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Tropospheric ozone sources and wave activity over Mexico City and Houston during MILAGRO/Intercontinental Transport Experiment (INTEX-B) Ozonesonde Network Study, 2006 (IONS-06)

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Abstract

During the INTEX-B (Intercontinental Chemical Transport Experiment)/MILAGRO (Megacities Initiative: Local and Global Research Observations) experiments in March 2006 and the associated IONS-06 (INTEX Ozone-sonde Network Study; <http://croc.gsfc.nasa.gov/intexb/ions06.html>), regular ozone-sonde launches were made over 15 North American sites. The soundings were strategically positioned to study inter-regional flows and meteorological interactions with a mixture of tropospheric O₃ sources: local pollution; O₃ associated with convection and lightning; stratosphere-troposphere exchange. The variability of tropospheric O₃ over the Mexico City Basin (MCB; 19 N, 99 W) and Houston (30 N, 95 W) is reported here. MCB and Houston profiles displayed a double tropopause in most soundings and a subtropical tropopause layer with frequent wave disturbances, identified through O₃ laminae as gravity-wave induced. Ozone-sondes launched over both cities in August and September 2006 (IONS-06, Phase 3) displayed a thicker tropospheric column O₃ (~7 DU or 15–20%) than in March 2006; nearly all of the increase was in the free troposphere. In spring and summer, O₃ laminar structure manifested mixed influences from the stratosphere, convective redistribution of O₃ and precursors, and O₃ from lightning NO. Stratospheric O₃ origins were present in 39% (MCB) and 60% (Houston) of the summer sondes. Comparison of summer 2006 O₃ structure with summer 2004 sondes (IONS-04) over Houston showed 7% less tropospheric O₃ in 2006. This may reflect a sampling contrast, August to mid-September 2006 instead of July–mid August 2004.

1 Introduction

A number of multi-national, multi-platform field experiments have been conducted to quantify intra- and intercontinental pollution transport. The 2004 ICARTT (International Consortium on Atmospheric Research on Transport and Transformation)/INTEX (Intercontinental Transport Experiment – North America) (Fehsenfeld et al., 2006; Singh et

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al., 2006) and 2006 MILAGRO/INTEX-B (Fast et al., 2007; Molina and Artaxo, 2007; Singh et al., 2007) used multiple aircraft to follow pollutant transport into and out of North America. With the launch of the Aura spacecraft carrying four sensors in 2004 (<http://aura.gsfc.nasa.gov>; Schoeberl et al., 2007), tropospheric O₃, a major pollutant, could be measured with some of its precursors (Zhang et al., 2006), providing a larger view of pollution transport. Although surface monitoring for pollutants like O₃, CO and aerosol, is useful, trans-boundary and intercontinental transport often take place in the free troposphere (FT), just above the mixed layer (1–3 km) or in the mid-troposphere. The mid-troposphere is also subject to direct injection of pollutants from convection, introduction of NO from lightning (Pickering et al., 1992; Ridley et al. 1996; Huntrieser et al., 2002) and stratospheric O₃, frequently leading to thin layers and transport over long distances (Newell et al., 1999; Oltmans et al., 2004).

Neither satellite nor aircraft instruments resolve pollutant structure from surface through the lower stratosphere, something that combined ozonesonde-radiosonde packages from balloons offer for O₃. Thus, strategically designed sounding networks are an integral part of field campaigns. IONS-06 (INTEX Ozonesonde Network Study) operating during MILAGRO/INTEX-B (Phase 1=March 2006; Phase 2=mid-April-mid-May 2006; Phase 3=August-mid-September 2006), supplied >700 O₃ and pressure-temperature-humidity (PTU) profile sets from surface to ~35 km (5 hPa) at 23 North American stations (Table 1, circles in Fig. 1). The IONS-06 soundings provided profiles for Aura instrument validation (e.g. Jiang et al., 2007; Schoeberl et al., 2007), for integrating field measurements and for evaluation of models (e.g. Pierce et al., 2007; Parrington et al., 2008).

The IONS-06 sonde network built on a similar effort, IONS-04, during the INTEX-A campaign; refer to the special INTEX-A/ICARTT Journal of Geophysical Research-Atmospheres issues, Vol. 111 (D23; 2006) and 112 (D12; 2007). IONS-04 was initiated to complement North American aircraft sampling and satellite measurements of O₃, temperature and humidity (Thompson et al., 2007a, b), with eleven sites and sonde launches timed for Aura overpasses. IONS-04 configured sampling from the south

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central US through New England and the maritimes (Fig. 1, solid dots) to encompass major pollution sources and the route of eastward export from North America (* symbol in Table 1 gives IONS-04 station coordinates). Combining forecasts with near-real-time O₃ data permitted INTEX-A and ICARTT aircraft to target sampling over North America, the Atlantic and Europe.

Unexpected findings in IONS-04 were: (1) the prevalence of high-O₃ concentrations in the middle and upper troposphere (UT) (Cooper et al., 2006; Thompson et al., 2007a) and (2) signatures of wave activity in stable laminae within most O₃ profiles (Thompson et al., 2007a, b). INTEX-A/ICARTT and IONS-04 took place in summertime (early July to mid-August 2004). Boreal fires contributed to O₃ pollution layers in the free troposphere (FT) over North America and beyond (e.g. Mao et al., 2006; Pfister et al., 2006; Val Martín et al., 2007). These layers were interleaved with O₃ laminae associated with convection, lightning and a robust stratospheric O₃ source. Over northeastern North America, tracer correlations and trajectories (Thompson et al., 2007b) assigned a tropospheric O₃ budget: stratospheric injection (25%); boundary-layer pollution (10%); regional convective redistribution of O₃ and/or its precursors, including lightning (15%); advected pollution and/or background O₃ (50%). The tropospheric O₃ column over northeastern North America averaged 45 DU (Dobson Units; 1 DU=2.69×10¹⁶ cm⁻²), with ~22–26 DU consisting of advected and/or background air. Values at Houston in 2004 were similar, 48 DU (total troposphere) and 28 DU (advected).

The three-phase IONS-06 (sample distribution in Table 1), addressed the broad set of transport issues posed by INTEX-B/MILAGRO (<http://www.eol.ucar.edu/projects/milagro>; Molina et al., 2008; <http://croc.gsfc.nasa.gov/intexb>; Singh et al., 2008) and TEXAQS (Texas Air Quality Study/GOMACCS (Gulf of Mexico Atmospheric Composition and Climate Study <http://esrl.noaa.gov/csd/2006>)). During MILAGRO/INTEX-B (March 2006), Phase 1 of IONS-06 coincided with aircraft sampling over Texas, Mexico and the Gulf of Mexico, to investigate export from the Mexico City Basin (MCB). Phase 2 supported INTEX-B aircraft sampling over Pacific and north-western North

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American inflow regions. A seasonal contrast was effected by IONS Phase-3 soundings in August and September 2006 when aircraft, ground and shipboard sampling were concentrated over Houston during the TEXAQS-06/GOMACCS.

In this paper, we quantify wave activity and tropospheric O₃ budgets over the MCB and Houston urban areas, using analytical approaches similar to the IONS-04 studies (Thompson et al., 2007a, b). With a focus on FT O₃ in Phase 1 (early spring) and Phase 3 (late summer), the following questions are addressed:

- How does O₃ vertical structure over MCB and Houston vary day-to-day? Do we detect a similar mixture of influences (stratosphere, regional convection and lightning, advection) to those computed in INTEX-A and IONS-04?
- How do mean profiles and O₃ budgets between spring and summer compare? Are these consistent with meteorological patterns over Houston and the MCB, where the MILAGRO period has been summarized by Fast et al. (2007)?
- What is the role of gravity waves, a prevalent dynamical signal, in determining upper troposphere and lower stratosphere O₃ over MCB and Houston?
- How do the wave-affected laminae, tracers and trajectories translate into O₃ sources? Are there links in pollution between Houston and MCB?

Over Houston, tropospheric O₃ during Phase 3 of IONS-06, August–mid-September 2006, is compared with O₃ profiles from the summertime IONS-04 sounding activity over Houston (Morris et al., 2006; Thompson et al., 2007a).

2 Experimental IONS-06 observations methods of analysis

Ozone profiles over Houston (30 N, 95 W) and MCB (20 N, 99 W) were taken as part of IONS-06 using electrochemical concentration cell instruments coupled with standard radiosondes. The total distribution of IONS-06 sites and sample numbers appear in

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Table 1. The sample number designations combine the Phase 1 and 2 sondes under Spring; Phase 3 is referred to as Summer. For Phase 3 analyses of O_3 over Houston, profiles are used from the University of Houston launch site (August 2006) and the R/V Ronald H. Brown oceanographic ship (September 2006). Most launches took place in early afternoon local time. Vertical resolution is effectively 50–100 m (Smit et al., 2007), sufficient to detect stable layers of locally elevated (or suppressed) O_3 . Images of all IONS O_3 are at: <http://croc.gsfc.nasa.gov/intexb/ions06.html> (hereafter referred to as the IONS-06 website).

As for IONS-04 (Thompson et al., 2007a), tracers for O_3 origins include: (1) radiosonde PTU profiles; (2) Ertel's potential vorticity (pv; $1\text{ pvu}=10^{-6}\text{ m}^2\text{ s}^{-1}/\text{K}$) computed from the Goddard Earth Observing System Assimilation Model (GEOS-version 4; Bloom et al., 2005); (3) forward and backward air-parcel trajectories for each launch location and date, calculated with the kinematic version of the GSFC trajectory model (Schoeberl and Sparling, 1995) using GEOS meteorological fields at a 1×1 -degree grid. For the MCB care is taken to use terrain following trajectories only. Additional meteorological data, satellite imagery and trajectory-based domain-filling products are taken from <http://croc.gsfc.nasa.gov/intexb>.

Ozone and potential temperature (θ) laminae, as described in Teitelbaum et al. (1994) and Pierce and Grant (1998) are used to identify signatures of Rossby (RW) or Gravity waves (GW). Wave frequencies at a given altitude for spring and summer are determined from the percentage of soundings within the season that have laminae with the RW or GW designation. The RW and GW classifications are refined in the “laminar identification (LID)” method to determine tropospheric O_3 budgets according to four contributions, as outlined in Thompson et al. (2007a): (1) stratospheric injection (ST, from a subset of RW-identified layers, based on H_2O and pv tracers); (2) regional redistribution of convection and/or lightning (RCL, taken as equivalent to GW over the tropics and sub-tropics); (3) local BL, based on integrating O_3 from the surface to the top of the mixed layer, with the latter parameter determined from the PTU; (4) advected O_3 (AD), the residual contribution, after (1)–(3), computed in column thickness (Dobson

Units), have been subtracted from total tropospheric O₃. AD represents either recently transported or aged, background O₃. The use of an individual, dynamically derived BL height is an update to the Thompson et al. (2007a,b) method. Over MCB, the BL at mid-day launch averages ~1.8 km above the surface. Fast et al. (2007) note that later in the day, under convective influence, the MCB BL may extend to 2.6 km above surface. Over Houston, the BL is ~1 km, the value used in Thompson et al. (2007a, b).

3 March 2006 (MILAGRO/INTEX-B) profiles and wave activity

3.1 MCB and Houston ozone and relative humidity structure

Curtains of O₃ mixing ratio in 0.25-km averages over the MCB and Houston below 17 km (95 hPa), with the tropopause in white, appear in Fig. 2. The FT over MCB has moderate O₃, 50–60 ppbv (Fig. 2a) with some clean layers, <35 ppbv, between 5 and 13 km. These layers originate mostly from tropical marine areas, with occasional recirculation over the nearby Caribbean (refer to trajectories at the IONS-06 website, <http://croc.gsfc.nasa.gov/intexb/ions06.html>). Marine origins are implicated by locally moist layers above 9 km in a number of the soundings. The FT over Houston (Fig. 2b) is sometimes as clean as over MCB but O₃ mixing ratios between 6 and 11 km average 10 ppbv higher over Houston than MCB (contrast Fig. 2c and d).

Over MCB the tropopause is consistently high, averaging 15.5 km (Fig. 2a). The O₃ structure between 13 and 16 km describes a subtropical tropopause transition layer with O₃ mixing ratio varying between 90 and 130 ppbv. All the March MCB and Houston profiles display a double ozonopause (see individual profiles at the IONS-06 website). On some days there are multiple laminae with stratospheric influence throughout the FT. For example, on 11 March 2006 two relatively dry layers over Houston (Fig. 3a), at 250 and 350 hPa, are distinguished by back-trajectories that pass over regions of elevated pv gradient over the Pacific (Fig. 3b). (In Sect. 5, it is shown that the LID method computes significant ST O₃ for the 11 March 2006 Houston profile). The uv-

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DIAL O₃ instrument on the NASA DC-8 noted stratospheric air to 4 km over Houston on its 11 March 2006 MILAGRO/INTEX-B flight (Butler et al., 2007).

Sondes and trajectories were used to explore the hypothesis that MCB and Houston O₃ pollution are linked (see profiles and trajectories on the IONS-06 website).

Note that the sondes are somewhat limited compared to aircraft observations with multiple tracers. However, direct links between MCB and Houston are not easy to establish because comprehensive aircraft data coverage throughout the MILAGRO/INTEX-B region is uneven. For the 11 March 2006 case noted above, there appears to be MCB influence in the Houston pollution O₃ layer at 750 hPa (corresponding trajectory in Fig. 3b), but origins are actually north of MCB. The NASA DC-8 flight report for 11 March 2006 sampling over Houston, the Gulf of Mexico and MCB noted a complex O₃ structure and a mixture of influences in the larger region (<http://catalog.eol.ucar.edu/milagro/report/dc-8/20060311>). An extended range of MCB impact was noted, based on uv-DIAL O₃ and aerosols (Butler et al., 2007), as well as other tracers. In some places biomass burning was implicated. In others, clean layers throughout the FT are noted. The complexity of flows and shifts in overall meteorology during MILAGRO are discussed by Fast et al. (2007; see also Fast and Zhong (1998) for a larger context). The IONS-06 soundings encompass two periods of northerly flow, “Norte” events, discussed by Fast et al. (2007); they ended prior to the third Norte episode.

Trajectories forward from the MCB O₃ sounding on 16–18 March 2006, suggest MCB flows toward Houston and the northern Gulf of Mexico, but the layers were not notably polluted (see website). Forward trajectories from MCB on 17 March 2006 (700 hPa level, O₃ mixing ratio 50–60 ppbv, were predicted to pass over Houston 1–2 days later, but no Houston soundings were taken on 18 and 19 March 2006. The DC-8 detected moderate pollution influences from MCB near Houston and the Gulf (see http://catalog.eol.ucar.edu/milagro/report/dc-8/20060319/report.DC-8.200603191635.mission_summary.pdf).

Mean O₃, *T* and relative humidity (RH) profiles over MCB show that a moderately

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polluted mixed layer (to 5 km; Fig. 2c) gives way to a cleaner layer with relatively invariant O_3 up to 10 km (Fig. 2c). Above 10 km, the MCB O_3 mean and standard deviation increase. RH increases from 7 km to 10.5 km over MCB, suggesting surface origins, ie convective outflow in the mid-troposphere. A more complex tropopause structure over Houston, relative to MCB, displays a highly structured mean O_3 profile above 13 km (Fig. 2b, d), with a wide range of mixing ratios, 45–180 ppbv. Consistent with the general pattern of higher O_3 over Houston above 11 km than over MCB (also drier over Houston, suggesting stratospheric influence), the mean T over Houston is greater than T over MCB. It is inferred that MCB has more tropical air with a colder tropopause layer.

3.2 Wave activity over MCB and Houston

Stable laminae associated with wave activity, as well as thin layers of advected pollution, are standard features in O_3 soundings. The morphology of wave types provides insight into processes affecting O_3 soundings. Higher correlation of θ - O_3 laminae, classified as GW by Teitelbaum et al. (1994) represents vertical displacements and potential convective activity. An RW designation indicates horizontal displacements, i.e. flow along isentropes, introducing stratospheric air into the troposphere. Figure 4 displays the frequency of GW and RW activity during March 2006 over the MCB and Houston. Combined GW and RW activity is most effective above 8 km for Houston (black lines), giving rise to more O_3 laminae and variability in the tropopause layer (Fig. 2d) than in the corresponding region over MCB (Fig. 2c). Over MCB, the increase of GW and RW in the subtropical tropopause layer is most pronounced above 12 km. The signature of RW activity above 12 km for MCB (Fig. 4) is consistent with the appearance of the very dry, high- O_3 layer at ~ 160 hPa in virtually all the March soundings (see IONS-06 website). Over the MCB there is also a high GW frequency at 3–4 km, presumably where dry convection mixes BL and lower FT air. This is where Fast et al. (2007) place the top of the convective BL. Note two O_3 maxima over the MCB at 3.5 and 5 km (Fig. 2c), corresponding to the location of the GW feature (Fig. 4) and to the green shading, 50–60 ppbv, in the mixing ratio curtain (Fig. 2a).

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4 August–September 2006 ozone structure

Figure 5a, b displays MCB and Houston O_3 mixing ratio curtains below 17 km in Phase 3 of IONS-06 along with mean O_3 , T and RH profiles (Fig. 5c, d). Below 10 km over the MCB (Fig. 5c), RH increases greatly in August–September, the onset of the North American monsoon, compared to spring, the late dry season. For MCB the March 2006 RH averages 10–20% near the surface (Fig. 2c); the August–September mean exceeds 60% below 7 km (Fig. 5c). Compared to spring, mean O_3 mixing ratios at 8–12 km over both cities increase 20–35 ppbv. Over Houston (Fig. 5b) in August there is more yellow-orange-red above 6 km (>90 ppbv) as well as a more sharply defined tropopause (Fig. 5d), relative to spring (Fig. 2b, d).

In summer, Houston O_3 profiles (August–September mean is the solid line in Fig. 5d) fall into a bimodal pattern. UT O_3 profiles with >100 ppbv features above 8 km are rare after 25 August 2006 (Fig. 5b). Pre- and post-25 August mean O_3 profiles (depicted as dashed (1–25 August) and dotted (26 August–11 September) with their respective RH profiles in Fig. 5d) have little overlap to within $1-\sigma$ (not shown).

Cooper et al. (2006, 2007), compositing IONS-04 and IONS-06 profiles with MOZAIC (Measurements of Ozone by Airbus In-service Aircraft) landing/takeoff data from major airports (e.g. Dallas-Fort Worth, Atlanta), note that elevated UT O_3 appears to be a broad feature over summertime eastern North America. For the southern US IONS sites (Houston, Huntsville in 2004 and 2006), Cooper et al. (2006, 2007) use lightning flash data with FLEXPART to attribute most of the elevated O_3 in the 10–12 km layer to lightning. For IONS-06, the southern US lightning interpretation (and significant UT O_3 from stratospheric sources over eastern North America), is further supported by analysis with a coupled chemical-transport model (Cooper et al., 2007).

Lightning-produced NO, along with post-convective redistribution of O_3 and/or O_3 precursors, has also been linked to FT O_3 layers over MCB in August–September 2006 (Ladino et al., 2007). This interpretation is consistent with a spring-to-summer RH increase above 12 km (deep convective outflow layers) over both cities (cf. Figs. 2c,

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d and 5c, d). Trajectory analysis (see IONS-06 website) shows that transport is even more decoupled between the MCB and Houston in August–September 2006 than in March 2006. Most of the air parcels from MCB head west; origins of O₃ over MCB tend to be relatively localized. A few back-trajectories from Houston are from recirculation over the Gulf of Mexico; most are from the west. Flows away from Houston head north.

5 Tropospheric wave influence and ozone budgets

5.1 Spring 2006

The amount of tropospheric O₃ in layers associated with RW and GW combined is 40% and 44% over MCB and Houston, respectively, in March 2006. Similar values hold for August–September 2006. When filtered with tracers, the corresponding March O₃ amounts translate to a combined ST and RCL fraction equivalent to 22% (MCB) and 32% (Houston) of the tropospheric column (Table 2). At Houston the ST fraction is twice that over MCB; convective influence (RCL in Table 2) is about the same over both cities. For the MCB the average tropospheric O₃ column is 11 DU lower than for Houston (Table 2), even though the 2-km thick BL at MCB has 1/3 more O₃ than the Houston BL. This appears to be due to a smaller FT O₃ column over MCB.

During March approximately half the O₃ is designated AD (Table 2), representing a mixture of recently imported O₃ pollution and a background amount of indeterminate origins, presumably mixed ST, RCL and earlier imported O₃ pollution. Figure 6 depicts tropospheric O₃ budgets computed by LID for daytime profiles over MCB and Houston in the March and August–September 2006 periods. In all cases there is considerable day-to-day variability in total tropospheric O₃ amounts as well as in individual budget fractions. The large and variable amounts of BL O₃ over MCB on 5–7 March and on 13 March 2006 (Fig. 6a) are partly due to a BL height >2 km above ground (not shown). Figure 6a indicates considerable ST O₃ over MCB on five days in March 2006. Over Houston ST O₃ is a consistent feature after 8 March 2006 (Fig. 6b). Individual GW

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fractions are ~30% in early March 2006. After 8 March, a greater ST O₃ amount sets in, driven by a 20–40% RW frequency over the remainder of MILAGRO/INTEX-B.

5.2 Summer 2006

Both the MCB and Houston increased in tropospheric O₃ column 7–8 DU in the spring-to-summer transition (Table 2). The BL O₃ amounts account for only 1 DU of the increase. Summer ST O₃ amounts (Table 2) did not change, although the ST fractions of total and free tropospheric O₃ decline 15–20% compared to the spring values. Stratospheric influence in the summer O₃ budgets (Fig. 6c, d) is significant, appearing in 39% of the MCB profiles and 60% of the Houston soundings.

Comparing Fig. 2c and d with Fig. 5c and d reveals that most of the spring-to-summer O₃ increases occur between 5 and 12 km. Table 2 shows corresponding enhanced AD amounts, 5 DU over MCB and 8 DU over Houston. Recall that the AD term consists of recently imported or recirculated O₃ as well as an O₃ background that includes earlier ST, RCL and imported O₃. Increases in relatively fresh imported or recirculated O₃ and an elevated background O₃ are both reasonable explanations for the higher AD O₃. Between the March (MILAGRO, IONS-06 Phase 1) and August–September 2006 sampling periods, RCL and ST fractions over Houston decline ~30% relative to their March contributions (Table 2). This is likely to be a consequence of a highly convective spring over Houston (Fast et al., 2007). The 500 hPa geopotential anomalies over southeastern Texas (not shown; refer to the website <http://www.cdc.noaa.gov>) show >50 m greater-than-normal heights in March 2006. The RCL and ST O₃ decreases from March to August–September 2006 over Houston are compensated for with higher AD, from 53% of the tropospheric O₃ column to 63%. All four O₃ component fractions change relatively little in the seasonal transition over MCB.

Budget variations over Houston in August–September (Fig. 6c, d) reflect wave activity and the bimodal O₃ profile comparisons (Sect. 4, Fig. 5c, d). Enhanced convection and lightning (higher O₃ concentrations and RH above 5 km) are more concentrated through 25 August (Fig. 5d, dashed profiles), corresponding to greater GW activity and

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