimated from the average ozone measured during transects upwind of an urban area. The average upwind ozone concentration will be referred to here as the background ozone concentration. It should be noted that this upwind ozone concentration is background only in a local spatial and short-term temporal sense, and does not refer to a continental-scale background. This background likely is influenced by urban plumes from other nearby and distant cities, or even recirculation of ozone produced from the same city on an earlier day. The contribution of the local ozone formation is then defined as the difference between this background ozone and the maximum 8-h average ozone reported from the surface monitoring network within that urban area. The contribution of the regional transport of ozone is also estimated from 8-h average ozone measured at a monitoring site on the upwind side of the urban area, although with less confidence owing to the relatively sparse coverage of the monitoring network. The results from these two methods of determining the regional transport contribution are compared. Model-predicted ozone concentrations show agreement with the measurements on the importance of regional transport of ozone relative to local ozone formation. The model's ozone source apportionment capability is used to determine the geographic regions contributing to high-ozone days in Dallas. The results show that these two major metropolitan areas in Texas can be brought close to exceeding the 8-h National Ambient Air Quality Standard for ozone of 0.08 ppm solely by the ozone contribution of regional transport before additional contribution from local sources.

2. Methods

2.1. Aircraft Measurements

[6] During the Texas Air Quality Study (TexAQS, held in 2000) and the second Texas Air Quality Study (TexAQS II, held in 2006; http://www.tceq.state.tx.us/nav/eq/texaqsII.html) field experiments, the Earth System Research Laboratory (formerly Aeronomy Laboratory) of the National Oceanic and Atmospheric Administration (NOAA) conducted research flights in the eastern Texas region. In 2000, 14 flights were flown between 16 August and 13 September aboard the National Center for Atmospheric Research L-188C Electra aircraft leased by NOAA. In 2006, 16 flights were flown between 11 September and 12 October aboard the NOAA WP-3D. Both aircraft were based at Ellington Field in Houston, Texas.

2.1.1. Measurement Techniques

[7] Instrumentation aboard both aircraft included 1-Hz measurements of O_3 by NO-induced chemiluminescence (CL). The 1-Hz O_3 CL measurements aboard the Electra were calibrated by and compared to a separate TEI model 49 UV-absorption instrument on the Electra in 2000, and compared to additional UV-absorption measurements during overflights of an instrumented ground site. These comparisons showed the 1-Hz O_3 measurements to be accurate within the stated uncertainty of ± (0.3 ppbv + 3%) [*Ryerson et al.*, 1998]. The 1-Hz O_3 measurements aboard the WP-3D in 2006 were made using a newer CL instrument; its calibration was quantified using a custom-built UV photometer to perform routine, in-flight standard addition of ozone to the CL instrument inlet. Comparisons of the newer CL instrument performance to the NOAA ER-2 UV ozone photometer in the lab, and to a NASA DC-8 CL instrument in flight, suggest the 1-Hz O3 measurements in 2006 are accurate to the stated uncertainty of ± (0.050 ppbv + 3%).

2.1.2. Description of Analysis

[8] During each of the TexAQS field studies, several aircraft flights were conducted to characterize the ozone distribution upwind, across, and downwind of the Houston–Galveston Bay (HGB) and Dallas–Fort Worth (DFW) metropolitan areas. Figures 1–4 illustrate the flight paths for the four flights that were conducted on the two flight days in each urban area when the highest 8-h maximum ozone averages were reported from the monitoring network in the respective urban area. These flights characterize the upwind ozone distribution transported into the urban areas as well as the downwind transport of the ozone plume produced by the urban area. The goal here is to derive the best possible approximation of the average background ozone concentration that was transported into the urban area on the day of

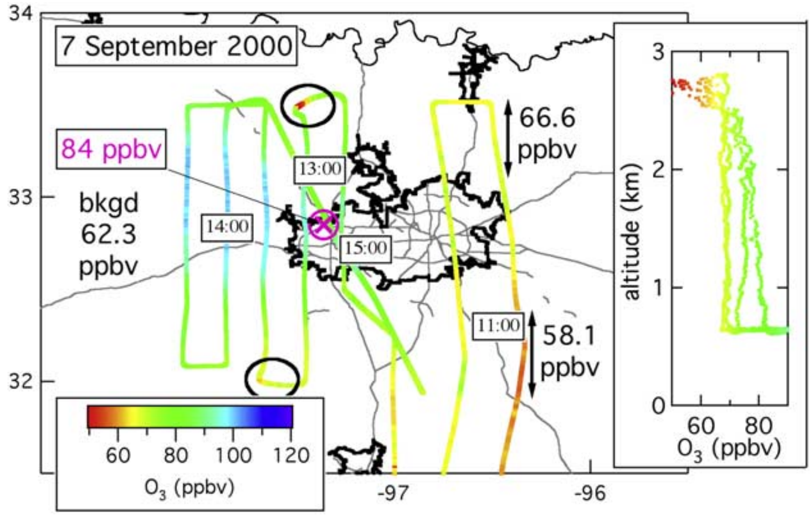

Figure 1. (left) Path of aircraft flight over the Dallas−Fort Worth metropolitan area (outlined by heavy black line) on 7 September 2000 color-coded according to measured ozone concentration. The prevailing wind during the flight was east-northeasterly. The local standard times of the flight are indicated along the flight path. The arrows indicate the flight path segments selected for calculating the background ozone transported into the urban area. The average ozone concentrations over those segments are shown beside the arrows, and the derived background is indicated on the left. The pink cross-in-a-circle indicates the location of the station in the urban area that recorded the highest maximum 8-h ozone average (indicated in pink in the tagged box) on the flight day. The black ovals indicate locations where vertical profiles were performed. (right) Ozone data collected during the two vertical profiles.

the flight. Each of these four flights is from the 2000 field study when higher ozone concentrations were observed. The following paragraphs discuss the derivation of the average background ozone transported into the respective urban areas for each of these four example flights. They are discussed in order of increasing complexity of the analysis. These four examples serve to exemplify the analysis of all 21 flights, which were conducted in a similar manner. Generally, the background ozone is taken to be the average upwind ozone concentration, with relatively minor adjustments made for the ozone entrained into the boundary layer as its depth evolves through the day. This entrainment is estimated from a few vertical profiles conducted by the aircraft during the urban characterization transects. These profiles were often not conducted at the same location and time as the upwind transect, but they do serve to adequately quantify the small boundary layer evolution adjustment.

[9] The analysis is most straightforward for the Dallas flight of 7 September (Figure 1). The wind direction in the convective boundary layer (CBL) was generally east-northeasterly (66 ± 18°: average and standard deviation here and elsewhere) and steady (4.8 ± 1.2 m/s) throughout the flight track shown. Two double (up and back down) vertical profiles (positions indicated by ovals in Figure 1) indicate that the CBL was well-developed with a relatively stable height near 2.5 km throughout this midday flight. Figure 1 (right) shows the measured vertical profiles with the same color-coding as the flight track. Thus, the profiles conducted at different locations can be distinguished by the color if significantly different ozone concentrations were measured. The measured ozone concentration varied across the upwind transect, and was approximately 8.5 ppbv higher on the north side of the city. This difference is preserved as the air passes over the Dallas urban area as indicated by the difference in the two vertical profiles (Figure 1, right). The best estimate of the background ozone concentration (62.3 ppbv) transported into the DFW urban area is taken to be the average of the ozone measured during the two indicated flight segments on the upwind transect. The ozone concentration between these two segments is not included in the background average since it was higher, presumably owing to photochemical formation from local precursors emitted over the outskirts of the Dallas urban area. The downwind urban plume reached maximum average ozone concentrations of 99 ppbv in the central ≈75 km width of the plume transect conducted near 14:00 local standard time (LST), and the maximum 8-h average ozone (84 ppbv) was reported at the Fort Worth Northwest monitor on the downwind side of DFW within that observed ozone plume.

[10] For the Houston flight of 6 September (Figure 2) evaluation of the effect of increasing CBL depth must be made. In the boundary layer the wind direction was again generally east-northeasterly (69 ± 18°), with the wind speed gradually increasing from 4.6 ± 1.2 m/s on the east transects to 7.2 ± 1.3 m/s on the west transects. The average ozone concentration in the CBL and in the marine boundary layer (MBL) through the upwind transect were quite similar as indicated in Figure 2. However, above the top of the boundary layer at about 1.3 km, the ozone concentration was somewhat larger, and over the urban area the CBL increased to about 2 km depth. Since the air above the boundary layer is mixed down as the boundary layer grows, a height-weighted average of the ozone concentration through the lower 2 km on the upwind transect is taken as the best estimate of the background ozone concentration (65.4 ppbv) transported into the HGB urban area. A similar

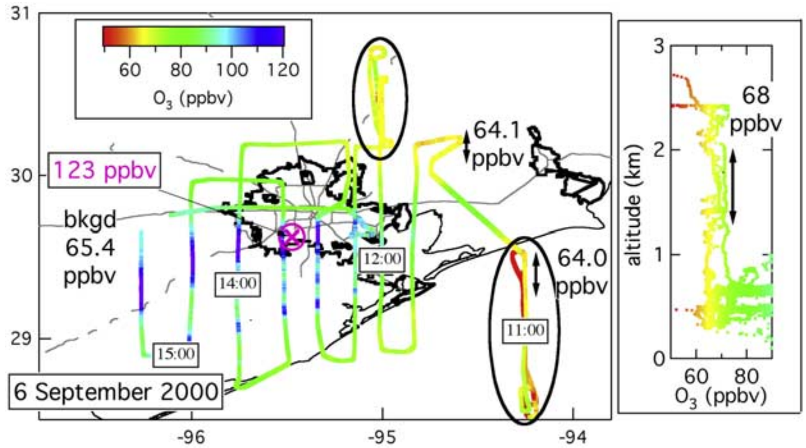

Figure 2. Path of aircraft flight over the Houston−Galveston Bay area and vertical profile data on 6 September 2000 in same format as Figure 1. The prevailing wind during the flight was east-northeasterly. On this flight, the background ozone concentrations aloft were somewhat larger than within the boundary layer. The arrow in the right-hand plot indicates the air aloft that was estimated to have mixed down into the boundary layer as the air moved across the urban area. A height-weighted average of the ozone concentrations in the boundary layer and aloft yielded the indicated background ozone concentration.

evaluation of the depth of the CBL upwind and over the urban area was made for each flight, and all estimated background ozone values are a height-weighted average over the CBL depth over the urban area. In Figure 2 a clearly defined ozone plume propagated downwind from the Houston Ship Channel (HSC) area. During the transect conducted near 14:00 LST the plume averaged 134 ppbv ozone over a width of about 16 km (much narrower than the relatively broad Dallas plume in Figure 1), and reached a maximum 1-s average ozone concentration of 157 ppbv. The maximum 8-h average ozone (123 ppbv) was reported from a station on the downwind side of HGB directly in the path of the observed ozone plume.

[11] For the Dallas flight of 23 August (Figure 3) determination of the appropriate background ozone concentration transported into the DFW urban area is more difficult because a strong gradient in the ozone concentration was observed during the upwind transect. On this flight the boundary layer winds were south-southwesterly (156 ± 25°) and the speed increased from 2.3 ± 0.9 m/s on the first transect to 5.8 ± 1.3 m/s on the later two transects. There was a difference of 23 ppbv in the average ozone concentration between the two upwind flight track segments illustrated. The average of these two averages (with a small correction for the illustrated change in the CBL depth) is taken as the best approximation of the background ozone concentration (71.4 ppbv) transported into the DFW urban area. Again, the Dallas urban plume was broad, averaging 119 ppbv over about 70 km width (maximum 1-s ozone average concentration of 129 ppbv) in the transect with the strongest violet color, and the maximum 8-h average ozone (98 ppbv) was reported from a station on the north side of DFW. This maximum 8-h average ozone is clearly located to the north side of the urban area where the background air with the highest ozone concentrations entered the city.

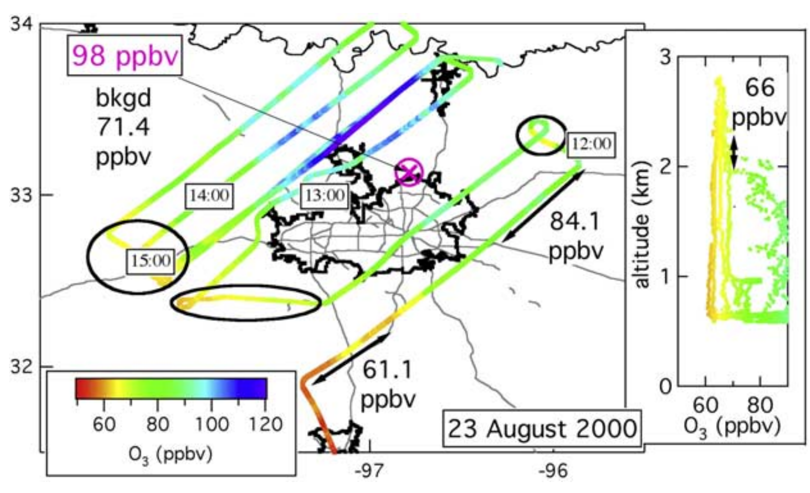

Figure 3. Path of aircraft flight over the Dallas−Fort Worth area and vertical profile data on 23 August 2000 in same format as Figure 1. The prevailing wind during the flight was south-southeasterly.

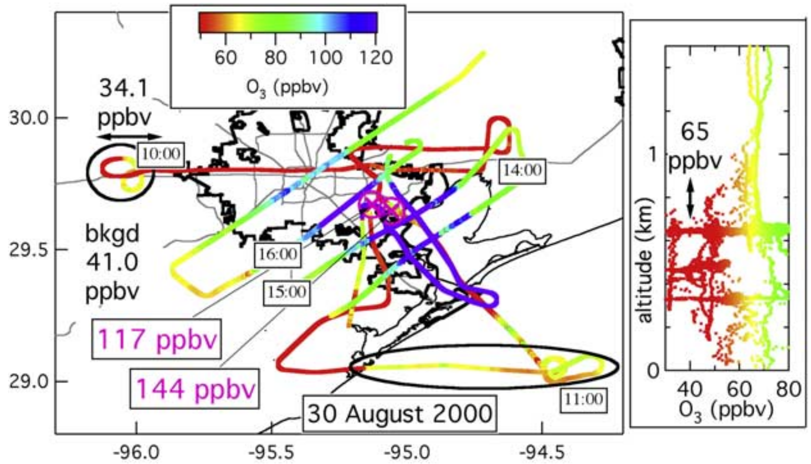

Figure 4. Path of aircraft flight over the Houston–Galveston Bay area and vertical profile data on 30 August 2000 in same format as Figure 1. The prevailing wind during the flight was generally northwesterly and quite slow.

[12] On the Houston flight of 30 August (Figure 4) a lower background ozone concentration was observed, but very high maximum ozone concentrations were reached during the day. Overall the winds were from the northwest, but they were quite variable owing to a land-sea breeze cycle that developed on that day. The ozone concentrations northwest of the HGB urban area were low (34.1 ppbv) but higher aloft (65 ppbv), which yields 41.0 ppbv as the best estimate of the background ozone concentration transported into the HGB urban area. The urban plume reached maximum 1-s average ozone concentrations of 223 ppbv very close to the two surface stations that recorded the highest maximum 8-h average ozone concentrations (117 and 144 ppbv). These maximum 8-h average ozone concentrations occurred very close to the HSC under these light and variable wind conditions. *Banta et al.* [2005] discuss the role of small-scale meteorological processes that lead to the production of these very high, local ozone concentrations.

[13] The background ozone concentration transported into the HGB or DFW urban area was calculated in a similar manner for each of the 21 flights that investigated these two urban areas. In each case, flight segments from an upwind transect provided an estimate of the background ozone concentration in the CBL. In cases when the CBL reached to higher elevations over the urban area than was the case upwind, then an height-weighted average of the ozone concentration within and above the CBL in the upwind region was taken as the best estimate of the background ozone concentration transported into the respective urban area.

2.2. Ground Level Monitoring Data

[14] The Texas Commission on Environmental Quality (TCEQ) in cooperation with other state and local entities maintains an extensive network of Continuous Ambient Monitoring Stations (CAMS) in Houston and Dallas. These monitors measure ambient ozone concentrations. The TCEQ calculates the daily maximum 8-h average ozone for each CAMS monitor according to EPA guidelines and places these averages on their website (http://www.tceq.state.tx.us/cgi-bin/compliance/monops/8hr_monthly.pl). The background ozone concentrations estimated from the aircraft data were subtracted from the TCEQ monitor network daily maximum 8-h average ozone to determine the local contribution of the metropolitan area to peak ozone on each flight day. The aircraft estimates of the regional background ozone are compared with estimates of the regional background determined from surface monitor measurements.

[15] The Houston and Dallas surface monitoring networks are relatively dense, with monitors located on the perimeter of each metropolitan area as well as in the urban core. Depending upon the wind direction, specific outlying monitors may be used to determine the regional background ozone concentration upwind of the city. *Nielsen-Gammon et al.* [2005] have investigated which outlying monitors provide a reliable estimate of regional background ozone. They excluded some monitors in each area that were determined to be overly influenced by local sources of emissions. For example, on the days of the 8 TexAQS 2000 aircraft flights over Houston, the Clute ozone monitor always recorded the lowest maximum 8-h average ozone regardless of wind direction, and was often significantly lower (by as much as 24 ppb) than the monitor recording the next lowest maximum 8-h average ozone value. *Nielsen-Gammon et al.* [2005] found that the Clute monitor is likely influenced by emissions from a nearby petrochemical processing facility, and does not give reliable estimates of the background ozone. *Nielsen-Gammon et al.* [2005] have developed a list of monitors in the Dallas and Houston areas that are relatively free of the strong influence of fresh emissions from local sources, are located on the periphery of the metropolitan areas, and may be used to estimate the background ozone. The lowest maximum 8-h ozone reading among the monitors on this list on a given day was taken to be representative of the regional background; these values were used for comparison with aircraft- and model-derived estimates of background ozone. The monitors used to estimate the background ozone are shown in Figure 5.

2.2.1. Photochemical Grid Model

[16] In this section, we present an overview of the Comprehensive Air Quality Model with Extensions (CAMx) photochemical grid model [*Environ*, 2007], which was used to investigate the relative contributions of regional and local ozone in the Dallas–Fort Worth area. CAMx is a

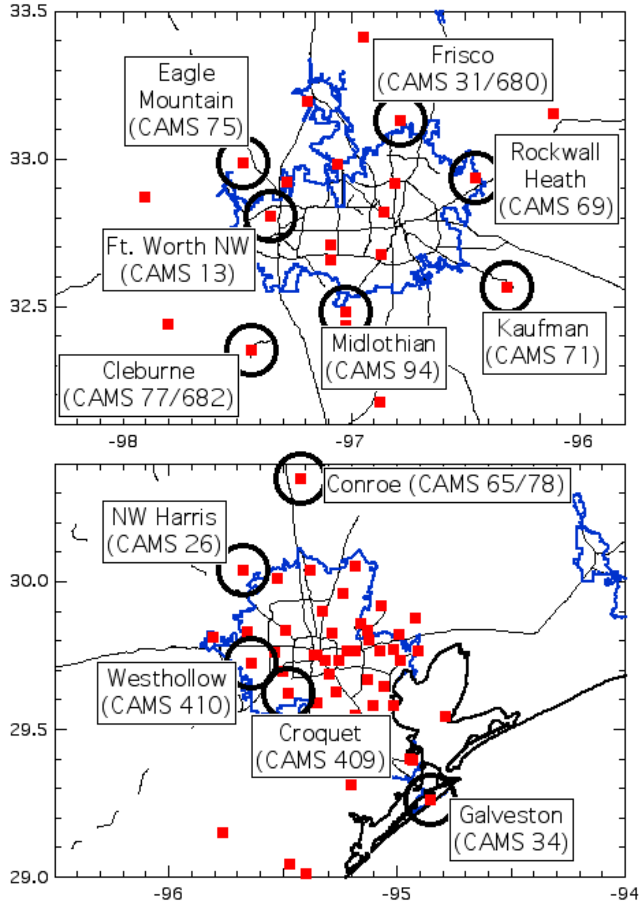

Figure 5. TCEQ surface ozone monitors in the (top) Dallas–Fort Worth and (bottom) Houston–Galveston Bay metropolitan areas. Monitors used to determine background ozone concentrations in this analysis are circled in black.

three-dimensional photochemical grid model, and simulates physical and chemical processes governing the formation and transport of ozone, particulates, and toxics in the troposphere. Version 4.30 of CAMx, which is available at www.camx.com, was used for this study. The model was exercised on two nested domains shown in Figure 6. A Lambert Conformal grid with horizontal resolution of 36 km covered much of the eastern United States. A two-way nested fine grid was placed over East Texas and nearby states; the horizontal resolution of each cell on the fine grid was 12 km × 12 km. The layer depth varied with height. The model's vertical resolution was finest near the ground, with a 36 m surface layer, and extended to the lower stratosphere in 20 layers. The vertical structure was the same on the 36 km and 12 km grids.

[17] The CAMx model input data were originally developed for regulatory modeling of visibility and particulate matter in the Central U.S. Meteorological data for CAMx were developed using the Pennsylvania State University/National Center for Atmosphere Research Mesoscale Model version 5 (MM5) [*Dudhia*, 1993] for the Central Regional Air Planning Association (CENRAP) by the Iowa Department of Natural Resources using the MM5 [*Johnson*, 2004]. The MM5 provides CAMx with hourly, gridded data for wind vectors, pressure, temperature, diffusivity, humidity, clouds and precipitation. Emissions of VOC, NO_x, CO, SO_2, NH_3 and aerosols were developed from the Environmental Protection Agency's 2002 National Emissions Inventory. Biogenic emissions were determined using the GloBEIS model [*Yarwood et al.*, 2003] with MM5 temperatures and solar radiation derived from analysis of GOES satellite data and land use/land cover data developed by the TCEQ. The modeling was performed using the Carbon Bond 4 chemical mechanism [*Gery et al.*, 1989] with updates to extend the inorganic reactions and add NO_x recycling reactions [*Yarwood et al.*, 2005].

[18] Model ozone transport was analyzed using the CAMx Anthropogenic Precursor Culpability Assessment (APCA) ozone source apportionment tool [*Environ*, 2007]. APCA uses tracers (tagged species) to track precursor emissions and ozone formation within the CAMx model and can attribute ozone production by geographic region and emissions category [*Dunker*, 2002; *Environ*, 2007]. Tracers are emitted along with ozone precursors, and travel with them as the precursors are transported within the modeling domain. If the emitted precursors are involved in ozone formation, ozone tracers are generated in the grid cell where that formation occurs, and the ozone tracers are then tracked as well, undergoing transport and destruction along with the modeled ozone. The precursor and ozone tracers are spectators to the model integration, and do not affect its outcome; their source-receptor relationships are consistent with those of the chemical species they follow in the model. In this way, the source apportionment tool takes into account the nonlinear photochemistry of ozone formation and the mechanisms of ozone transport and destruction.

[19] APCA differs from the standard CAMx Ozone Source Apportionment Tool (OSAT) [*Environ*, 2007] in recognizing that certain emission groups are not controllable (e.g., biogenic emissions) and that apportioning ozone production to these groups does not provide information that is relevant to development of control strategies. To address this, in situations where OSAT would attribute ozone production to noncontrollable (i.e., biogenic) emissions, APCA reallocates that ozone production to the controllable portion of precursors that participated in ozone formation with the noncontrollable precursor. For example, when ozone formation is due to biogenic VOC and anthropogenic NO_x under VOC-limited conditions (a situation in which OSAT would attribute ozone production to biogenic VOC), APCA redirects that attribution to the anthropogenic NO_x precursors present. The use of APCA instead of OSAT results in more ozone formation attributed to anthropogenic NO_x sources and less ozone formation attributed to biogenic VOC sources, but generally does not change the partitioning of ozone attributed to local sources and the transported background for a given receptor.

3. Results

3.1. Aircraft Results for Houston

[20] Aircraft measurements were made in the Houston–Galveston Bay (HGB) area on 17 days during 2000 and 2006. Of these 17 days, 10 qualified as 8-h ozone exceedance days since the daily maximum 8-h average ozone in the HGB area was greater than 84 ppbv. Figures 7 and 8 show the apportionment of the maximum 8-h average

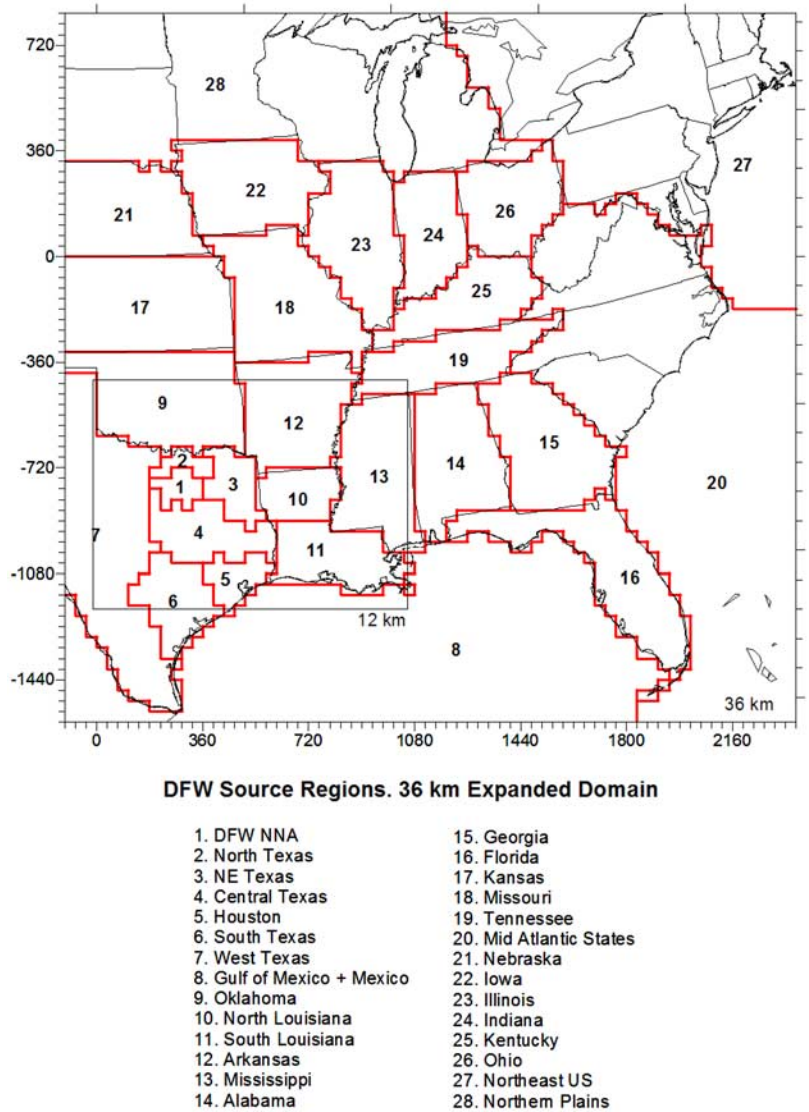

Figure 6. CAMx 36 km modeling domain, the nested 12 km modeling domain, and the 28 source areas used in the ozone transport analysis.

ozone measured by the TCEQ monitoring network into contributions from local sources and regional transport for all 17 days when aircraft measurements are available. Data from the TexAQS 2000 flights are shown by the green symbols, and data from the TexAQS II 2006 flights are shown by red symbols. In Figure 7, the vertical axis shows the maximum 8-h ozone concentration among all monitors in the TCEQ monitoring network in the HGB area on the day of the aircraft flight; the horizontal axis shows the calculated contribution from local HGB sources derived from the difference between the measured 8-h peak and the background determined from aircraft measurements as described above. The local contribution ranged from 8 to 103 ppbv on the flight days. There is a significant contribution from regional background ozone, which ranged from a minimum of 22 ppbv to a maximum of 72 ppbv. This background maximum concentration can lead to an exceedance of the 8-h ozone NAAQS with a relatively small additional contribution of 13 ppbv from local sources, neglecting removal and destruction of the transported ozone. The linear least squares fits give slopes near unity for the separate years (0.93 with $r^2 = 0.77$ for 2000 and 0.91 with $r^2 = 0.17$ for 2006), and for the total data sets (1.0 with $r^2 = 0.68$). The fact that the slopes are near unity suggests that the local and transported background contributions are largely independent of one another in the HGB area.

[21] Figure 8 shows the local contribution and the transported contribution to the daily maximum 8-h ozone over HGB on the day of the flights. Data from the TCEQ monitors as well as the aircraft measurements are displayed. $r^2 = 0.0004$ for the entire aircraft data set, and $r^2 = 0.07$ for the TCEQ data. The small values of r^2 in Figure 8 support the finding noted above that the local and transported background contributions are independent for the entire data set on the HGB flight days.

[22] We averaged the values of the local and background contributions derived from the aircraft data for all flights that occurred on exceedance days in 2000 and 2006 in order

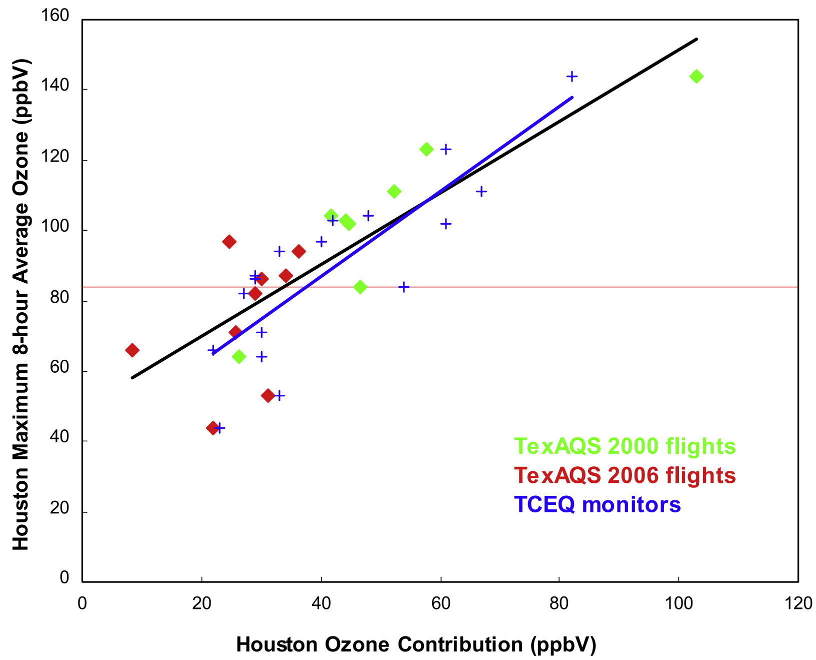

Figure 7. Vertical axis shows the measured peak 8-h average O_3 in the Houston–Galveston Bay (HGB) area measured by the TCEQ monitor network as a function of the local HGB contribution to that peak (horizontal axis). The HGB contribution is derived from the difference between the measured peak 8-h average and the background. For the aircraft data points, the background ozone was determined from aircraft transects upwind and across the HGB region on each of 17 days of aircraft flights in 2000 and 2006. Aircraft data from 2000 (2006) are indicated by green (red) diamonds. The blue crosses represent data from the TCEQ monitor network. For the TCEQ monitor data, the background contribution was derived from measurements by monitors located upwind of the urban area. The black line is the best fit line for the entire aircraft data set including both 2000 and 2006. The best fit line for the TCEQ monitor data is shown in blue. The red dashed line shows the 84 ppbv ozone level required for an exceedance of the NAAQS.

to determine the average contribution from local and transported ozone on high-ozone days. During 2000, local and transported ozone each contributed about 57 ppbv to the average total 8-h ozone maximum of 115 ppbv on the six Houston exceedance days investigated by aircraft measurements. Thus, local emissions and transport each contributed about equally to these average 8-h ozone exceedances. During 2006, there were four exceedance days investigated by aircraft measurements. TCEQ surface monitoring network measurements for the HGB area show that the average daily maximum 8-h ozone for these 4 days was 91 ppbv. HGB area emissions contributed an average of 31 ppbv on these four exceedance days and transported ozone contributed an average of 60 ppbv. In 2006, the transported contribution was slightly larger and the local contribution was significantly smaller than in 2000. The results for 2000 and 2006 indicate that, on average, transported ozone alone brought the Houston area within 25–30 ppbv of an exceedance of the 8-h ozone standard.

[23] Comparison of results when the regional background is derived from the TCEQ surface monitor data (Figure 7) shows reasonable agreement (i.e., within 10 ppbv) on most of the aircraft flight days, with 30 August 2000 (the day with the highest maximum 8-h average) a notable exception. On this day, the estimated background from the lowest of the perimeter monitors was 62 ppbv, while the aircraft

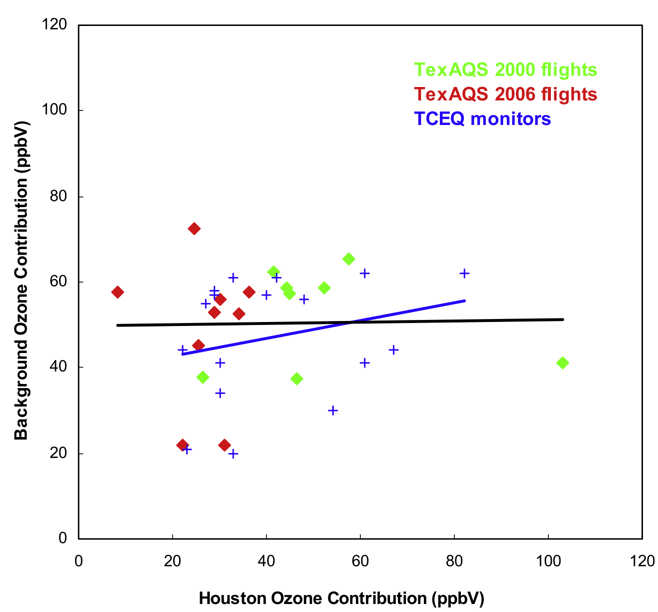

Figure 8. As in Figure 7, except vertical axis shows 8-h average background O_3 in the HGB area on the day of the flights as derived from aircraft and TCEQ monitor data.

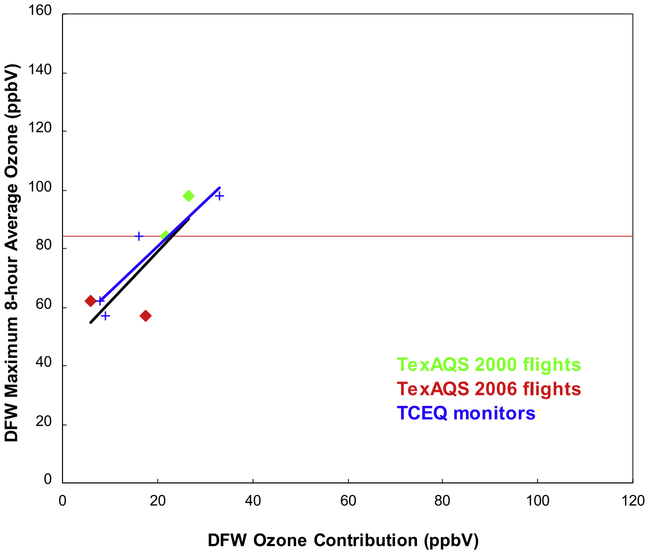

Figure 9. As in Figure 7, for the Dallas–Fort Worth flight days in 2000 and 2006.

measured background ozone of 41 ppbv. Note that Houston's complex meteorology, with its sea breeze that tends to recirculate pollutants, can make isolation of the background ozone from surface monitors very difficult. The slopes of the best fit lines for the monitor and aircraft data are within 20% of one another (1.0 versus 1.2).

[24] We average the monitor data for the 2000 and 2006 exceedance days that coincided with aircraft flights, as was done with the aircraft data, above. For 2000, the average background contribution derived from the monitor data was 54 ppbv, which is within 3 ppbv of the aircraft estimate of this quantity. The average local contribution estimated from the monitor data is 60 ppbv, which is likewise within 3 ppbv of the average local contribution derived from aircraft data. For 2006, the monitor data indicate that contributions from local sources and the transported background are 33 and 58 ppbv, respectively, and the corresponding values from the aircraft data are 31 ppbv and 60 ppbv. The aircraft and monitor data, therefore, show good agreement on the relative contributions of local sources and the regional background to the average exceedance day in both 2000 and 2006.

3.2. Aircraft Results for Dallas

[25] Only four flights were made over the Dallas–Fort Worth (DFW) region, two in 2000 and two in 2006. The only one of these days when the DFW area had an exceedance of the ozone NAAQS with a peak value of 98 ppbv is 23 August 2000. On that day, the background ozone transported into Dallas was 71 ppbv or 72% of the total measured maximum 8-h ozone concentration. The 71 ppbv contribution due to transport brought the monitor to within 13 ppbv of the 84 ppbv NAAQS.

[26] The results from the DFW aircraft flights are summarized in Figure 9. Results from the 23 August and 7 September 2000 flights are shown by the green symbols, and results from the 13 and 25 September 2006 flights are shown by red symbols. Peak ozone values were higher in 2000 than in 2006, but this can be largely attributed to meteorological differences rather than an improvement in overall air quality in the DFW area. The 2006 flight days were cooler and cloudier than those in 2000, and overall maximum ozone levels have changed only very little since 2000 (see page 18 of the Final Rapid Science Synthesis Report: Findings from the Second Texas Air Quality Study (TexAQS II) www.tceq.state.tx.us/assets/public/implementation/air/am/texaqs/rsst_final_report.pdf). Each of the 4 days shows a significant impact from regional transport, which ranged from 40 to 71 ppbv, while the local contribution ranged from 17 to 27 ppbv. As in Houston, the aircraft data show that transported ozone can bring the Dallas area close to an 8-h exceedance before the additional contribution from local emissions. The positive slope in Figure 9 shows that the value of the 8-h daily maximum ozone increases as the transported background ozone increases. Note that it is not possible to draw broad conclusions regarding the relative importance of the transported and local contributions from the DFW flights because of the small number of data points.

[27] As for Houston, comparison with TCEQ monitor data shows reasonable agreement. The maximum difference between the aircraft and monitor-derived estimates of the background ozone was 8 ppbv. The slopes (1.7 with r^2 = 0.62 for the aircraft data versus 1.5 for the monitor data) agree within 12%. This suggests that the aircraft and monitor estimates of the regional background are reasonably consistent. The slopes for both the aircraft and monitor data are both significantly different from 1, suggesting that the local and transported contributions are not independent, as was the case for the Houston flight days. When the Dallas local and transported contributions are plotted against one another as was done for Houston in Figure 8, r^2 = 0.22 for the aircraft data and r^2 = 0.46 for the TCEQ monitors (Figure 10). This confirms that on the aircraft flight days over Dallas, the local and transported contributions were not independent of one another.

3.3. Modeling Results for Dallas

[28] The CAMx model has been used to investigate the origin of transported ozone and precursors from regions outside of the Dallas–Fort Worth area. The benefit of using a model is that source attribution for an ozone exceedance at a particular monitor is possible. The contributions of local and regional sources can then be separated and compared, and the model contributions can be compared with those derived above from the aircraft and surface monitoring data.

3.3.1. Source Apportionment for a Single Exceedance Day

[29] We focus first on a single DFW 8-h ozone exceedance day (7 August 2002) in order to show an example of source apportionment using CAMx. The CAMx APCA tool was used to perform source apportionment at the time of peak modeled ozone at the DFW area monitor with the highest 8-h ozone concentration. Figure 11b displays the modeled ozone transport contributions from the major source regions; for the sake of clarity, we include only source regions that contributed ≥ 1 ppbv ozone. The source regions are shown in Figure 6.

[30] We compare air parcel back trajectories with the APCA analysis in order to assess whether there is agreement between the back trajectories and the model's estimate

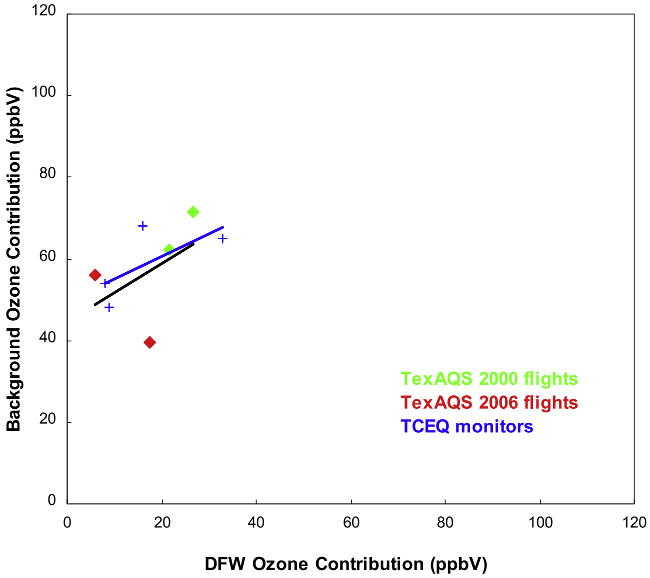

Figure 10. As in Figure 8, for the Dallas–Fort Worth flight days in 2000 and 2006.

of the importance of regional transport from a particular region. The back trajectories shown in Figure 11a were prepared using online tools provided by the National Oceanic and Atmospheric Administration (NOAA) at http://www.arl.noaa.gov/ready/hysplit4.html (R. R. Draxler and G. D. Rolph, HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) Model, 2003). These tools are based on application of NOAA's HYSPLIT model with archived weather forecast model data. The back trajectories were computed using wind data from the National Center for Environmental Prediction's EDAS forecast model which have 80 km resolution. The trajectories extend 72 h backward in time. Trajectories for air parcels ending at 500 m, 1000 m, and 2000 m were calculated in order to assess the importance of vertical wind shear.

[31] On 7 August 2002, the peak ozone measured by the regional monitoring network was 106 ppbv at the Fort Worth NW monitor at 1400 local time. The background concentration was approximately 69 ppbv as measured at the Frisco monitor. The local DFW contribution derived from the TCEQ monitor data is therefore 37 ppbv. The modeled peak of 95 ppbv is 11 ppbv lower than the observed peak. The modeled DFW contribution of 36 ppbv agrees well with the monitor-derived DFW contribution, and shows that the model's underestimate of the peak ozone is due to its underprediction of the contribution of the transported background ozone (59 ppbv predicted versus 69 ppbv at the Frisco monitor).

[32] The HYSPLIT 5-day back trajectories cross Northeast Texas, Arkansas, and Tennessee, extending to the mid-Atlantic region; the CAMx modeling finds these same regions contributing to the Dallas area 8-h ozone exceedance at the Fort Worth NW monitor on 7 August 2002. There is also a significant contribution from biogenics. For 7 August as well as other exceedance days in 2002, there was good agreement between the APCA source region attribution and back trajectories calculated with HYSPLIT.

3.3.2. CAMx Modeling of the 2002 Ozone Season

[33] The CAMx model was run for the period extending from 1 June to 30 September 2002. Although this period does not correspond to the time of the aircraft observations, it was used because of the availability of recently developed, high-quality emissions and meteorological databases for the entire 2002 ozone season. The fact that the aircraft and model data are from different years means that no direct day-by-day comparison is possible, and is a source of

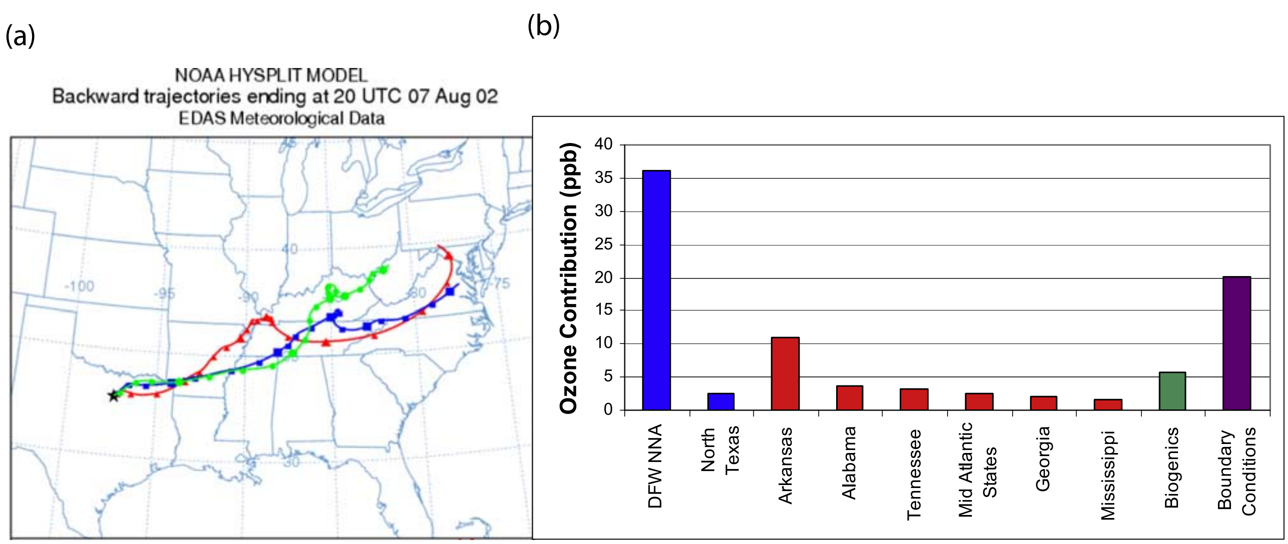

Figure 11. (a) Trajectories from three different altitudes over Fort Worth to characterize upwind areas for the column of air at 2000 UTC on 7 August 2002. The star shows the location of the Fort Worth NW monitor. Red trajectory ends at 500 m altitude. Blue (green) trajectory ends at 1000 m (2000 m) altitude. (b) CAMx APCA source apportionment showing contributions to the modeled total ozone concentration at the Fort Worth NW monitor at the time of the maximum 8-h ozone concentration on 7 August 2002. Source regions are defined in Figure 6.

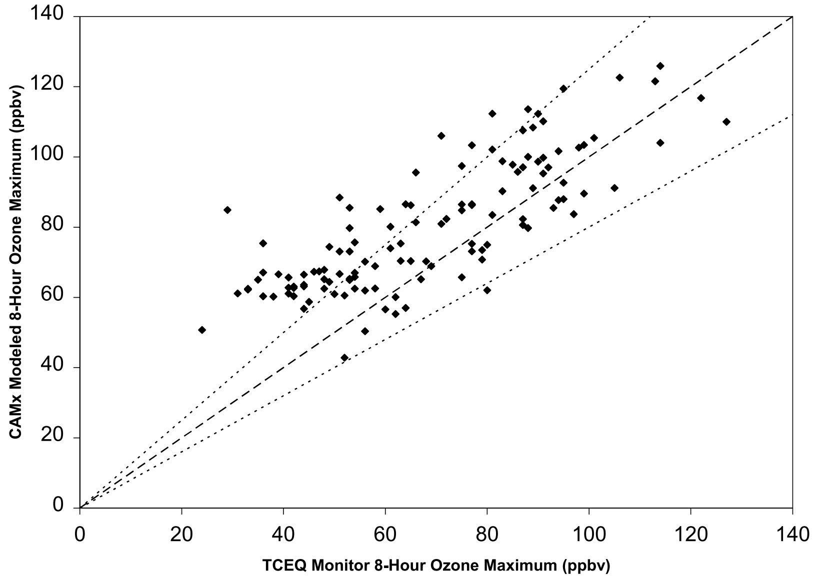

Figure 12. CAMx model performance evaluation for 1 June to 30 September 2002. Each point on the plot corresponds to an observed and modeled concentration pair at the specific monitor and time that reported the day's maximum 8-h average ozone. Heavy dashed line is the 1:1 line indicating agreement between modeled and observed values. Lighter dashed lines indicate deviations of ±20%.

uncertainty in this analysis. Model performance for ozone and some precursors was evaluated in detail and was found to be reasonably good for the DFW area and Northeast Texas on days of interest when the maximum 8-h ozone was higher than 80 ppbv [*Yarwood et al.*, 2006; *Kemball-Cook et al.*, 2006]. Figure 12 shows a measure of the model performance on the 12 km grid in the DFW region. For each day during the entire summer modeling period, the modeled and observed DFW area maximum 8-h ozone are plotted against one another. Each point on the plot corresponds to an observed and modeled concentration pair at the specific monitor and time that reported the day's maximum 8-h average ozone. In Figure 12, good performance would be characterized by most of the scatterplot points lying close to the 1:1 line and preferably within the ± 20% lines. The model tends to overpredict the daily peak ozone values, and this tendency is more pronounced at monitored values below 80 ppbv. When the monitored values are higher than 80 ppbv, the modeled/observed points generally fell within the ± 20% lines.

[34] CAMx was run with a constant boundary condition of 40 ppbv ozone, which makes it difficult for the model to reproduce observed ozone concentrations on days when particularly clean air was transported into the Dallas area. In Figure 12, there are more than 25 data points with 8-h average ozone from the TCEQ monitors lower than 55 ppbv, but CAMx simulated ozone this low for very few of them. To accurately simulate low-ozone days, a time-varying boundary condition based on observed conditions or a global model simulation would be required.

[35] Local and regional transport contributions to Dallas area ozone were quantified for the summer ozone season and the contributions of individual geographic regions were calculated using the APCA source apportionment tool. The source regions used in the APCA analysis are shown in Figure 6. For each day during the summer of 2002, the APCA tool was applied for the time of the maximum modeled 8-h ozone value at the monitor with the highest observed daily maximum 8-h ozone concentration; the contribution of each source region to the peak ozone at that monitor was determined. The APCA source apportionment results are shown along with the aircraft and monitor data in Figure 13. The model result for each day is shown as a purple diamond; for these points, the vertical axis is the modeled DFW maximum 8-h average ozone, and the horizontal axis is the modeled local DFW contribution. For the aircraft and monitor data, the vertical axis is the DFW maximum 8-h average ozone derived from the TCEQ monitor network. The monitor data are shown as blue crosses, and aircraft data are as in Figures 7 and 9.

[36] Figure 13 compares the ozone apportionment in the DFW area from 2000 and 2006 aircraft data, the CAMx model results for 2002, and the TCEQ monitor data for 2002. The slopes of the linear fits to the aircraft data (black line, slope = 1.7 with r^2 = 0.62) and to the TCEQ monitor data (blue line, slope = 1.7 with r^2 = 0.66) are in excellent agreement, but this is inconclusive since the aircraft slope is poorly defined by the four available flights and the limited surface monitoring network may not have the spatial resolution to characterize either the upwind background or downwind maximum ozone on any given day. The aircraft line does lie about 15 ppbv above the TCEQ monitor data. However, Figure 9 shows good agreement between the results from the aircraft and the TCEQ monitor data on the specific days of the flights, which indicates that the aircraft data were all collected on days of particularly large contributions from regional transport. The slope of the linear fit to the CAMx APCA data (purple line, slope = 1.2 with r^2 = 0.88) is approximately 30% lower than those derived from the measurements. This lower slope is due to both the tendency of the model to overpredict the daily peak ozone values particularly below 80 ppbv (see discussion of

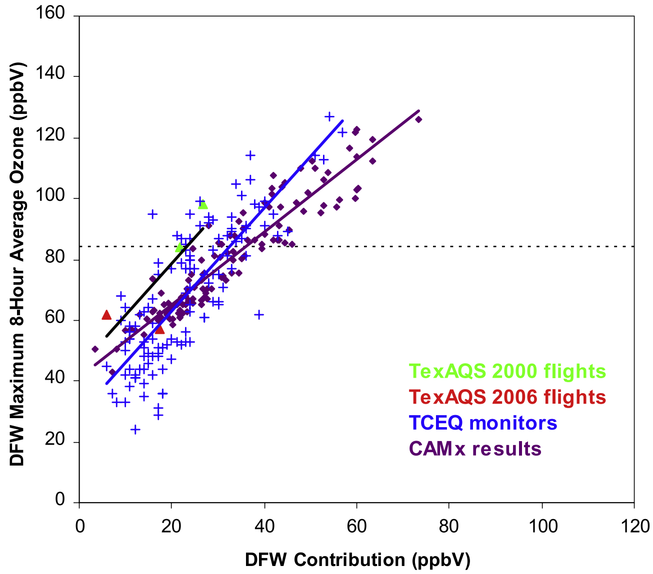

Figure 13. Measured and modeled peak 8-h average O_3 in the DFW area as a function of the DFW contribution to that peak. Purple symbols show modeled maximum 8-h average ozone and the corresponding DFW contribution from the APCA method for each day 1 June to 30 September 2002. Data from aircraft transects upwind and across the DFW region on 4 days in 2000 (2006) are shown in green (red) as in Figure 9. The linear least squares best fit line for the 2000 and 2006 aircraft (APCA) data is shown in solid black (purple). The blue crosses represent data from the TCEQ monitor network for each day 1 June to 30 September 2002. The best fit line for the TCEQ monitor data is shown in blue. Black dashed line shows the 84 ppbv ozone level required for an exceedance of the NAAQS.

Figure 12 above), and a tendency of the model to overpredict the local DFW contribution on days of higher peak ozone, even though it predicts the total peak ozone with reasonable accuracy. For values of maximum 8-h average ozone between approximately 55 ppbv and 90 ppbv, the CAMx results and the monitor data generally overlap. The best fit lines for all three data sets have positive slopes, indicating that the value of the 8-h daily maximum ozone increases as the transported background ozone increases.

[37] Statistical descriptions of a hypothetical average exceedance day for the DFW area can be developed from the monitoring data and from the CAMx APCA model results. The DFW area had 35 days during the period 1 June to 30 September 2002 with monitored 8-h ozone levels greater than 84 ppbv. For these 35 days the average apportionment of the maximum 8-h average ozone between local production and regional transport was derived from the model and the monitoring data. Table 1 compares the results. On average the model overestimated the total ozone by about 5% (101 versus 96 ppbv) and attributed a significantly lower fraction to regional transport (54% versus 65%) and a correspondingly higher fraction to local production (46% versus 35%). This 2002 average exceedance day may be compared with the 23 August 2000 flight, which was the sole aircraft flight made on an ozone exceedance day. This day had a nearly average maximum 8-h ozone average of 98 ppbv, and the aircraft measurements showed an even greater importance of regional transport (72%) compared to local production (28%).

[38] The CAMx APCA model, TCEQ monitoring data and aircraft results all agree that on an average exceedance day, regional transport brings the DFW area to within 20 to 30 ppbv of an exceedance of the NAAQS for ozone. Transport and local production both played a critical role in determining the peak ozone in DFW on exceedance days during 2002.

[39] The APCA results for the average exceedance day may be analyzed to determine the origins of the transported ozone. The average modeled transport contribution from all other parts of Texas was 9 ppbv, and there were days when northeast Texas, the Houston area, south Texas, and central Texas individually made contributions as large as 13 ppbv. The boundary conditions accounted for an additional 20 ppbv. The boundary conditions can be a major contributor because there is assumed to be a tropospheric background ozone concentration of approximately 40 ppbv, so that air with 40 ppbv ozone concentration is constantly advected into the domain through the model boundaries. The APCA boundary contribution is less than 40 ppbv due to chemical destruction of ozone and its deposition to the earth's surface. The average modeled transport contribution from other states was 25 ppbv, and the largest contributing states were Louisiana (4 ppbv) and Arkansas (4 ppbv), with contributions of approximately 2 ppbv each coming from Mississippi, Oklahoma, Alabama, Tennessee, and the mid-Atlantic states.

4. Discussion

[40] We have utilized three approaches to approximately apportion the maximum 8-h average ozone between that transported into, and that produced from local precursor emissions within the Houston and Dallas urban areas. These three approaches include analysis of CAMx model calculations, aircraft measurements and surface monitoring data. The main conclusion from this study is that these two major metropolitan areas in Texas can be brought close to exceeding the 0.080 ppm 8-h ozone standard solely from the ozone contribution from regional transport and before any contribution from local sources. The implementation of the recently revised NAAQS to 0.075 ppm, which effectively reduces the standard by 10 ppbv, will further strengthen this

Table 1. Summary of Ozone Apportionment Between Regional Transport and Local Production on Exceedance Days in the DFW Area

	Average Local Ozone Production	Average Regional Transport of Ozone	Average Maximum 8-h Average Ozone
CAMx model (all 2002)	46 ppbv (46%)	55 ppbv (54%)	101 ppbv
TCEQ monitors (all 2002)	34 ppbv (35%)	62 ppbv (65%)	96 ppbv
Aircraft (23 August 2000)	27 ppbv (28%)	71 ppbv (72%)	98 ppbv

conclusion. These results emphasize the benefits of regional control strategies such as the Clean Air Interstate Rule (CAIR; issued by EPA in 2005 and vacated by the U.S. Court of Appeals in 2008), because local controls, while important, will very likely not be sufficient to ensure attainment of the new 8-h ozone standard in either the Houston or Dallas urban areas.

[41] It is important to note that each of the three apportionment approaches has significant uncertainty, but that the results from the three are reasonably consistent. Comparison to measurements suggests that this CAMx seasonal model tended to overpredict the daily peak ozone values, particularly at lower concentrations, and to bias high the fraction of the total ozone attributed to local production. At least part of this discrepancy is due to the lack of time-varying boundary conditions for the CAMx model. Aircraft measurements during transects upwind of the urban area can quantify the ozone concentration entering the urban area, but there are limits on the accuracy and precision of this quantification. On some occasions (e.g., Figure 3) there are strong gradients in the regional ozone distribution, and the upwind transect represents conditions at the time the transect is made, which may not remain constant over the 8-h averaging period required for the NAAQS calculation. Further, as the height of the CBL evolves during the day, air is mixed down from above, which affects the average ozone concentration transported into the urban area. The ozone vertical profile is usually only characterized from a very small number of vertical profiles during the flight. Analysis of monitoring network measurements also can apportion the observed ozone, but these networks provide only a limited characterization of the urban ozone distribution. Generally, they cannot be instrumented extensively enough to accurately capture the highest maximum 8-h ozone average, particularly in the Houston area, which is characterized by narrow ozone plumes emanating from the concentrated industrial areas in that urban region. Further, the ozone transported into the urban area is taken from the one upwind station with the lowest reported maximum 8-h average, but this average can be affected by local conditions. Nevertheless, the results obtained from these three approaches are in reasonable agreement, as discussed in section 3. This agreement increases our confidence in the conclusion presented above, which is consistent with the results from all three approaches.

[42] Important findings include the following.

[43] 1. On average ozone exceedance days, the regional background ozone transported into both the Houston and Dallas urban areas averaged 55 to 60 ppbv. Thus, each area requires a local net ozone production of only 25 to 30 ppbv to reach exceedance levels. The maximum contribution from transport on a single exceedance day was 75% for Houston and 73% for Dallas.

[44] 2. In the Houston urban area, the ozone concentration transported into the urban area and that produced from local emissions were approximately independent. This independence may reflect the important roles in the Houston-area local ozone production played by the unique ozone precursor mix (i.e., industrial NO_x and highly reactive VOC emissions) and the small-scale meteorological features (e.g., land-sea breeze circulation).

[45] 3. In the Dallas area, the ozone produced from local emissions was higher on days of higher regional background ozone. This dependence may reflect the importance of large-scale meteorological conditions (i.e., stagnant, hot, sunny) in raising both the regional background and local ozone production for the Dallas area.

[46] 4. The CAMx model results show that the regions outside Texas most frequently impacting the Dallas urban area via transport of ozone and precursors during the summer of 2002 were Louisiana, Arkansas, Mississippi, Oklahoma, Alabama, Tennessee, and the mid-Atlantic states, with a smaller total contribution coming from other parts of Texas.

[47] One possible avenue for future work is to extend the model analysis to Houston to take advantage of the fact that the aircraft data sets for both 2000 and 2006 are larger for Houston than for Dallas. This would require development of a new model that would have a different nested grid structure with a fine-scale grid centered over Houston. Note that the model used in the present study was originally designed to focus on the Dallas−Fort Worth area, and is configured accordingly. The synoptic-scale meteorological features that contribute to the occurrence of high-ozone days in Dallas−Fort Worth are well-resolved at the model's present 12 km grid size, as are on-road mobile sources of emissions, which are the main source of emissions in the area. Houston, on the other hand, has a more complicated meteorology that includes a local-scale sea breeze circulation. The sea breeze circulation cannot be accurately simulated at 12 km resolution. In addition, ozone formation in Houston is strongly influenced by local point source emissions of highly reactive VOCs; simulating ozone over Houston requires model resolution of 4 km or higher to describe the fate of these point source emissions with reasonable accuracy. A new emission inventory incorporating adjusted highly reactive VOC emissions from Houston industrial sources would be required, as well as a new, higher resolution meteorological database.

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