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# Alaskan and Canadian forest fires exacerbate ozone pollution over Houston, Texas, on 19 and 20 July 2004

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[1] On Monday, 19 July, and Tuesday, 20 July 2004, the air over Houston, Texas, appeared abnormally hazy. Transport model results and data from the Atmospheric Infrared Sounder (AIRS), the Moderate Resolution Imaging Spectrometer (MODIS), the Measurement of Ozone by Airbus In-service airCraft (MOZAIC) experiment, and the Total Ozone Mapping Spectrometer (TOMS) indicate that an air mass originating on 12 July 2004 over forest fires in eastern Alaska and western Canada arrived in Houston about 1 week later. Ozonesonde data from Houston on 19 and 20 July show elevated ozone at the surface (>125 ppbv) and even higher concentrations aloft ( $\sim$ 150 ppbv of ozone found 2 km above the surface) as compared to more typical profiles. Integrated ozone columns from the surface to 5 km increased from 17-22 DU (measured in the absence of the polluted air mass) to 34-36 DU on 19 and 20 July. The average on 20 July 2004 of the 8-hour maximum ozone values recorded by surface monitors across the Houston area was the highest of any July day during the 2001-2005 period. The combination of the ozone observations, satellite data, and model results implicates the biomass burning effluence originating in Alaska and Canada a week earlier in exacerbating pollution levels seen in Houston.

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#### 1. Introduction

[2] When combined with emissions of nitrogen oxides  $(NO_x)$  in the presence of sunlight, emissions of hydrocarbons (HC), including volatile organic compounds (VOCs), react to form ozone  $(O_3)$ . As an epicenter of petrochemical production plants and home to the fourth

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largest population in the United States (U.S.), Houston, Texas, has concentrated emissions of both HC and  $NO_x$ . Nearly colocated sources of both are found around the Houston Ship Channel and Galveston Bay, resulting in significant and rapid ozone production [*Daum et al.*, 2004].

[3] Houston's location near the Gulf of Mexico and Galveston Bay and the related summertime meteorology also play key roles in the development of the very high levels of ozone often observed in Houston [*Banta et al.*, 2005]. As a result, Houston provides an environment unique in the U.S. in the character and sources of its ozone pollution.

[4] Once again in 2004, Houston led the U.S. in the number of days on which the National Ambient Air Quality Standard (NAAQS) for ozone established by the Environmental Protection Agency (EPA) was violated. Surface monitors in the greater Houston area reported violations of the 1-hour ozone standard [*Environmental Protection Agency*, 1999] of 125 ppbv on 35 days and violations of the 8-hour standard of 85 ppbv on 45 days in 2004 (Texas Commission on Environmental Quality (TCEQ): http://www.tceq.state.tx.us). Numerous studies have demonstrated the negative impacts on human health caused by both acute high-level exposures to ozone and prolonged exposures at lower levels [e.g., *Lippmann*, 1991; *McConnell et al.*, 2002; *Bell et al.*, 2004].

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**Figure 1.** MODIS (Terra) data show a smoke cloud over Houston on 19 July 2004 when ozone levels in Houston were notably higher from the surface to 4 km altitude. Image is courtesy of MODIS Rapid Response Project at NASA GSFC.

[5] Observations from the Williams Tower on the west side of Houston during the Texas Air Quality Study (TexAQS) 2000 demonstrated that the highest levels of ozone are often found above the surface, and therefore remain undetected by the surface monitoring network [*Berkowitz et al.*, 2004]. In addition, imported pollution tends to appear above the boundary layer [e.g., *Forster et al.*, 2001; *Wotawa and Trainer*, 2000], with downward mixing bringing polluted air parcels to the surface.

[6] Despite the compelling nature of the problem, Houston's ozone pollution above the surface was not monitored before July 2004 [*Banta et al.*, 2005]. Accordingly, we initiated a sounding program to examine ozone profiles throughout the troposphere and participated in the IONS project (Intercontinental Transport Experiment (INTEX) Ozonesonde Network Study) (A. M. Thompson et al., IONS-04 (INTEX Ozonesonde Network Study, 2004): Perspective on summertime UT/LS (upper troposphere/ lower stratosphere) ozone over Northeaster North America), submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Thompson et al., submitted manuscript, 2006). Nine IONS stations operated from 1 July to 15 August 2004 along a south-central U.S. to Maritime Canada axis with Houston being the most southerly of these.

[7] Two unanticipated features during July and August 2004 contributed to an unusual ozone record over the U.S.

and maritime Canada. First, the normal "Bermuda highpressure" system that typically fosters serious ozone pollution events along the east coast of the U.S. did not materialize because of a persistent low-pressure system centered over Ontario and the Great Lakes region. This meteorological setup suppressed lower tropospheric ozone levels at most IONS sites (Thompson et al., submitted manuscript, 2006). Second, outbreaks of forest fires in Alaska and California added to the regional pollution burden [*Pfister et al.*, 2005].

[8] Links between biomass burning and pollution observations downwind are well established [e.g., *Wotawa and Trainer*, 2000; *Forster et al.*, 2001; *Rogers and Bowman*, 2001; *Thompson et al.*, 2001; *Lamarque et al.*, 2003; *Colarco et al.*, 2004]. The elevated levels of ozone,  $NO_x$ , and a wide range of hydrocarbons found in smoke plumes from biomass burning can lead to significant further ozone production downwind of the fires [*Goode et al.*, 2000].

[9] In this paper, we examine the impact of one smoke transport event on ozone levels over Houston, Texas. The combination of rich local sources of  $NO_x$  and VOCs in Houston along with the transport of additional  $NO_x$ , VOCs, and ozone from remote forest fires in Alaska and western Canada led to the ozone pollution event of 19 and 20 July that was characterized by 50-110% increases in ozone in the first 5 km over Houston. Below we present supporting evidence from in situ observations, satellite data, and transport models linking the enhanced pollution above Houston to the Alaskan and Canadian fires.

#### 2. Observations and Model Results

## 2.1. Surface Observations on 19 and 20 July

[10] The skies over Houston were noticeably hazier than normal on 19 and 20 July 2004. Figure 1 shows smoke over southeast Texas and Louisiana on 19 July as derived from



**Figure 2.** Microtops II observations of aerosol optical thickness were recorded from the Rice University ozone-sonde launch site by two different instruments at two different wavelengths (340 and 380 nm). Surface ozone measurements were made by a monitor in Katy, Texas. Both data sets are for July 2004.



**Figure 3.** Frequency distribution is shown for the 8-hour daily maximum ozone readings averaged over all monitors in the Houston, Texas, area as observed during the summers (1 June to 31 August) of 2001–2005 ("All") and during just the month of July each year ("July").

the Moderate Resolution Imaging Spectrometer (MODIS) instrument aboard the Terra satellite. Aerosol optical thickness measurements were taken regularly from the Rice University campus (29.72°N, 95.40°W) during the month of July using the Microtops II instrument [*Mimms*, 1992; *Flynn et al.*, 1996; *Labow et al.*, 1996]. Figure 2 indicates that the highest levels of aerosols over Houston for the month were found on 20 July 2004.

[11] Deployed around the Houston metropolitan area is a network of ground stations with instruments to measure

local air quality and meteorological parameters. Hourly data from the instruments in this network are available from the TCEQ. These surface ozone monitors recorded a broad event of elevated ozone concentrations on 19 and 20 July. The 8-hour NAAQS standard for ozone was violated at 9 of the 37 monitors operational in the Houston area on 20 July 2004 and all monitors in the Houston area reported 8-hour ozone levels > 63 ppbv, resulting in the broadest distribution of elevated 8-hour ozone for the entire month.

[12] Figure 2 also shows 1-hour and 8-hour ozone data for July 2004 from the surface monitor in Katy, Texas (29.81°N, 95.81°W), about 45 km west of downtown Houston and about 75 km west of the major petrochemical industries located around Galveston Bay on the east side of Houston. The 2 days with the highest levels of ozone at this site correspond to the 2 days with high levels of smoke in the atmosphere, and one of the 2 days corresponds to the highest aerosol optical thickness measured by the Microtops instruments.

[13] Figure 3 puts this event in the context of all surface observations over the last 5 summers in Houston. Figure 3 shows a frequency distribution for the Houston area average 8-hour maximum ozone concentrations for "All" summer data (1 June to 31 August) and for just "July" data as recorded by the network of surface monitors. As can be seen, the event of 20 July 2004 was exceeded less than 2% of the time (8 days) during the 5-year period and was the highest event during the July months of 2001–2005.

[14] According to daily burned area data available from a webpage at the Center for International Disaster Information (http://www.cidi.org/wildfire), the summer of 2004 set a new record, 2.7 million hectare, for the annual area burnt in



**Figure 4.** Seven-day back trajectory data generated with the Goddard Trajectory Model connect the air in Houston on 19 July to Alaska and western Canada 6–7 days prior. The gray-shaded region shows the approximate location of the forest fires burning in Alaska and western Canada during early July 2004.



**Figure 5.** TOMS aerosol index data are shown for 12, 14, 16, and 18 July. The data show the progression of an air mass with high aerosol content from western Canada down the Mississippi River Valley into the Houston area.

Alaska. This is more than ten times as much as the longterm annual average of about 0.2 million hectare [*French et al.*, 2002]. Another 3.1 million hectare burned in Canada, 50% above the long-term average [*Stocks et al.*, 2002]. Half of that burned in the Yukon Territory close to the border with Alaska. Fire count data from the NOAA 15 satellite and the MODIS instrument [*Giglio et al.*, 1999; *Justice et al.*, 2002; *Giglio et al.*, 2003] (data not shown) indicate that these fires were most intense during July, with fire events on 12-14 July producing the smoke that was transported across Canada to the Atlantic Ocean and down the Mississippi River Valley to Houston, as we will demonstrate below.

#### 2.2. Trajectories

[15] We use the NASA Goddard Trajectory Model [Schoeberl and Sparling, 1995] in a kinematic mode. The trajectory model employs meteorological data from the Goddard Earth Observing System, Version 4 (GEOS-4) data assimilation product [Bloom et al., 2005] to transport the parcels. The winds from this assimilation are produced every 6 hours on a  $1.0^{\circ}$  latitude  $\times 1.25^{\circ}$  longitude grid. Air parcel trajectories are computed using a time step of 1/50th of a day. At each time step in the model, the meteorological fields are interpolated in time and space to the location of each parcel before calculating the next spatial displacement along the trajectory.

[16] Figure 4 shows the representative 7-day kinematic back trajectories beginning in Houston on 19 July 2004. The trajectories link the air parcels found below 3 km over Houston on 19 and 20 July to initial positions over western Canada and Alaska 1 week earlier.

[17] To test sensitivity of the trajectories to initial parcel placement, the trajectory model was initialized with rings of parcels with radii of 50, 100, and 200 km centered on Houston. (Note that in Figure 4, only the trajectory of the parcel at the center of this ring is plotted.) As the rings advect backward they get distended, providing an indication of the uncertainty of the back trajectory calculations. In general, the evolution of these rings follows the same pattern as the evolution of the central parcels shown in Figure 4. The rings at initial pressures 650 hPa and higher (lower altitudes) passed through Alaska and western Canada while those at lower initial pressures (higher altitudes) originated over the Pacific Ocean.

#### 2.3. Satellite Instruments Track the Plume

[18] Further confirmation of the origins of the smoke observed in Houston on 19 and 20 July 2004 comes from several satellite instruments. Figure 5 shows a time series of aerosol index data from the Total Ozone Mapping Spectrometer (TOMS) aboard the Earth Probe satellite [*Herman et al.*, 1997; *Torres et al.*, 1998] for 12, 14, 16, and 18 July 2004 (see also the accompanying flash animation in the auxiliary material for a movie of the movement of the cloud with high aerosol indices).<sup>1</sup> On 12 July a large region with high aerosol index appears near the Alaskan/Canadian border. This region moves eastward on 14–16 July before dividing on 17 July (not shown) with part of the cloud moving east and part moving south down the Mississippi River Valley toward Houston on 18 July.

[19] Figure 6 shows CO data from the Atmospheric Infrared Sounder (AIRS) instrument on board the Aqua satellite [*McMillan et al.*, 2005] from 12, 14, 16, 18, and 19 July (see also the accompanying flash animation in the

<sup>&</sup>lt;sup>1</sup>Auxiliary material are available in the HTML. doi:10.1029/2006JD007090.



**Figure 6.** CO data from AIRS on 12, 14, 16, 18, and 19 July show the progression of an air mass high in CO across Canada, down the Mississippi River Valley into the Houston area. The AIRS CO data reinforce the TOMS observations seen in Figure 5.

auxiliary material for a movie of the movement of the cloud with high aerosol indices).<sup>1</sup> The AIRS data indicate transport of air from Alaska/western Canada to the Houston area in a manner well correlated with the TOMS data (Figure 5) and the calculated air mass trajectories (Figure 4). Slight differences between the patterns seen in the AIRS CO and TOMS aerosol index data may be attributed to both measurement sensitivities (AIRS CO typically represents midtropospheric values while TOMS aerosol index is related to the top of the aerosol layer and the density of aerosols), differences in the times of the observations (AIRS observations occur at about 1330 local time (LT) while TOMS observations occur at about 1100 LT), and to the fact that CO and aerosols may be subject to different wind fields.



## Houston, TX - July 2004



This difference in transport may be related to the heights at which the two plumes are advected (the particulate matter will typically sink as its transported). As a result, vertical wind shear of the type indicated by the trajectories (Figure 4) would cause some separation of the CO and aerosol plumes. Although we have no direct evidence that such separation occurred in this case, the observed separation of the smoke and ozone plumes associated with Indonesian fires in 1997 reported by *Thompson et al.* [2001] suggests the possibility of such separation in this case as well.

#### 2.4. In Situ Ozone and CO Profile Data

[20] Ozone profiles in Houston are measured using the electrochemical concentration cell (ECC) type [Komhyr, 1986]. Intercomparisons with other ozone measuring instruments have demonstrated the ECC sonde precision to be  $\pm 6\%$  near the ground and -7% to +17% in the upper troposphere [Kerr et al., 1994; Komhyr et al., 1995; Reid et al., 1996].

[21] Pressure, temperature, and humidity measurements are recorded on the ozonesonde payload by Vaisala RS80-15N radiosondes. 350-gram balloons carried our payloads to altitudes of 22–24 km before bursting. We launched 26 ozonesondes, prepared using the Southern Hemisphere Additional Ozonesondes (SHADOZ) protocol [*Thompson et al.*, 2003], between 8 July and 13 August 2004 from the Rice University campus (29.72°N, 95.40°W), located about 5 km south of downtown Houston. Most launches occurred around 1900  $\pm$  30 min Universal Time (UT). The timing of the launches coincides with the typical midafternoon ozone maximum observed by local ground-monitoring stations. The ozonesonde data can be found at http://www.espo. nasa.gov/intex-na/ and http://www.ruf.rice.edu/~ozone.

[22] Comparison of the data from our ozonesondes just before launch with hourly averaged readings from the nearest two ground-level monitors (Texas Avenue and Central Office) shows good agreement over a range of ozone concentrations from near 0 to ~120 ppbvv. Regression analysis indicates an offset of 9 ( $\pm$ 5) ppbv (with the ozonesondes high) and a slope of 0.93 ( $\pm$ 0.09). (Quoted uncertainties represent one standard deviation statistical errors only.) The observed differences reasonably can be attributed to real spatial gradients in ozone across Houston, temporal differences between the ground-level station data and the time of our launches, and the difference in time resolution of the data sets, with our ozonesonde data being nearly instantaneous while the ground-level data we examined were averaged over 1-hour time periods.

[23] We launched two ozonesondes during the period of interest: one at 1932 UT on 19 July and the other at 1900 UT on 20 July. We also launched ozonesondes at 1930 UT on 16 July and 1900 UT on 21 July, before the arrival of and after the departure of the smoke respectively.

[24] Figure 7 summarizes the Houston ozonesonde data from July and August 2004. In Figure 7, the thick black curve represents the mean profile from the 24 afternoon launches between 8 July and 13 August 2004. The gray shaded region represents the observed variability of our ozone profiles at the one standard deviation level.

[25] The thin black curve represents the profile from 16 July, before the arrival of the smoke. It is quite typical of our observations in Houston during IONS, with a well defined mixed layer extending up to 1.5 km altitude.

[26] The black-starred curve represents the ozone profile from 19 July, the first day on which the smoke from the biomass burning was noticeable in the air above Houston. Although ozone below ~1.5 km is only slightly elevated, ozone levels of around 110 ppbv between 2 and 4 km are two to three standard deviations (about 50 ppbv) above the mean. If we define the tropospheric background ozone to be the mixing ratio observed above the enhanced layer, our profiles show an ozone enhancement of ~30 ppbv. Similar enhancements of 30–50 ppbv near 2 km were observed in a forest fire smoke plume during the Southern Oxidants Study in 1995 (SOS95) [*Wotawa and Trainer*, 2000] and of 20– 25 ppbv near 3 km in a smoke plume over Europe during August 1998 [*Forster et al.*, 2001].

[27] The gray-starred curve in Figure 7 represents the ozone profile on 20 July. Elevated levels of ozone of 110-120 ppbv, nearly two standard deviations above the mean, appeared in the mixed layer with still greater enhancements between the top of the mixed layer (~1.6 km) and 3.2 km. At these altitudes, ozone concentrations of nearly 150 ppbv, two to three standard deviations above the mean, are observed. The profile indicates enhancements in ozone of 30-90 ppbv above the tropospheric background level on that day and up to 100 ppbv above the mean profile between the surface and 3 km altitude. The vertical extent of the pollution corresponds well with the calculated trajectories (Figure 4).

[28] The thin gray curve in Figure 7 shows the ozone profile from 21 July, after the smoke has moved out of the Houston area. On this day, the profile looks quite clean, with ozone levels below average from the surface up to  $\sim$ 4.5 km altitude.

[29] Integrating these ozone profiles from the surface to 5 km altitude, we find 22 Dobson units (DU), 34 DU, 36 DU, and 17 DU for 16, 19, 20, and 21 July respectively, where 1 DU is equivalent to  $2.6 \times 10^{16}$  molecules/cm<sup>2</sup>. These data suggest that the transport of biomass burning materials into the atmosphere over Houston led to increases of 50–110% in the first 5 km compared to typical observations. We note that among the 26 ozone profiles collected during IONS in Houston, only the profiles of 19 and 20 July





**Figure 8.** Ozone and CO profiles based on MOZAIC data and output from the GEOS-4 DAS for the Dallas/Ft. Worth area on 18 July 2004 are shown. See the text for details.

showed ozone levels in excess of 100 ppbv at altitudes between the top of the mixed layer and 5 km.

[30] Figure 8 shows CO and ozone data from the Measurement of Ozone by Airbus In-service airCraft (MOZAIC) experiment [*Marenco et al.*, 1998; *Thouret et al.*, 1998] from a flight landing at DFW at ~1830 UT and a flight taking off from DFW at ~2045 UT. The MOZAIC data show enhancements in both the ozone and CO profiles on the afternoon of 18 July at 2–4 km altitude. While the observed ozone amounts are smaller, the vertical location of the pollution layer matches well with that seen in the ozonesonde profiles for 19 and 20 July over Houston (Figure 7). Trajectory analysis (Figure 4), TOMS data (Figure 5), AIRS CO data (Figure 6), and weather satellite images (not shown) all indicate the presence of the smoke-filled air mass in the Dallas/Ft. Worth vicinity on 18 July before its arrival in Houston on 19 July.

#### 2.5. Assimilated Meteorology and Transport Models

[31] To better assess the impact of the transported smokefilled air mass on the local air quality in Dallas on 18 July and in Houston on 19 and 20 July, we examined the output from two different models: FLEXPART, a passive Lagrangian particle transport and dispersion model [*Stohl et al.*, 1998], and the GEOS-4 general circulation model (GCM) and data assimilation system (DAS) [*Bloom et al.*, 2005].

[32] Driven by meteorological data from the European Centre for Medium-Range Weather Forecasts, FLEXPART tracks a forest fire CO tracer emitted between 0 and 3 km over active fires (see A. Stohl et al. (Pan-Arctic enhancements of light absorbing aerosol concentrations due to North American boreal forest fires during summer 2004, submitted to *Journal of Geophysical Research*, 2006) for a more detailed description of the FLEXPART forest fire CO tracer) and predicts a rise in CO levels at about 3 km over Houston beginning late on 18 July and subsiding late on 20 July.

[33] These results qualitatively complement the AIRS CO data and correspond well with the observed ozone increase

on 19 July and decrease on 21 July. Analysis of FLEXPART backward simulations from the ozone sonde profiles (see *Stohl et al.* [2002] for a description of methodology) indicates that for the lowest 5 km of the troposphere above Houston a negligible fraction of the particles originated in the stratosphere during the previous 20 days. Thus enhancements in ozone must be attributed to photochemistry within the air mass rather than descent of stratospheric air rich in ozone.

[34] The GEOS-4 GCM-DAS serve as an interpretive tool in this work. The GEOS-4 DAS produces four daily meteorological analyses that have been used in a number of atmospheric transport calculations, through either trajectory calculations (e.g., Thompson et al., submitted manuscript, 2006) or chemistry-transport modeling [e.g., *Kawa et al.*, 2004].

[35] For this study a number of CO "tracers" are added to the GEOS-4 DAS. Each is tagged with respect to region of origin and type of source. We examine two tagged CO tracers of North American origin, one with anthropogenic sources and one with biomass burning (BB) sources, along with a total CO that includes all global sources. For anthropogenic emissions climatology is used, but the BB emissions are updated daily with MODIS fire counts converted to CO surface fluxes following Heald et al. [2003]. The anthropogenic CO was initialized in March 2004, meaning that it is well "spun up" before the period of interest. The BB CO was set to zero on 12 July in order to isolate the impact of fires occurring during the week preceding the Houston pollution episode. Transport in GEOS-4 DAS is termed "online," since the CO transport is performed at the native model time steps, which means 3.75 min for the resolved winds and 30 min for subgrid processes. In contrast, the dynamics in the trajectory and the "off-line" chemistry and transport models (CTMs) are read from (archived) files, aggregated for 6-hour intervals. The dynamics for the "current" time step is then obtained by linear temporal interpolation. Compared to "off-line" tools, the "online" simulation with GEOS-4 DAS, should more accurately represent transport due to the nonlinear, rapidly evolving subgrid-scale processes, the most important of which are convection and vertical diffusion.

[36] Figure 9 shows the distribution of integrated column CO resulting from local, anthropogenic emissions (left panels) and a release of CO above the eastern Alaskan/ western Canadian forest fires burning during the period 12-20 July. The timing of the arrival of high CO air in Texas in the model coincides well with that seen in Dallas on 18 July and with the higher ozone and aerosol levels observed in Houston on 19 and 20 July.

[37] The gray-starred curve in Figure 8 shows a profile of total CO (all sources) projected by the GEOS-4 DAS over the Dallas area at 2000 UT on 18 July 2004. The CO concentrations predicted by the GEOS-4 DAS are about 10-20 ppbv less than the MOZAIC observations in the mixed layer (below ~1.8 km) and substantially smaller (by a factor of 2 or more) at 2–4 km altitude than the MOZAIC observations in Dallas on 18 July, completely missing the enhanced features seen in the MOZAIC profile at ~2.3 km and near 3.5 km.

[38] Regarding the CO partitioning, the model suggests that the Alaskan/Canadian fires of 12 July contributed about

# North American CO Column Integrals



**Biomass Burning Sources** 



**Figure 9.** Integrated total column carbon monoxide from (left) anthropogenic and (right) biomass burning sources on (top) 18, (middle) 19, and (bottom) 20 July as indicated by GMAO model results are shown. The red areas indicate large amounts while the white areas indicate small amounts. See the text for details.

one third of the total CO below 2.5 km at Dallas on 18 July and less than 25% of the total CO in Houston on 19 and 20 July. Although the timing of the arrival of enhanced CO in Dallas and Houston is reproduced well in the model (as indicated by Figure 9), the model is unable to reproduce the large enhancements in CO above the mixed layer as seen in the MOZAIC data over Dallas (Figure 8). This discrepancy may be attributed to uncertainty in the vertical distribution of emissions from the Alaskan fires (see S. Turquety et al., Inventory of boreal fire emissions for North America in 2004: The importance of peat burning and pyro-convective injection, submitted to Journal of Geophysical Research, 2006) or in the transport resulting from the assimilated meteorological fields. Such impacts will be investigated in future work. By missing this large enhancement over Dallas, the model may then underestimate the impact of the biomass burning pollution downwind in Houston on 19 and 20 July.

#### 3. Summary and Conclusions

[39] This paper has provided evidence for the impact of remote biomass burning on ozone pollution levels above Houston, Texas, on 19 and 20 July 2004. Forest fires burning in Alaska and western Canada during early July produced a cloud of smoke containing elevated levels of CO, ozone, and ozone precursor species. This air mass was transported across Canada from 12-17 July at which time it split, with part of the air mass being transported down the Mississippi River Valley. Satellite data from NOAA 15 indicate the generation of the smoke in Alaska and western Canada from fires burning 12-14 July. AIRS CO and TOMS aerosol index data tracked the plume across Canada 12–16 July, down the Mississippi River Valley 17–18 July, through Dallas on 18 July and into Houston on 19–20 July. MOZAIC data from flights into and out of DFW on 18 July showed enhanced ozone and CO between 2 and 4 km altitude. On 19 and 20 July, smoke was observed in Houston, where ozone levels between the surface and 5 km altitude increased 50-110%.

[40] Model results support our interpretation of the ozonesonde in situ data. Results from the NASA Goddard Trajectory Model link the air mass in Houston on 19 July to origins in Alaska and western Canada 6–7 days earlier. Results from FLEXPART indicate the arrival of higher CO levels in Houston late on 18 July with subsidence on 21 July, in good agreement with our ozonesonde data. In addition, FLEXPART suggests that the elevated ozone concentrations seen in our ozonesonde profiles are not of stratospheric origin. The GEOS-4 results demonstrate good agreement in the timing of the arrival of the polluted air mass over Dallas and Houston. The model seems to underestimate, however, the impact on the local air quality of the transported pollution, as indicated by the comparison of the model CO with in situ measurements from MOZAIC over Dallas.

[41] Of the possible explanations for our observations, our data suggest and are consistent with the following interpretation: pollution from the Alaskan and Canadian forest fires arrived in the Houston area at or just above the level of the mixed layer on 19 July. Some of this polluted air was entrained in the mixed layer over Houston during its daytime growth on 19 July, exacerbating the local surface pollution. Similarly, some locally generated pollution from Houston mixed into the lower free troposphere, enhancing the ozone concentrations above the mixed layer through reactions with the precursors transported into the Houston area along with the smoke. Enhancements in the ozone profile above the mixed layer observed in Houston on 19 and 20 July are more substantial than those seen above Dallas on 18 July.

[42] Similar events likely have occurred repeatedly in the past several decades, during which time Houston has struggled with ozone pollution levels. In the case of this event, the arrival in Houston of smoke containing ozone and ozone precursor species generated from forest fires burning in Alaska and western Canada can be associated with substantial increases in ozone pollution levels above Houston. Since such natural episodes will continue to occur in the future, state implementation plan designs will need to account for the influence of such remote events in order to attain compliance with EPA air quality standards in the Houston area.

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#### References

- Banta, R. M., C. J. Senff, J. Nielsen-Gammon, L. S. Darby, T. B. Ryerson, R. J. Alvarez, S. P. Sandberg, E. J. Williams, and M. Trainer (2005), A bad air day in Houston, *Bull. Am. Meteorol. Soc.*, 86, 657–669.
- Berkowitz, C. M., T. Jobson, G. Jiang, C. Spicer, and P. V. Doskey (2004), Chemical and meteorological characteristics associated with rapid increases of O<sub>3</sub> in Houston, Texas, *J. Geophys. Res.*, 109, D10307, doi:10.1029/2003JD004141.
- Bell, M. L., A. McDermott, S. L. Zeger, J. M. Sarnet, and F. Dominici (2004), Ozone and short-term mortality in 95 US urban communities, 1987–2000, *J. Am. Med. Assoc.*, 292, 2372–2378.
- Bloom, S., et al. (2005), Documentation and validation of the Goddard Earth Observing System (GEOS) data assimilation system—Version 4, *NASA Tech. Memo. NASA TM-2005-104606*, vol. 26.
- Colarco, P. R., M. R. Schoeberl, B. G. Doddridge, L. T. Marufu, O. Torres, and E. J. Welton (2004), Transport of smoke from Canadian forest fires to the surface near Washington, D.C.: Injection height, entrainment, and optical properties, J. Geophys. Res., 109, D06203, doi:10.1029/ 2003JD004248.
- Daum, P. H., L. I. Kleinman, S. R. Springston, L. J. Nunnermacker, Y.-N. Lee, J. Weinstein-Lloyd, J. Zheng, and C. M. Berkowitz (2004), Origin and properties of plumes of high ozone observed during the Texas 2000 Air Quality Study (TexAQS 2000), J. Geophys. Res., 109, D17306, doi:10.1029/2003JD004311.
- Environmental Protection Agency (1999), Guideline for reporting of daily air quality—Air quality index (AQI), *EPA-454/R-99-101*, Research Triangle Park, N. C.
- Flynn, L. E., G. J. Labow, R. A. Beach, M. A. Rawlins, and D. E. Flittner (1996), Estimation of ozone with total ozone portable spectroradiometer instruments. I. Theoretical model and error analysis, *Appl. Opt.*, 35, 6076–6083.
- Forster, C., et al. (2001), Transport of boreal forest fire emissions from Canada to Europe, *J. Geophys. Res.*, *106*, 22,887–22,906.
- French, N. H. F., E. S. Kasischke, and D. G. Williams (2002), Variability in the emission of carbon-based trace gases from wildfire in the Alaskan

boreal forest, J. Geophys. Res., 108(D1), 8151, doi:10.1029/2001JD000480.

- Giglio, L., J. D. Kendall, and C. O. Justice (1999), Evaluation of global fire detection algorithms using simulated AVHRR infrared data, *Int. J. Remote Sens.*, 20, 1947–1985.
- Giglio, L., J. Descloitres, C. O. Justice, and Y. Kaufman (2003), An enhanced contextual fire detection algorithm for MODIS, *Remote Sens. Environ.*, 87, 273–282.
- Goode, J., R. J. Yokelson, D. E. Ward, R. A. Susott, R. E. Babbitt, M. A. Davies, and W. M. Hao (2000), Measurements of excess O<sub>3</sub>, CO<sub>2</sub>, CO, CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>, HCN, NO, NH<sub>3</sub>, HCOOH, CH<sub>3</sub>COOH, HCHO, and CH<sub>3</sub>OH in 1997 Alaskan biomass burning plumes by airborne Fourier transform infrared spectroscopy (AFTIR), *J. Geophys. Res.*, 105, 22,147–22,166.
- Heald, C. L., et al. (2003), Asian outflow and transpacific transport of carbon monoxide and ozone pollution: An integrated satellite, aircraft and model perspective, *J. Geophys. Res.*, 108(D24), 4804, doi:10.1029/ 2003JD003507.
- Herman, J. R., P. K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier (1997), Global distribution of UV-absorbing aerosols from Nimbus-7/ TOMS data, J. Geophys. Res., 102, 16,911–16,922.
- Justice, C. O., L. Giglio, S. Korontizi, J. Owns, J. Morisette, D. Roy, J. Descloitres, S. Alleaume, F. Petitcolin, and Y. Kaufman (2002), The MODIS fire products, *Remote Sens. Environ*, 83, 244–262.
- Kawa, S. R., D. J. Erickson, S. Pawson, and Y. Zhu (2004), Global CO<sub>2</sub> simulations using meteorological data from the NASA data assimilation system, J. Geophys. Res., 109, D18312, doi:10.1029/2004JD004554.
- Kerr, J. B., et al. (1994), The 1991 WMO international ozonesonde intercomparison at Vanscoy, Canada, Atmos. Ocean, 32(4), 685–716.
- Komhyr, W. D. (1986), Operations handbook: Ozone measurements to 40 km altitude with mode 4A electrochemical concentration cell (ECC) ozonesondes (used with 1680-Mhz radiosondes), NOAA Tech. Memo. ERLARL-149.
- Komhyr, W. D., R. A. Barnes, G. B. Brothers, J. A. Lathrop, and D. P. Opperman (1995), Electrochemical concentration cell performance evaluation during STOIC, *J. Geophys. Res.*, 100, 9231–9244.
- Labow, G. J., L. E. Flynn, M. A. Rawlins, R. A. Beach, C. A. Simmons, and C. M. Schubert (1996), Estimation of ozone with total ozone portable spectroradiometer instruments. II. Practical operation and comparisons, *Appl. Opt.*, 35, 6084–6089.
- Lamarque, J.-F., et al. (2003), Identification of CO plumes from MOPITT data: Application to the August 2000 Idaho-Montana forest fires, *Geophys. Res. Lett.*, 30(13), 1688, doi:10.1029/2003GL017503.
- Lippmann, M. (1991), Health effects of tropospheric ozone, *Environ. Sci. Technol*, 25, 1954–1962.
- Marenco, A., et al. (1998), Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, an overview, *J. Geophys. Res.*, 103, 25,631–25,642.
- McConnell, R., K. Berhane, F. Gilliland, S. J. London, T. Islam, W. J. Gauderman, E. Avol, H. G. Margolis, and J. M. Peters (2002), Asthma in exercising children exposed to ozone: A cohort study, *Lancet*, 359, 386–391.
- McMillan, W. W., C. Barnet, L. Strow, M. T. Chahine, M. L. McCourt, J. X. Warner, P. C. Novelli, S. Korontzi, E. S. Maddy, and S. Datta (2005), Daily global maps of carbon monoxide from NASA's Atmospheric Infrared Sounder, *Geophys. Res. Lett.*, 32, L11801, doi:10.1029/ 2004GL021821.
- Mimms, F. M. (1992), How to measure the ozone layer, *Sci. Probe*, 2, 45–51.
- Pfister, G., P. G. Hess, L. K. Emmons, J.-F. Lamarque, C. Wiedinmyer, D. P. Edwards, G. Pétron, J. C. Gille, and G. W. Sachse (2005), Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data, *Geophys. Res. Lett.*, 32, L11809, doi:10.1029/2005GL022995.

- Reid, S. J., G. Vaughan, A. R. W. Marsh, and H. G. J. Smit (1996), Accuracy of ozonesonde measurements in the troposphere, *J. Atmos. Chem.*, 25, 215–226.
- Rogers, C. M., and K. P. Bowman (2001), Transport of smoke from the Central American fires of 1998, J. Geophys. Res., 106, 28,357–28,368.
- Schoeberl, M. R., and L. Sparling (1995), Trajectory modeling, in *Diagnostic Tools in Atmospheric Physics, Proceedings of the International School of Physics "Enrico Fermi"*, vol. 124, edited by G. Fiocco and G. Visconti, pp. 289–306, IOS Press, Washington, D. C.
- Stocks, B. J., et al. (2002), Large forest fires in Canada, 1959–1997, J. Geophys. Res., 108(D1), 8149, doi:10.1029/2001JD000484.
- Stohl, A., M. Hittenberger, and G. Wotawa (1998), Validation of the Lagrangian particle dispersion model FLEXPART against large scale tracer experiment data, *Atmos. Environ.*, 24, 4245–4264.
- tracer experiment data, *Atmos. Environ.*, 24, 4245–4264. Stohl, A., S. Eckhardt, C. Forster, P. James, N. Spichtinger, and P. Seibert (2002), A replacement for simple back trajectory calculations in the interpretation of atmospheric trace substance measurements, *Atmos. Environ.*, 36, 4635–4648.
- Thompson, A. M., J. C. Witte, R. D. Hudson, H. Guo, J. R. Herman, and M. Fujiwara (2001), Tropical tropspheric ozone and biomass burning, *Science*, 291(5511), 2128–2132.
- Thompson, A. M., et al. (2003), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology: 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, J. Geophys. Res., 108(D2), 8238, doi:10.1029/ 2001JD000967.
- Thouret, V., A. Marenco, J. A. Logan, P. Nedelec, and C. Grouhel (1998), Comparisons of ozone measurements from the MOZAIC airborne program and the ozone sounding network at eight locations, *J. Geophys. Res.*, 103, 25,695–25,720.
- Torres, O., P. K. Bhartia, J. R. Herman, Z. Ahmad, and J. Gleason (1998), Derivation of aerosol properties from satellite measurements of backscattered ultraviolet radion: Theoretical basis, *J. Geophys. Res.*, 103, 17,099–17,110.
- Wotawa, G., and M. Trainer (2000), The influence of Canadian forest fires on pollutant concentrations in the U.S., *Science*, 288, 324–328.
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