

Airborne lidar measurements of ozone flux downwind of Houston and Dallas

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[1] We use airborne lidar measurements of ozone collected during the Texas Air Quality Study (TexAQS) 2000 and TexAQS 2006 field campaigns to compute the horizontal flux of ozone downwind of the Houston and Dallas/Fort Worth metropolitan areas. Fluxes are computed for each aircraft transect by integrating excess ozone (plume ozone minus background ozone) in the urban plumes and multiplying the result by the horizontal wind speed provided by radar wind profilers. In addition, we use the lidar data to estimate ozone production rates and ozone enhancements in the Houston and Dallas/Fort Worth plumes. We found that the average horizontal flux of ozone emanating from the Houston area based on data from six research flights was $3.2 \cdot 10^{26}$ molecules per second. This was significantly higher than the flux measured downwind of Dallas/Fort Worth during a single flight. The Houston fluxes exhibited a strong dependence on wind direction. Under southerly or northerly flow, ozone fluxes were about twice as large as under westerly or easterly flow conditions. We estimate that a day's worth of export of ozone from the Houston area could raise regional background ozone by about 10 ppbv over a 40,000 km² area. This has important ramifications for air quality in communities downwind of Houston as it could raise background ozone levels enough that regions with little or no local pollution sources of their own may violate the federally mandated ozone standard.

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1. Introduction

[2] The two largest urban areas in Texas, Houston and Dallas/Fort Worth, often experience high ozone pollution events during the summer months. Compared to Dallas/Fort Worth or other large cities in the United States, high ozone events in the Houston area tend to be more frequent and severe [Kleinman *et al.*, 2002]. Houston is unique among metropolitan areas in the United States because it is home to a large concentration of petrochemical plants. These are predominantly located along the Houston Ship Channel (referred to as Ship Channel hereafter), which runs from the northwest corner of Galveston Bay to the eastern edge of downtown Houston (Figure 1, inset). These refinery complexes coemit highly reactive volatile organic compounds and nitrogen oxides (NO_x), which cause rapid and very efficient ozone formation during the summer months [Daum *et al.*, 2004; Ryerson *et al.*, 2003]. The most severe ozone episodes typically occur under stagnant conditions or, in the case of the Houston area, when pollutants are recirculated by the land-sea breeze system [Banta *et al.*, 2005; Darby, 2005]. However, even under steady flow conditions, when the

Houston and Dallas/Fort Worth urban areas are rather well ventilated, high ozone levels can occur downwind [Ryerson *et al.*, 2003]. Often, these high-ozone events escape detection because routine pollution monitoring stations are predominantly located in and near the urban areas. Ozone plumes emanating from the Houston and Dallas/Fort Worth urban areas add to the ozone loading downwind. Initially, the area impacted by the ozone plume tends to be rather limited, but eventually the plume is distributed over a larger area, often by nighttime transport processes [Tucker *et al.*, 2010; Banta *et al.*, 2005], and becomes part of the regional background. Background ozone in eastern Texas is highly variable and strongly influenced by synoptic scale transport [Langford *et al.*, 2009; Hardesty *et al.*, 2008; Nielsen-Gammon *et al.*, 2005]. Under certain flow conditions, background levels may be high enough that the ozone added by the Houston or Dallas plumes could cause a violation of the 8 h ozone standard. Thus, small to medium-sized urban areas that lie downwind of Houston or Dallas/Fort Worth, and even rural areas without any local pollution sources, may be pushed into noncompliance with the National Ambient Air Quality Standard (NAAQS) [EPA, 2008].

[3] The Houston and Dallas/Fort Worth pollution plumes have been examined rather extensively in the last decade as part of two large air-quality studies: the Texas Air Quality Study (TexAQS) 2000 and TexAQS 2006 [Parrish *et al.*, 2009]. In particular, ozone production rates and efficiencies were measured and compared to those of other urban or

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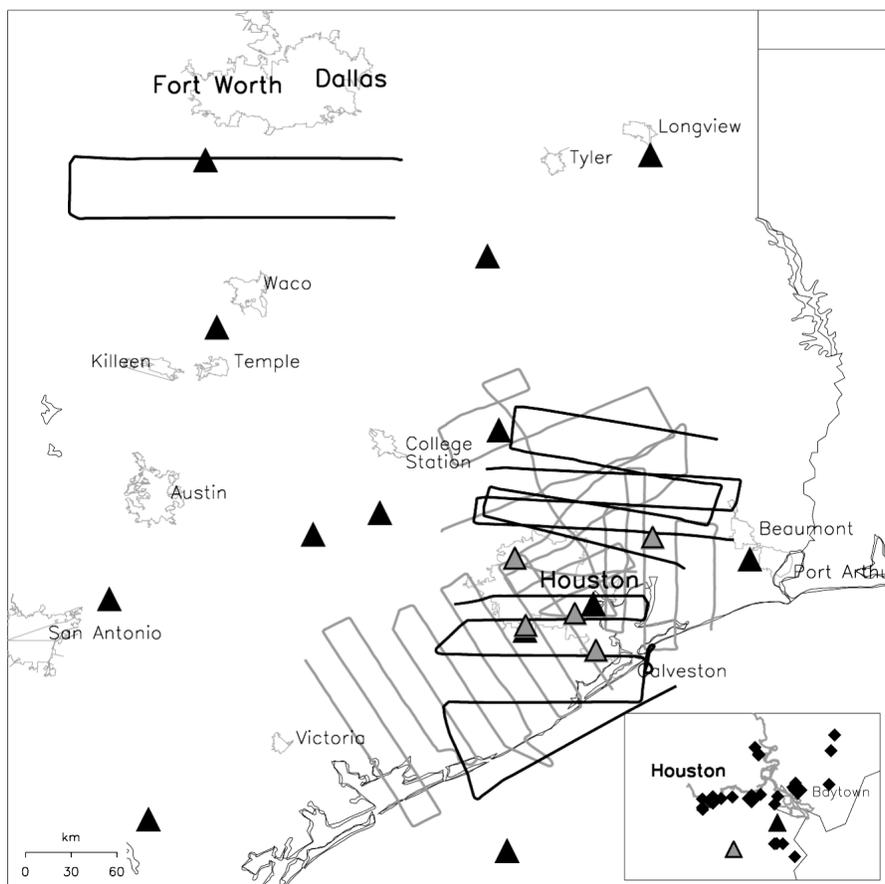


Figure 1. Flight tracks for the days chosen for ozone plume flux retrieval from the TexAQS 2000 study (gray lines) and the TexAQS 2006 study (black lines). Locations of radar wind profilers are indicated by filled triangles (gray, TexAQS 2000; black, TexAQS 2006). The inset shows the high concentration of petrochemical plants (filled diamonds) along the Ship Channel between downtown Houston to the west and Baytown to the east. The size of the diamond-shaped symbols scales with NO_x emission rates.

point source plumes [Neuman *et al.*, 2009; Daum *et al.*, 2004; Ryerson *et al.*, 2003; Kleinman *et al.*, 2002]. In this study, we present measurements of the amount of ozone transported downwind of the Houston and Dallas/Fort Worth metropolitan areas and estimate its impact on regional background ozone levels. To quantify the export of ozone from Houston and Dallas, we use data collected with NOAA airborne ozone lidars during the TexAQS 2000 and TexAQS 2006 studies. The NOAA lidars provided highly resolved, continuous ozone profiles as well as measurements of local mixing height along the flight path, making it ideally suited for studying the structure and properties of pollution plumes emanating from urban areas or other point sources. We use lidar data from seven flights (three from 2000 and four from 2006) during which we mapped out the Houston and Dallas/Fort Worth ozone plumes by flying multiple downwind transects across the plumes. Figure 1 shows the flight patterns for the TexAQS 2000 flights (gray lines) and the TexAQS 2006 flights (black lines). Only the portions of the flights during which the plumes were mapped out are shown. We use the lidar data in combination with wind data provided by networks of wind profilers to compute horizontal fluxes of ozone, ozone production rates, and ozone enhancements in these plumes. The locations of the radar wind profilers are

shown in Figure 1 as filled triangles (gray for TexAQS 2000, black for TexAQS 2006). We compare the characteristics of the Houston and Dallas/Fort Worth plumes and discuss Houston plume properties under different meteorological conditions. The flux measurements are used to assess the impact of ozone exported from the Houston and Dallas/Fort Worth metropolitan areas on air quality downwind.

2. Instrumentation

[4] Ozone measurements in the Houston and Dallas/Fort Worth plumes were obtained with two different versions of the NOAA airborne ozone and aerosol lidar. The TexAQS 2000 ozone data were collected with an excimer laser-based airborne ozone lidar that used five fixed wavelengths in the UV spectral region to detect ozone [Alvarez *et al.*, 1998]. The TexAQS 2000 missions were flown on a Douglas DC-3 aircraft. For the TexAQS 2006 study, we used NOAA's recently developed tunable optical profiler for aerosol and ozone (TOPAZ) differential absorption lidar (DIAL) [Alvarez *et al.*, 2008]. TOPAZ incorporates the latest solid-state laser technology, and its transmitter is tunable in the UV spectral region. The lightweight and compact system was deployed on a NOAA Twin Otter research aircraft during the TexAQS

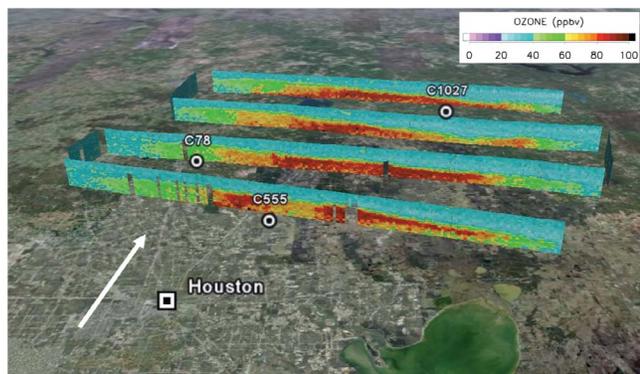


Figure 2. Airborne lidar ozone cross sections of the Houston plume measured 33–115 km downwind of Houston on 14 August 2006 overlaid on a Google Earth map. The ozone mixing ratio is color coded according to the scale shown in the top right corner. The white arrow indicates the southerly wind flow on that day. The locations of three CAMS surface monitoring sites (C78, C555, and C1027) along the flight path are marked on the map.

2006 study. Both lidars were flown in a nadir-looking configuration at altitudes of typically 3 km above sea level (ASL), yielding ozone profiles from ~ 300 m above the surface to about 2.5 km ASL. The vertical ozone profiles are calculated by first averaging the lidar signals vertically over 90 m and then applying the DIAL retrieval technique in a gliding fashion over five adjacent 90 m gates. The reported DIAL resolution is 90 m; however, completely independent data points are spaced 450 m apart. The integration time is 10 s, corresponding to a horizontal resolution of about 650 m, as both aircraft typically flew at speeds of 65 m s^{-1} . Both lidars also provided aerosol backscatter profiles at 15 m (excimer laser-based system) and 6 m (TOPAZ system) vertical resolution. The ozone and aerosol backscatter retrieval techniques for the two lidar systems are described by Alvarez *et al.* [1998, 2008].

[5] Knowledge of mixed layer depth is critical for the horizontal ozone flux calculation since it controls the vertical extent of the ozone plumes emanating from the Houston and Dallas urban areas. We could estimate mixed layer height from the ozone profile data, but their rather coarse vertical resolution and the inherent 450 m smoothing would introduce significant uncertainty in the mixed layer height determination. Instead, we use the highly resolved lidar backscatter profile data to retrieve mixed layer height by employing a Haar wavelet technique [Davis *et al.*, 2000]. This method relies on a strong contrast in aerosol backscatter between the convective mixed layer and the free troposphere and assigns the local mixed layer height to the altitude at which the gradient in aerosol backscatter is largest. When the aerosol contrast is sufficient, this approach provides mixed layer height measurements at high spatial resolution, capable of resolving individual convective eddies. Previous studies [Nielsen-Gammon *et al.*, 2008; Cohn and Angevine, 2000; White *et al.*, 1999] have demonstrated that mixing heights retrieved with the lidar backscatter method compare well with those computed from wind profiler backscatter or radiosonde temperature and humidity data.

[6] The horizontal wind measurements needed to compute horizontal fluxes of ozone were provided by networks of radar wind profilers that were deployed in eastern Texas during the TexAQS 2000 and 2006 campaigns. Boundary layer radar wind profilers typically provide profiles of wind speed and direction at 30 or 60 min intervals between about 100 m to a few kilometers above ground level (AGL) at vertical resolutions of 50–100 m [Carter *et al.*, 1995].

3. Analysis

[7] To estimate the amount of ozone emanating from Houston and Dallas/Fort Worth, we chose days when steady, moderate winds of several meters per second transported pollutants downwind of the two metropolitan areas, resulting in well-defined ozone plumes oriented parallel to the mean flow. We typically mapped these plumes by flying at least one upwind and multiple downwind transects across the plume with the airborne ozone lidar. An onboard real-time display of the lidar ozone profiles allowed us to adjust the length of the plume transects until we had crossed the entire width of the plume and were sampling background air before beginning the next cross-plume flight leg. In addition to computing the horizontal flux of ozone in the Houston and Dallas/Fort Worth plumes for each plume transect, we also determined the corresponding ozone production rates and peak excess ozone for the same transects.

3.1. Horizontal Ozone Flux

[8] To illustrate the retrieval technique, we show ozone profile data from the flight on 14 August 2006 (Figure 2). On that day, southerly winds of about 4 m s^{-1} transported the Houston/Ship Channel ozone plume to the north, where we flew four transects across the plume at distances of 33–115 km downwind of Houston. The ozone cross section for the second downwind transect from Figure 2 is shown in more detail in Figure 3. Also plotted is the mixed layer height (Figure 3, thick black line) determined from the lidar aerosol backscatter profiles. Ozone data in the lowest ~ 300 m above the surface are omitted because of rather large uncertainties and potential biases in the ozone retrieval caused by poor signal-to-noise ratio and possible signal contamination by the ground return. To compute the horizontal plume ozone flux, we extrapolate the ozone data from the lowest usable altitude to the ground and interpolate the data across short time gaps caused by clouds that prevented the downward-looking lidar from obtaining a full ozone profile down to the ground (e.g., first transect in Figure 2). These are reasonable approximations since the boundary layer was convectively well mixed to at least several hundred meters above ground in all the cases that we used to estimate ozone fluxes. To test our assumption of constant ozone mixing ratio profiles in the lower portion of the mixed layer, we compared the lidar ozone measurements at ~ 300 m AGL (the lowest consistently usable altitude) with ozone measurements at three surface sites, which were close to the flight track on 14 August 2006 (Figure 2). The three surface sites (C555, C78, and C1027) are part of the Texas surface ozone monitoring network of Continuous Ambient Monitoring Stations (CAMS). The lidar ozone measurements at ~ 300 m AGL were averaged within a 5 km radius of each surface station and compared to the 5 min average surface

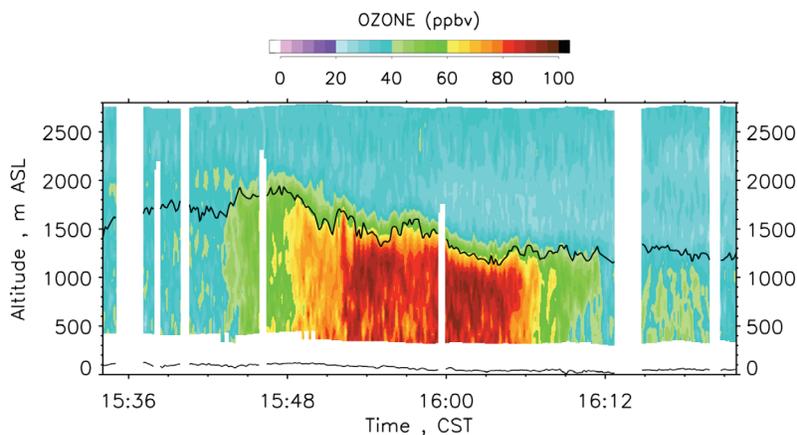


Figure 3. Airborne lidar ozone cross section of the second downwind transect of the Houston plume (see Figure 2) measured on 14 August 2006 about 58 km north of Houston. The thick black line indicates mixed layer height, and the black line near the bottom of the plot represents the ground altitude.

ozone data at the time of the flyby. Table 1 shows that the ozone mixing ratios measured with the lidar at ~ 300 m AGL are about 2–5 ppb or ~ 4 –8% higher than the ozone values measured at the surface. The slightly lower surface ozone values are probably due to ozone deposition or titration and are likely limited to the surface layer, which typically occupies the lowest 5–10% of the mixed layer (e.g., *Driedonks and Tennekes*, 1984). Therefore, the assumption of constant vertical ozone profiles below ~ 300 m AGL will only result in a very slight overestimation of ozone contained in the plume.

[9] The next step of the ozone flux retrieval consists of determining the background ozone level, which must be subtracted from the plume ozone values to quantify the amount of ozone added to the atmosphere by the Houston and Dallas/Fort Worth metropolitan areas. Figure 4 shows a time series plot of ozone for the same transect as in Figure 3. This ozone time series was generated by extrapolating the ozone profiles shown in Figure 3 to the ground and averaging the resulting profiles vertically from the surface to the top of the mixed layer. From this line plot of mean mixed-layer ozone, we determine plume width and local background levels. The plume edges are chosen as those locations where the ozone trace begins to level off on either side of the plume. We determine local background ozone by averaging several minutes (i.e., ~ 10 km) worth of data just beyond each plume edge. Background ozone levels may be different for the two sides (i.e., the background beneath the

plume may be sloped). Therefore, we interpolate across the plume between the background estimates on either side. We then subtract for each time step the interpolated background ozone from the corresponding plume ozone profile, which yields a 2-D time height cross section of excess plume ozone. Total excess ozone in the plume is determined by integrating the data from the surface to the top of the mixed layer and between the horizontal plume edges. Figure 3 shows that the mixing height (and thus the upper limit for the integration of excess plume ozone) varies significantly across the plume, from about 1200 m on the left (western) side of the plume to about 2000 m on the right (eastern) side of the plume. The higher mixing depths on the western side of the plume may be attributed to advection of the Houston urban heat island. This case illustrates the advantage of using a lidar for this type of study as it provides ozone profile and mixing height data simultaneously, which allows us to properly account for any mixing height inhomogeneities.

[10] The horizontal wind speeds needed for the flux calculation are derived from the hourly wind speed profiles measured by the network wind profilers. Wind speed data from all available profilers for the hour closest to the middle time of a given transect are averaged and interpolated to the center location of the transect using an inverse distance squared-weighted average [*White et al.*, 2006]. The resulting wind speed profile is then averaged vertically between the surface and the mean mixing height for the transect, yielding

Table 1. Comparison of TOPAZ Ozone Measurements at ~ 300 m AGL With Texas Commission on Environmental Quality (TCEQ) Surface Ozone Measurements on 14 August 2006^a

Station	TCEQ Surface Sites			TOPAZ Lidar at ~ 300 m AGL		
	Altitude (m ASL)	Time (LT)	O ₃ (ppbv)	Distance (km)	Time (LT)	O ₃ (ppbv)
Kingwood Library C555	12	152000	62.2	1.5	152233	66.9
Conroe Relocated C78	67	154500	54.6	2.2	154643	56.8
Alabama-Coushatta C1027	91	171500	80.5	2.7	171753	85.0

^aThe 300 m AGL data are used to extrapolate TOPAZ ozone profiles to the surface. TOPAZ data are an average of all data points that fell within a circle of 5 km radius around each surface station. The closest distance of TOPAZ to the surface site and the corresponding time are denoted. TCEQ surface ozone values are 5 min averages closest in time to the TOPAZ flyby. TCEQ time is the begin time of the 5 min average.

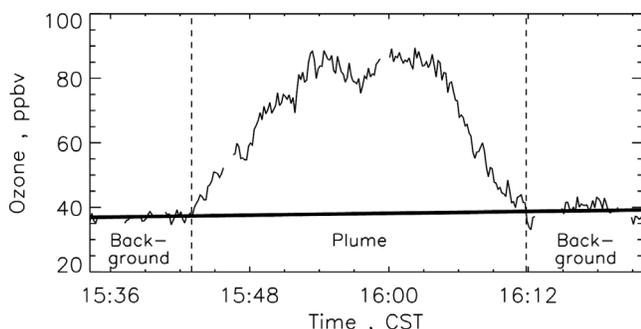


Figure 4. Time series plot of ozone lidar data averaged vertically from the surface to the top of the mixed layer for the same transect as in Figure 3. The thick horizontal line indicates the background ozone level.

an average horizontal wind speed within the mixed layer for each transect. The vertical averaging is performed to reduce errors in the wind speed measurements. By multiplying the horizontal wind speed and the integrated excess plume ozone, we compute the horizontal ozone flux in molecules per second for each transect. In the flux calculation, the angle between the flight path and the travel direction of the plume is accounted for by multiplying the flux by the sine of the angle between aircraft and plume headings. The aircraft heading is known from the onboard GPS data. Plume heading is determined from the center locations of adjacent plume transects. If transects are spaced closely together, this method of computing plume heading may introduce significant uncertainties. In these cases, plume heading for all transects is determined from the center locations of the first and last transects.

3.2. Ozone Enhancement and Production Rates

[11] To further characterize the Houston and Dallas/Fort Worth plumes, we also determine the ozone enhancement and ozone production/loss rate for each plume transect. To compute the ozone enhancement for a given transect, we use the plume ozone data vertically averaged between the surface and the top of the mixed layer (Figure 4, data points between the vertical dashed lines), from which we subtract the corresponding background values to yield vertically averaged excess plume ozone. We define plume ozone enhancement as the 95th percentile of these vertically averaged data points. To calculate the ozone production rate, we first take the difference in integrated excess plume ozone between adjacent transects and divide by the average cross-sectional plume area of the two transects. The cross-sectional area is calculated by summing the area of all range gates that lie between the surface and the boundary layer height and the lateral plume boundaries. The difference in integrated excess plume ozone is a measure of ozone production within the plume between the two transects. Dividing by the plume travel time between the transects yields the ozone production rate in ppbv h⁻¹. The plume travel time is simply derived from the distance between the center locations of adjacent transects and an estimate of the wind speed in the area between the transects. The latter was approximated by taking the mean of the interpolated and boundary layer averaged profiler wind speeds at the two transects. The

measured ozone production rates are time stamped with the middle time between the two transects. This method to compute ozone production rate from airborne lidar data is similar to that described by *Senff et al.* [1998]. The production rate that we measure can be viewed as a net ozone production rate since it represents the net total of photochemical ozone production and destruction and ozone losses caused by deposition or detrainment processes.

3.3. Error Estimates

[12] Errors in the ozone flux, production rate, and enhancement retrievals are influenced by a number of factors. The lidar ozone measurements at 90 m vertical and at 650 m horizontal resolution have a precision of about 3–10% and an accuracy of a few percent [*Alvarez et al.*, 1998, 2008]. For the retrievals described here, the ozone data were either integrated vertically and horizontally across the plume (ozone flux, production rate) or averaged vertically (ozone enhancement). Therefore, the statistical uncertainty of the retrievals is proportional to $1/\sqrt{N}$, with N being the number of ozone data points that are integrated or averaged together. The number of data points within the plume cross sections ranges from about 40 to 250 in the horizontal to about 10–30 in the vertical. Thus, the statistical uncertainties in the integrated ozone are less than 0.5% and less than about 1.5% for the vertically averaged ozone. The total errors of these quantities are dominated by the systematic errors of the ozone measurements (which are not reduced by integration or averaging procedures), and we estimate them to be on the order of several percent. This includes the slight bias introduced by extrapolating the measured ozone profile from ~300 m AGL to the ground (see section 3.1). The relative errors of the ozone production rate retrievals may be significantly larger in those cases when integrated plume ozone at two adjacent transects is of similar magnitude and the difference of these similar values is taken to compute ozone production rate. Errors in the determination of plume width and ozone background also contribute to uncertainties in the ozone flux, production rate, and enhancement retrievals. Since measurements of other chemical plume tracers such as CO or NO_y were not available for most of the flights, we use the ozone measurements to determine horizontal plume dimensions. This approach will yield unbiased results in most cases, except when the sampled plume is a conglomerate of the plume to be sampled and another pollution plume that originated from a source outside of the Houston or Dallas urban areas. If our data indicate that another plume may be interfering, we use wind profiler-based trajectory analysis [*White et al.*, 2006] or, if available, collocated in situ chemistry data from another research aircraft to delineate the plume pieces and adjust the main plume's width and background sampling regions accordingly. Biases introduced in these “conglomerate plume” cases are difficult to estimate and depend on the particular circumstances. The largest uncertainties in the flux and production rate retrievals are introduced by the wind measurements. The precision of the wind speed measurements derived from the profiler networks is on the order of a few percent (after the wind speed data have been averaged vertically within the boundary layer at each transect). However, the fact that the wind profiler measurements may not be representative for the location of a particular plume transect

can introduce much larger errors. This is especially of concern when the Houston or Dallas/Fort Worth plumes have been transported to areas with no wind profilers in the vicinity. We estimate these wind speed sampling errors to be as large as 30% in some cases. Taking into account all error sources, we estimate that total errors for the ozone enhancement measurements are typically about 10% and for the horizontal flux and production rate retrievals on the order of 25%.

4. Results

[13] We performed horizontal ozone flux, plume ozone enhancement, and ozone production rate retrievals for a total of seven flights, when steady winds at moderate speeds resulted in well-defined pollution plumes downwind of the Houston and Dallas/Fort Worth urban areas. Three flights are from the TexAQS 2000 campaign (28 August, 1 September, and 6 September 2000), and four are from the TexAQS 2006 study (12 August, 14 August, 30 August, and 13 September 2006). All but one flight (13 September 2006) were flown downwind of Houston. Figure 5 shows plan view plots of ozone concentration measured with the lidar and averaged from the surface to the top of the mixed layer for each of the seven flights. Ozone is plotted along the flight tracks and overlaid over a map of the area. The plume measurements were mostly made in the afternoons with sampling times falling between 1120 and 1744 LT. The time spent characterizing the Houston or Dallas/Fort Worth plumes ranged from about 1.5 to 4.5 h (nine transects on 6 September 2000) but was typically ~2 h. Only the plume sampling portions of the flights are shown in Figure 5. In all cases, the pollution plume can be easily identified by elevated ozone levels downwind of the urban areas. Mean wind speeds for the duration of the plume sampling period ranged from 2.6 to 6.5 m s⁻¹, coming from southerly, westerly, northerly, or northeasterly directions. Plume sampling was not performed in a strictly Lagrangian sense since the plume travel time between the first and last downwind transects was usually larger than the sampling duration. All of the days shown had strong convective potential with negative values of the Lifted Index in the afternoon sounding from the nearby National Weather Service station in Lake Charles, Louisiana. Maximum daytime temperatures at Houston or Dallas/Fort Worth ranged from 30°C to 42°C. On all seven flights, the weather was clear to partly cloudy. All flights occurred on non-holiday weekdays, except the 12 August 2006 flight, which fell on a Saturday. Table 2 summarizes logistical and weather-related information for all flights.

4.1. Plume Characteristics

[14] Figure 6 depicts horizontal ozone flux downwind of Houston and Dallas/Fort Worth in molecules per second as a function of plume age (i.e., time of emission until time of arrival at the location of the particular aircraft transect) for all transects and all seven flights. Plume age for a given transect was computed by adding the plume travel times (see section 3.2) between transect pairs. Plume travel time to the first downwind transect was determined using the distance from an assumed plume emission point to the center location of the first transect and the wind speed interpolated from the

wind profiler measurements. As the origin of the emissions, we used a location in the middle of the Ship Channel area for the Houston cases and a location between Dallas and Fort Worth for the Dallas flight. Since the Houston/Ship Channel and Dallas/Fort Worth pollution plumes originate from rather large areas instead of individual points, the derived plume age is to be viewed as an approximation of the mean plume age. The Houston data show that ozone fluxes increased rather quickly during the first 3 h after emission and, for most cases, stayed rather constant after that. Similarly, plume ozone enhancements increased initially and then leveled off or decreased slightly as the chemically aged plume dispersed (Figure 7). These observations are supported by the measured ozone production rates shown for all transects and flights in Figure 8, and they are consistent with the finding that ozone is rapidly produced in the Houston plume shortly after emission [Ryerson *et al.*, 2003]. For plume ages of ≤ 3 h, the production rates were all positive and ranged from about 3–13 ppbv h⁻¹. Transects very close to the strong sources in the Ship Channel area were either not flown or not used for this analysis. Therefore, we did not find the very high instantaneous ozone production rates of ≥ 80 ppbv h⁻¹ that others have reported very close to the industrial sources in the Ship Channel area [Daum *et al.*, 2004; Kleinman *et al.*, 2002]. For plume ages of 3 h or longer, the measured production rates dropped significantly and ranged from about 5 to -4 ppbv h⁻¹. With ozone production rates tending to zero for plume ages greater than 3 h, the corresponding horizontal fluxes should level off and stay rather constant as the plumes travel downwind of the source areas. This was the case for all flights except for the one on 6 September 2000. The slight increase in flux for the last transect on 14 August 2006 is within the stated uncertainties (see section 3.3).

[15] The plume sampled on 6 September 2000 exhibited a different behavior. After leveling off at around 3 h plume age, the flux increased again at about 5 h plume age and beyond. The ozone production rate dropped to below zero at 3.5 h and jumped back up to about 4 ppbv h⁻¹ at 4.5 h plume age. On that day, the Houston/Ship Channel plume passed over the W. A. Parish power plant (Figure 5c). The fourth transect (4 h plume age) was the first transect downwind of the power plant. In 2000, the W. A. Parish plant was one of the largest NO_x point sources in the United States emitting 33,000 tons of NO_x per year. The additional NO_x injected into the Houston/Ship Channel plume likely caused titration of ozone in the near field and then additional ozone production farther downwind. This may explain the negative ozone production rate measured between the third and fourth transect (just downwind of the Parish plant) and at least some of the increased production rate and flux for the transects farther downwind. Also, measured wind speeds were higher by about 20% for transects 5 and 6, increasing the measured flux there. Since the nearest profiler was about 80 km away, the extrapolated profiler winds may not be representative and may have been overestimated.

[16] The bar graph in Figure 9 shows the average horizontal ozone flux for plume ages of ≥ 3 h (after the bulk of ozone production had ceased) for each of the seven flights. The fluxes range in magnitude from 0.9 to 4.7 · 10²⁶ molecules of ozone per second. The mean ozone flux for all six Houston cases is about 3.2 · 10²⁶ molec s⁻¹ compared to

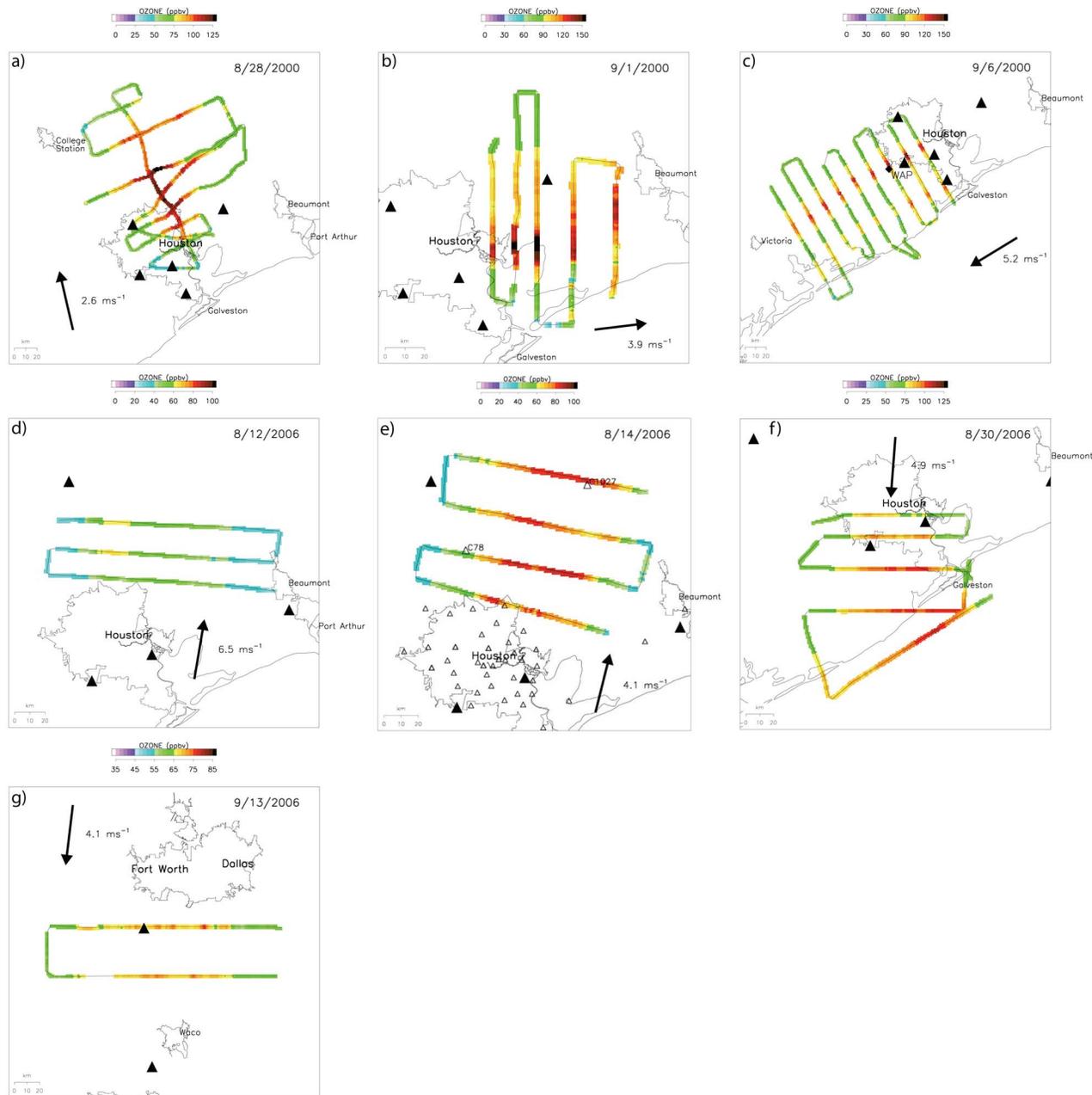


Figure 5. Ozone averaged from the surface to the top of the mixed layer plotted along the flight track and overlaid over a map of the surrounding area for (a) 28 August 2000, (b) 1 September 2000, (c) 6 September 2000, (d) 12 August 2006, (e) 14 August 2006, (f) 30 August 2006, and (g) 13 September 2006. Only those portions of the flights are shown when the Houston or Dallas/Fort Worth plumes were sampled. The ozone mixing ratio is color coded according to the scale shown on each plot (note that color scales vary). For each flight, the mean wind direction (arrow) and mean wind speed are indicated. Filled triangles represent the locations of radar wind profilers. The filled diamond in Figure 5c marks the W. A. Parish power plant (WAP), and the open triangles in Figure 5e show the locations of the Houston area CAMS stations.

$0.9 \cdot 10^{26}$ molec s⁻¹ for the Dallas plume case (13 September 2006). Maximum plume ozone enhancements are 13 ppbv for the Dallas/Fort Worth flight and range from 34 to 77 ppbv for the Houston/Ship Channel plume (Figure 7). Our data indicate that the horizontal ozone flux and ozone enhancements associated with the Houston pollution plume

were significantly larger than those downwind of the Dallas/Fort Worth area. However, our observations from the single Dallas plume flight may not have been representative for typical summertime ozone pollution conditions in the Dallas area. The Dallas flight occurred rather late in the summer, and the maximum temperature on that day was a relatively

Table 2. Logistical and Meteorological Data for all Seven Flights^a

Flight Date	Day of the Week	Plume Sampling Time (LT)	Urban/Industrial Plume Sampled	Mean Wind Speed (m s ⁻¹)	Mean Wind Direction	Maximum Temperature (°C)
28 Aug 2000	Mon	1256–1722	Houston/Ship Channel	2.6	S	37
1 Sep 2000	Fri	1300–1501	Houston/Ship Channel	3.9	W	42
6 Sep 2000	Wed	1120–1551	Houston/Ship Channel	5.2	NE	36
12 Aug 2006	Sat	1538–1736	Houston/Ship Channel	6.5	S	33
14 Aug 2006	Mon	1506–1727	Houston/Ship Channel	4.1	S	35
30 Aug 2006	Wed	1431–1744	Houston/Ship Channel	4.9	N	34
13 Sep 2006	Wed	1445–1634	Dallas/Fort Worth	4.1	N	30

^aMaximum temperature is taken from the National Weather Service stations at Houston Intercontinental Airport and Dallas/Fort Worth (on 13 September 2006).

cool 30°C (Table 2), which may have contributed to lower ozone mixing ratios and fluxes downwind of Dallas/Fort Worth. Other measurements [e.g., Luria *et al.*, 2008] suggest that the Dallas/Fort Worth area may produce stronger ozone fluxes earlier in the ozone season. However, higher ozone fluxes and greater plume ozone enhancements downwind of the Houston area are entirely consistent with the fact that the mix of urban and petrochemical pollution sources in the Houston area leads to more rapid ozone formation and, subsequently, more frequent and more severe violations of the ozone standard than in the Dallas/Fort Worth area [McKeen *et al.*, 2009; Neuman *et al.*, 2009; Daum *et al.*, 2004; Ryerson *et al.*, 2003; Kleinman *et al.*, 2002].

[17] Figures 6–8 show differences in the Houston plume properties between the 2000 and 2006 measurements. Our data, which are based on a rather small sample size of three cases from each study, indicate that ozone fluxes were significantly lower for two of three 2000 cases (1 September 2000 and 6 September 2000) compared to the 2006 observations. Ozone enhancements and production rates for plume ages up to 3 h were consistently higher in 2000. The observed differences in plume properties could be due to a number of factors, but the most likely causes may be

changes in source emissions and differences in meteorological conditions between the 2 years. Sullivan [2009] reports that NO_x and VOC concentrations in the Houston area declined between 2000 and 2006 by roughly 20–30% based on data from the CAMS surface monitoring network. These trends were corrected for interannual differences in meteorological conditions and probably indicate a reduction in source emissions. Model and experimental studies [Ryerson *et al.*, 2001, 2003; Sillman and He, 2002; Sillman, 2000; Liu *et al.*, 1987] have shown that ozone formation rates and yields (molecules of ozone produced per molecule NO_x emitted) depend in a complex, nonlinear way on NO_x and VOC concentrations and the ratio of these two species. Lower VOC concentrations, in particular reductions in highly reactive alkene concentrations, and lower NO_x concentrations are consistent with the lower ozone enhancements and ozone production rates that were observed in 2006 in the Houston/Ship Channel plume during the first few hours after emission. However, differences in these plume properties between the 2 years may also be due to changes in meteorological conditions. Ozone production rates are affected by ambient temperature, and ozone enhancement is strongly influenced by plume dispersion, which depends on the height of the mixed layer, the strength

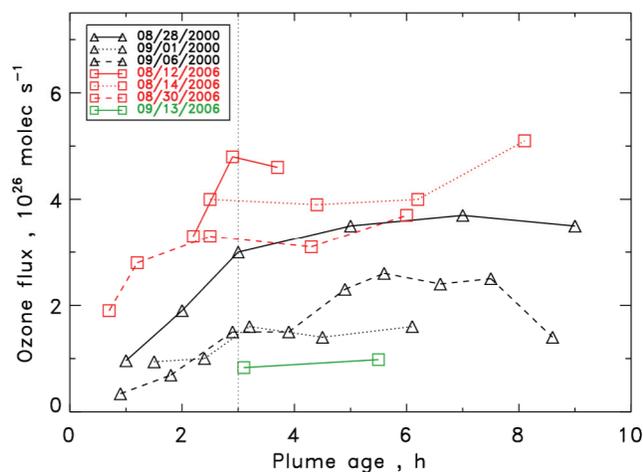


Figure 6. Horizontal ozone flux as a function of plume age for all plume sampling flights (Houston 2000 data are shown in black, Houston 2006 data are shown in red, and Dallas/Fort Worth 13 September 2006 data are shown in green). Each symbol represents the flux at a particular flight transect.

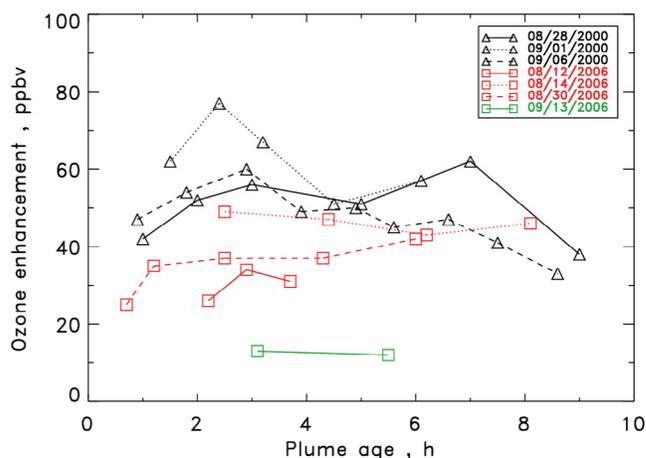


Figure 7. Plume ozone enhancement as a function of plume age for all plume sampling flights (Houston 2000 data are shown in black, Houston 2006 data are shown in red, and Dallas/Fort Worth 13 September 2006 data are shown in green). Each symbol represents the ozone enhancement at a particular flight transect.

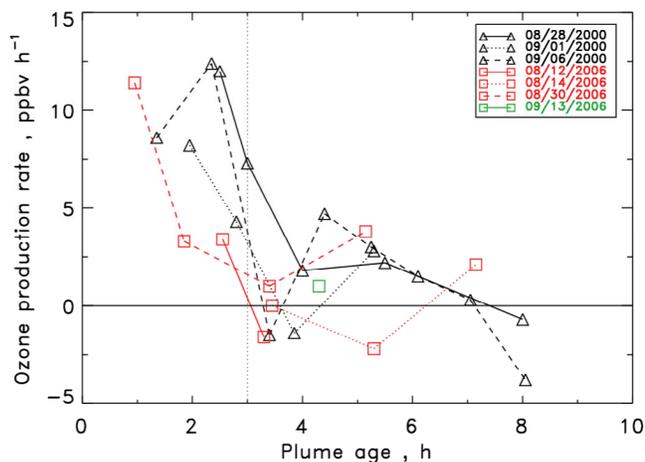


Figure 8. Ozone production rate as a function of plume age for all plume sampling flights (Houston 2000 data are shown in black, Houston 2006 data are shown in red, and Dallas/Fort Worth 13 September 2006 data are shown in green). Each symbol represents the production rate at a particular flight transect.

of the mixing, and the horizontal wind speed. Surface observations show slightly higher ambient temperatures during the 2000 flights (Table 2), mean wind speeds were lower for the 2000 cases (3.5 m s^{-1} , on average) than for the 2006 cases (5.2 m s^{-1} , on average) (see Table 2), and mixing heights were slightly higher for the 2000 flights compared to the 2006 flights. In Figure 10, the highest measured ozone enhancement for each of the six Houston flights is plotted versus cross-sectional plume area multiplied by the horizontal wind speed. This quantity is a proxy for the dilution of the plume in all three dimensions. As one might expect, Figure 10 shows a strong inverse relationship between the maximum plume ozone enhance-

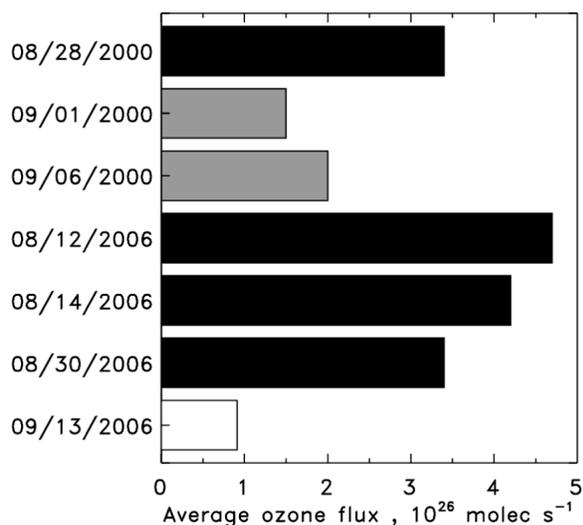


Figure 9. Mean horizontal ozone flux for plume ages greater than 3 h for each flight (Dallas/Fort Worth plume on 13 September 2006, Houston plume on all other days).

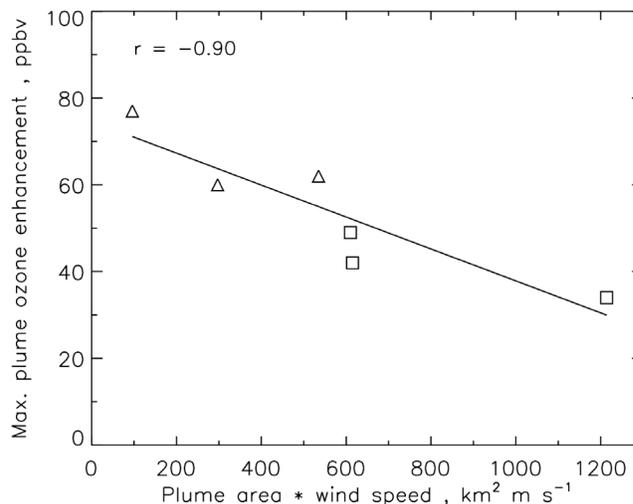


Figure 10. Maximum plume ozone enhancement versus the product of plume cross-sectional area and horizontal wind speed. Plume area and wind speed are for the transect where the maximum ozone enhancement occurred. Triangles denote results from the TexAQS 2000 flights, and squares denote results from the TexAQS 2006 flights.

ment and the product of plume size and horizontal wind speed. Despite the likely change in emissions between 2000 and 2006, we have included both sets of data in the regression analysis to maintain an acceptable sample size. Figure 10 clearly shows that the inverse relationship between ozone enhancement and the product of plume size and horizontal wind speed holds true for data from the individual years as well. The higher ozone enhancements observed for the 2000 cases (triangles) compared to the 2006 flights (squares) are likely caused by a combination of higher emissions (as discussed above) and less dilution because of lower horizontal wind speeds and smaller plume sizes (see Figures 5b and 5c).

[18] The fact that the ozone fluxes on 1 September 2000 and 6 September 2000 were lower than the fluxes observed in 2006 despite larger ozone enhancements seems at first counterintuitive. The differences in wind speed and plume dimensions between the 2 years should not affect plume flux since it takes into account the dilution effects of horizontal wind speed and plume dispersion. In the following, we examine whether differences in wind direction (Table 2) may explain the changes in observed fluxes. Figure 9 shows that on days with southerly or northerly winds (28 August 2000, 12 August 2006, 14 August 2006, 30 August 2006; black bars), ozone fluxes are significantly higher (average of $3.9 \cdot 10^{26} \text{ molec s}^{-1}$) compared to days when the wind was blowing from westerly or northeasterly directions (1 September 2000 and 6 September 2000; gray bars), when ozone fluxes averaged only $1.7 \cdot 10^{26} \text{ molec s}^{-1}$. The major pollution sources along the Ship Channel form a west-to-east line that stretches over approximately 50 km from downtown Houston in the west to Baytown in the east (Figure 1, inset). When the wind is coming from southerly or northerly directions, the air crosses this line of pollution sources roughly at a 90° angle. Downwind of each individual source, ozone is produced rapidly, and as the individual plumes disperse laterally, they

merge into a single, wide plume. For example, in the first downwind transect on 14 August 2006 (Figures 2 and 5e), one can distinguish the Houston urban plume near the western edge of the broad plume and the Ship Channel plume in the middle and near the eastern edge. Some structure within the Ship Channel plume is also visible. Farther downwind, the different plume segments have all blended together. On the other hand, when winds are coming from westerly or easterly directions, the air is streaming roughly parallel to the Houston/Ship Channel pollution sources and is picking up NO_x and VOC precursors from these sources one after the other. This results in a much narrower pollution plume (e.g., 6 September 2000 in Figure 5c) with likely higher ozone precursor concentrations compared to the southerly or northerly wind cases. Previous studies [Ryerson *et al.*, 2001, 2003; Sillman and He, 2002; Sillman, 2000; Liu *et al.*, 1987] have shown that plumes with high ozone precursor concentrations have lower ozone yields compared to plumes with lower precursor concentrations and that a single concentrated pollution plume tends to produce less ozone than several separate, more dilute plumes combined. This may explain why the narrow, conglomerate Ship Channel plume, which forms under easterly or westerly flow conditions, produces less total ozone and thus a smaller flux than the sum of all plumes originating from individual petrochemical facilities under southerly or northerly winds. We lack data on NO_x and VOC concentrations in the plumes and thus cannot prove or disprove our hypothesis, but it provides a plausible explanation for the observed dependence of Houston/Ship Channel ozone fluxes on wind direction and, in turn, the lower fluxes measured on 1 September 2000 and 6 September 2000.

[19] Given this apparent dependence of ozone flux on wind direction, only the results from 28 August 2000, a southerly flow case, can reasonably be compared with the 2006 fluxes, which were all measured under northerly or southerly flow conditions (we did not compute fluxes under easterly or westerly flow in 2006 that could be compared to the east-west flow cases from 2000). The 28 August 2000 ozone flux is very similar to the fluxes measured in 2006. Based on this very limited comparison, we find no evidence for a significant change in the total amount of ozone produced by the Houston area between 2000 and 2006. We also did not observe any significant differences in the horizontal flux on a Saturday (12 August 2006) compared to weekdays under similar flow directions. This was to be expected since the major pollution sources in the Houston area, the petrochemical plants, are operated continuously regardless of day of week. Any changes in traffic emissions appear to have had no discernable effect on the horizontal ozone flux. Given the small sample size, our findings regarding flux comparisons between the 2 years and between weekdays and weekend days are to be seen more as circumstantial evidence rather than statistically robust results.

4.2. Implications for Air Quality Downwind of Houston

[20] The ozone plume emanating from the Houston area clearly has a detrimental impact on air quality in communities downwind of Houston. For example, our measurements from 14 August 2006 (Figures 2–4 and 5e) show that peak ozone concentrations in the Houston plume up to about 100 km downwind of Houston approached 90 ppbv

although the Houston area was well ventilated and background ozone concentrations were rather low (about 40 ppbv in this case). Since the wind direction did not change much over the course of the day, it is conceivable that locations in the path of the Houston ozone plume may have experienced an exceedance of the 8 h ozone standard [EPA, 2008], which was 84 ppbv at the time. With the 8 h ozone standard now lowered to 75 ppbv, an ozone exceedance downwind of Houston under the scenario observed on 14 August 2006 would be likely. The CAMS surface ozone monitoring network in southeast Texas is concentrated within the Houston metropolitan area (Figure 5e). Only two CAMS stations, Conroe (C78) and Alabama-Coushatta (C1027), were located to the north of the Houston metropolitan area in or near the path of the Houston pollution plume on 14 August 2006. The locations of these two CAMS stations are shown in Figure 5e. The plume grazed the Conroe site, so the CAMS measurements from there are only representative of ozone concentrations at the edge of the plume. The Alabama-Coushatta site, which reported a 1 h ozone maximum of 79 ppbv, was closer to the plume core but still missed the highest ozone values in the Houston plume measured with the airborne lidar. This site was deactivated in October 2008. As a consequence, under conditions similar to the ones observed on 14 August 2006, the Houston ozone plume would now go largely undetected by the surface ozone monitoring network. This illustrates that under steady flow conditions, when the Houston area is well ventilated, the CAMS network in the greater Houston area will likely report rather low ozone values while higher ozone values and possible violations of the ozone standard may occur downwind. In cases with moderate to high background ozone levels of ≥ 60 ppbv (e.g., 6 September 2000 or 30 August 2006) (Figures 5c and 5f), ozone exceedances in the plume path downwind of Houston are a virtual certainty, and again, it is very likely that these exceedances would not be registered by the CAMS network because of the sparsity of stations outside of the Houston metropolitan area.

[21] During the six Houston flights, we mapped out the Houston/Ship Channel plume to downwind distances of up to 160 km or about 9 h of plume age. In all cases, the plume was still well defined and easily identifiable even at the farthest transects. As the Houston plume travels farther downwind, transport and mixing processes are likely to disperse it over a larger area. Convective venting will help disperse the plume during the day [Langford *et al.*, 2010], and the low-level jet, a regularly occurring phenomenon in southeast Texas, tends to spread the plume very effectively at night [Tucker *et al.*, 2010; Banta *et al.*, 2003, 2005, 2006]. The next morning, the remnants of the Houston plume are mixed vertically as the new turbulent boundary layer develops. Eventually, the Houston plume becomes part of the background and increases regional background ozone levels [Banta *et al.*, 1998, 2005].

[22] To quantify the contribution of the Houston ozone plume to regional background ozone levels, we use the mean measured horizontal ozone flux of $3.2 \cdot 10^{26}$ molec s⁻¹. We assume that this rate of transport is sustained over a 12 h period (during which photochemical ozone production occurs) and that the plume is released into a 1.5 km deep mixed layer. Mixing heights vary with time of day and

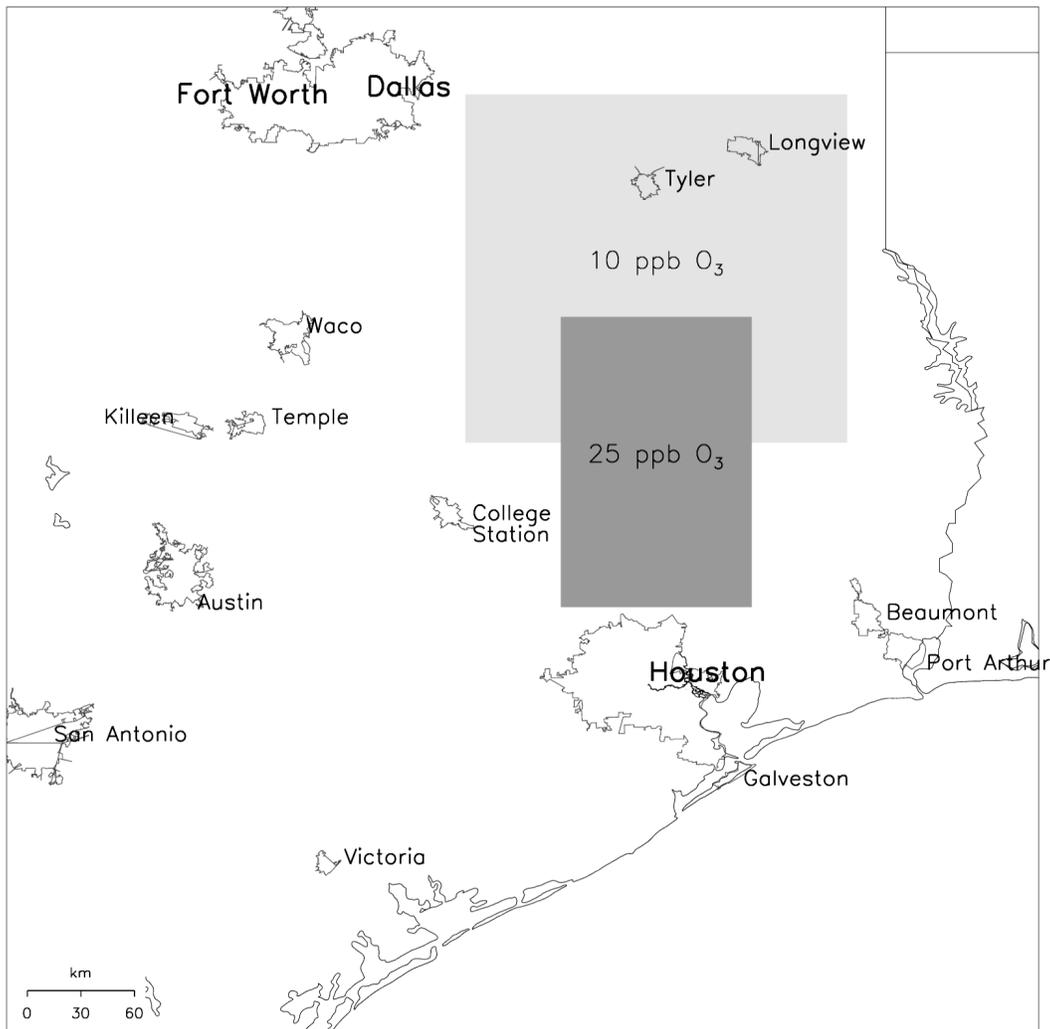


Figure 11. The gray rectangle and square illustrate the area and average amount of ozone by which regional background ozone would be augmented if the excess plume ozone transported away from the Houston area at a rate of $3.2 \cdot 10^{26}$ molec s^{-1} dispersed uniformly over these two areas. For comparison, the two shaded areas are overlaid on a map of east Texas.

spatially across southeast Texas, with generally lower heights near the Gulf of Mexico and Galveston Bay and greater mixed layer depths farther inland [Nielsen-Gammon *et al.*, 2008]. Mixed layer height measurements in the greater Houston area described by Nielsen-Gammon *et al.* [2008] indicate that 1.5 km is a reasonable value to use for a mean regional-scale daytime mixed layer height. Using the above values for horizontal ozone flux, duration of photochemical ozone production, and mixing height, we compute the ozone enhancement due to the Houston plume per unit area. By estimating the area occupied by the plume at various plume ages and assuming uniform dispersion, we calculate the amount of ozone added to the background by the Houston plume. Figure 11 shows the mean ozone enhancement for two assumed areas covered by the plume. The dark gray rectangle indicates the area the Houston plume might occupy in the evening of the day it was emitted after a full day of transport at wind speeds of several meters per second. The plume covers about 16,000 km², resulting in a mean ozone enhancement of approximately 25 ppbv. This value is

lower than the ozone enhancements shown in Figure 7 because the latter represent peak rather than mean values. As the Houston plume ages and disperses further, the areal coverage can vary widely depending on the particular atmospheric conditions it encounters. Assuming an increase in plume area to 40,000 km² or a factor of about 2.5 over the area covered by the plume at the end of the day when it was emitted, the average ozone enhancement over the regional background would be about 10 ppbv. A plume area of this magnitude is not unreasonable for a day-old plume that has undergone nighttime transport and next-day turbulent mixing. The light gray square in Figure 11 indicates the size of the area in relationship to east Texas. Under this scenario, the Houston plume would increase background ozone levels by approximately 10 ppbv over an area half the size of northeast Texas. Obviously, for areas smaller or larger than this, the ozone background enhancement would vary proportionally to the inverse of the area. In reality, the aged Houston plume would not be spread uniformly across a given area but rather exhibit some degree of inhomogeneity.

Therefore, the estimates for ozone background enhancement are to be viewed as an area-wide average. In computing the ozone background enhancement, we neglect ozone loss processes, such as titration caused by the plume passing over fresh NO_x sources or detrainment of ozone into the free troposphere. However, ozone losses due to deposition are accounted for since the ozone flux data inherently include losses of ozone caused by surface deposition.

[23] The estimated ozone background enhancements resulting from ozone transport out of the Houston area have important implications for air quality in communities downwind of the Houston metropolitan area. *Nielsen-Gammon et al.* [2005] reported that background ozone concentrations in east Texas typically peak in late summer. Average late-summer background concentrations on days without precipitation are above 50 ppbv for the Houston-Galveston-Brazoria area and approach 60 ppbv for the Dallas/Fort Worth area. Background ozone concentrations for individual days can be significantly higher than that, approaching the 8 h ozone standard of 75 ppbv [Langford et al., 2009; Hardesty et al., 2008; Nielsen-Gammon et al., 2005]. Therefore, the additional 10–25 ppbv that may be added to the regional ozone background by the Houston plume could increase the background close to or above the 8 h ozone standard. As a consequence, rural areas without any local pollution sources or small urban areas in eastern Texas that produce some ozone locally, but not enough to exceed the 8 h standard, may be pushed into noncompliance with the National Ambient Air Quality Standard [EPA, 2008].

5. Summary and Conclusions

[24] We computed horizontal ozone fluxes, ozone production rates, and ozone enhancements for the Houston/Ship Channel and Dallas/Fort Worth pollution plumes using airborne ozone lidar and radar wind profiler data that were collected during the TexAQS 2000 and TexAQS 2006 air-quality studies. Based on six case studies of the Houston plume and one flight during which we investigated the Dallas plume, the horizontal ozone flux produced by the Houston metropolitan area was significantly higher than the ozone flux emanating from the Dallas/Fort Worth area. However, our observations from the single Dallas plume flight may have underestimated typical summertime ozone fluxes downwind of Dallas/Fort Worth, since the flight occurred toward the end of the ozone season. The Houston fluxes exhibited a strong correlation with wind direction: on southerly or northerly flow days, horizontal ozone fluxes were approximately twice as high as on days with winds coming from the west or east. A plausible explanation for this finding is that westerly or easterly flow results in rather narrow plumes with higher ozone precursor concentrations and, consequently, lower ozone yields, because the wind flow is parallel to the Ship Channel, where the majority of large pollution sources in the Houston area are located. On the other hand, southerly or northerly flow is perpendicular to the Ship Channel causing wide plumes with likely lower precursor concentrations and higher ozone yields. Despite the lower total amount of ozone produced for Ship Channel parallel flow, we found that plume ozone enhancement was often higher than on southerly or northerly flow days because the plumes were much more concentrated.

[25] The amount of ozone exported by the Houston area clearly has a detrimental impact on air quality in communities downwind of Houston. We have found that ozone concentrations in the Houston plume can exceed the 8 h ozone standard up to more than 100 km downwind of the metropolitan area, even on days when background ozone levels are low and the region is well ventilated by a steady synoptic flow. As the Houston plume is carried farther downwind, it disperses and increases regional ozone background levels. We estimate that ozone transported out of Houston during the course of a summer day may raise background ozone levels over a 40,000 km² area by as much as 10 ppbv. This is a significant increase in regional background ozone over a large area, which may lead to violations of the 8 h ozone standard far downwind of Houston in regions that have no or only modest pollution sources of their own.

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