Use of tethersonde and aircraft profiles to study the impact of mesoscale and microscale meteorology on air quality

Gina M. Mazzuca [a, *], Kenneth E. Pickering [a, b], Richard D. Clark [c], Christopher P. Loughner [g, h], Alan Fried [e], Deborah C. Stein Zweers [d], Andrew J. Weinheimer [f], Russell R. Dickerson [a]

[a] Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD 20742, USA
[b] NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA
[c] Department of Earth Sciences, Millersville University, Millersville, PA 17551, USA
[d] KNMI-Royal Netherlands Meteorological Institute, De Bilt, The Netherlands
[e] Institute for Arctic & Alpine Research, The University of Colorado Boulder, CO 80309, USA
[f] National Center for Atmospheric Research, Boulder, CO 80307, USA
[g] Air Resources Laboratory, National Oceanic and Atmospheric Administration, College Park, MD 20740, USA
[h] Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD 20740, USA

Highlights

- Thermally direct circulations can strongly alter air quality near bodies of water.
- Large ozone concentration gradients could exist between water and its adjacent land.
- Bay/gulf breeze circulations of varying magnitudes have profound impacts on ozone.

ABSTRACT

Highly-resolved vertical profiles of ozone and reactive nitrogen in the lower troposphere were obtained using Millersville University's tethered balloon system and NASA's P-3B aircraft during the July 2011 Baltimore, MD/Washington DC and the September 2013 Houston, TX deployments of the NASA DISCOVER-AQ air quality field mission. The tethered balloon and surface measurement sites were located at Edgewood, MD and Smith Point, TX. The balloon profiles are used to connect aircraft data from the lowest portion of NASA's P-3B spirals (300 m AGL) to the surface thus creating complete profiles from the surface to 3e5 km AGL. The highest concentrations of surface ozone at these coastal sites resulted from mean flow transport of polluted air over an adjacent body of water followed by advection back over land several hours later, due to a bay or gulf breeze. Several meteorological processes including horizontal advection, vertical mixing, thermally direct circulation (i.e., bay, gulf, and, sea breezes) combined with chemical processes like photochemical production and deposition played a role in the local ozone maxima. Several small-scale, but highly polluted layers from the Chesapeake Bay advected landward over Edgewood, MD. The Houston Metro area was subject to large-scale recirculation of emissions from petrochemical sources by the Gulf of Mexico and Galveston Bay breezes.

1. Introduction

Boundary-layer ozone is a secondary photochemical pollutant formed by a reaction mechanism involving nitrogen oxides ($NO_x = NO + NO_2$), volatile organic compounds (VOCs), carbon monoxide (CO), and sunlight (UV radiation). Since ozone is harmful to both the human respiratory system and the photosynthetic processes of vegetation, the United States

Environmental Protection Agency (EPA) has implemented air quality standards for ozone as a criteria pollutant (Krupa and Manning, 1988; Burnett et al., 1997). Surface ozone is regulated according to the current primary National Ambient Air Quality Standard (NAAQS) of 70 parts per billion by volume (ppbv), calculated as the daily maximum of an 8-h running mean.

The concentration of ozone at or near the surface is also contingent upon meteorological conditions such as the synoptic scale circulation, boundary-layer height and turbulence, advection, incoming solar radiation, temperature, and humidity (Seaman and Michelson, 2000; Hegarty et al., 2007). Areas most commonly affected by high ozone concentrations are downwind of metropolitan centers. Additionally, coastal regions are frequently subject to poor air quality due to bay or sea breezes that can effectively recirculate pollution in the lower boundary layer (Banta et al., 2005; Loughner et al., 2011, 2014).

The same meteorological conditions that yield thermally direct circulations can also lead to ozone events in the right chemical regime: weak winds, warm temperatures, intense solar radiation, and subsidence inversions. Under these atmospheric conditions pollutants accumulate leading to ozone formation as well as allow these mesoscale circulations to compete with synoptic forcing. With pressure gradients in place, air near the surface moves from water to land during the day due to differential heating. The reversal of this occurs at night when the land cools much more quickly than water and causes a pressure gradient force in the opposite direction. This sequence forces early morning emissions over land to be transported over the adjacent body of water, and then re-circulated back to the land in the afternoon (Jacob, 2000; Wang et al., 2001).

Several studies have shown that sea, bay, and gulf breezes can contribute to poor air quality (Banta et al., 2005; Evtyugina et al., 2006; Darby et al., 2007; Loughner et al., 2011). The 2011 DISCOVER-AQ (Deriving Information on Surface conditions from Column and Vertically Resolved Observations Relevant to Air Quality) campaign yielded data demonstrating the influence of the Chesapeake Bay breeze as it enhanced pollution inland of the coastline (Stauffer et al., 2015; Stauffer and Thompson, 2015; Loughner et al., 2014). During the July 2011 DISCOVER-AQ campaign, the 2008 8-h ozone standard of 75 ppbv was violated at Edgewood, MD on ten days, and a bay breeze was observed on eight of these days (Stauffer et al., 2015). Studies during the 2011 DISCOVER-AQ campaign showed that concentrations of surface ozone tended to be higher over the Chesapeake Bay than upwind land areas due to a shallower boundary layer, ship emissions, lower deposition rates, higher photolysis rates, and decreased boundary layer venting due to a decrease in cloud cover compared to the nearby land (Goldberg et al., 2014).

Concentrations of background ozone in eastern Texas tend to be higher in late summer and early fall due to the synoptic circulations of northerly and easterly flow transporting continental high ozone air to the area. Higher background concentrations could contribute to the frequency and magnitude of ozone episodes (Langford et al., 2009). The Texas Commission on Environmental Quality (TCEQ) uses the background ozone concentration to estimate the local contribution of ozone as the difference between the 8-h maximum background ozone and the 8-h maximum measured ozone (Nielsen-Gammon et al., 2005). High ozone in the Houston area is often a result of small-scale circulations with advection of pollutants from the Houston Ship Channel to the southwestern part of the Houston Metro area (Ngan and Byun, 2011) and in many cases is the result of wind shifts in a postfrontal environment (Rappenglück et al., 2008). When a gulf or Galveston bay breeze sets up after these pollutants are advected over the water behind the front, the Houston Metro area can experience a second dose of pollution.

Studies performed in Houston, TX, showed that ozone episodes begin when the synoptic-scale winds transport pollutants from the land to water before a bay or gulf breeze sets up (Darby, 2005). As the bay or gulf breeze develops, pollutants are recirculated over the adjacent land adding to the pollution generated locally in these areas. Banta et al. (2005) discussed an ozone episode where the gulf/bay breeze contributed to surface hourly ozone concentrations of 200 ppbv.

Similarly in this paper, we focus on the effects of thermally direct circulations and local meteorology on air quality in Edgewood, MD and Smith Point, TX, as measured during DISCOVER-AQ.

2. The DISCOVER-AQ field project

It remains a challenge to accurately detect and resolve near surface pollution with Earth observations from space (Liu et al., 2005; Fishman et al., 2008; Martin, 2008; Chatfield and Esswein, 2012). DISCOVER-AQ, a five-year NASA Earth Venture campaign, was designed to advance satellite observation capabilities by investigating the relationship between column-integrated trace gas quantities and pollution in the near-surface environment (http://discover-aq.larc.nasa.gov). Goals of DISCOVER-AQ include assessing uncertainties in column, surface trace gas, and aerosol observation correlations, characterizing the diurnal variation of the column and surface observations, and to investigate how much horizontal variability can be captured in satellite retrievals and model calculations. The P-3B aircraft provided profiling of meteorological, trace gas, and aerosol variables centered over surface air quality sites.

The ability to understand and predict air pollution events has been limited in part by the lack of vertical meteorological and chemical profile observations. With this unprecedented DISCOVERAQ data set in terms of horizontal, vertical and temporal coverage, the spatial-temporal variability of air pollution can be better addressed.

2.1. P-3B aircraft measurements

NASA's P-3B aircraft typically spiraled over each ground site three to four times within an operational day at altitudes from 300 to >3000 m AGL, and did not measure air pollutants near the surface. However in some DISCOVER-AQ deployments, missed approaches were used to fill this gap between 300 m and the surface. In the Maryland deployment, the tethered balloon at Edgewood is used in this study. In the Houston deployment, the Millersville University tethered balloon was used at Smith Point. Onboard the P-3B there was continuous ozone, NO, NO_2, and NO_y measurements made using the National Center for Atmospheric Research (NCAR) 4-Channel Chemiluminescence Instrument with 1-s averages with 5% uncertainty for ozone and NO, 10% for NO_2, and 20% for NO_y. CH_2O measurements were made on the P-3B using the Difference Frequency Generation Absorption Spectrometer (DFGAS) with 30 s averaging and 13% uncertainty (Weibring et al., 2007).

Some differences were observed between the P-3B measurements and those of the tethersonde. These differences are likely due to the horizontal distance between the aircraft and the balloon along a convoluted coastline near the Edgewood site, and/or the timing between the flyover and the tethered balloon position. Based on inter-comparisons between the P-3B and the tethersonde, differences due to representativeness (timing and exact location) are likely greater than differences associated with instrument errors or operations. The timing between the flyover and when the tethered balloon reached the aircraft altitude was sometimes as different as 30 min.

2.2. Ground sites for tethersonde deployment

2.2.1. Edgewood, Maryland

The Baltimore/Washington metropolitan area is vulnerable to exceeding the 70 ppbv; 2015 (75 ppbv; 2008) EPA ozone standard due to the abundance of precursor emissions along with meteorological conditions favorable for ozone production (He et al., 2013). The highest ozone design value in the Baltimore Non-Attainment Area (NAA) has been consistently measured at the air-monitoring site in Edgewood. This site experienced the highest ozone measured on the U. S. east coast region for 2011 and was many times the only monitoring station within the NAA that exceeded the ozone standard of 75 ppbv standard at that time. This is due in part to its location in a bay breeze convergence zone.

2.2.2. Smith Point, Texas

Houston, TX has large emissions of ozone precursors from power plants, refineries, and petrochemical industrial plants coupled with meteorological conditions favorable for ozone production, typically during late summer. Emissions are particularly large along the Ship Channel and western shore of Galveston Bay (Banta et al., 2005). Aircraft observations from Kleinman et al. (2005) found that NOx and light olefins emitted from petrochemical facilities led to the highest ozone production observed in the study. Smith Point, on a peninsula extending into Galveston bay from the eastern shore, is susceptible to both bay and gulf breeze pollution recirculation. The combination of high emissions and gulf and bay breeze circulations lead to ozone exceedances.

2.3. Methods

2.3.1. Edgewood measurements (Table 1)

Millersville University deployed its mobile lab including a suite of instruments and equipment in support of boundary layer and atmospheric chemistry research (Millersville Atmospheric Research and Aerostat Facility (MARAF; see http://www.millersville.edu/esci/maraf). One hundred-sixty seven tethered balloon soundings captured the temporal and vertical evolution of ozone on P-3B flight days and some non-flight days throughout the campaign. The continuous profiles provide a useful data set to characterize profile shapes and how they vary as a result of meteorological conditions such as bay breezes, the amount of boundary layer turbulence, and influences of local plumes versus longer range transport. MARAF was deployed at Eagle Point on the Edgewood side of the Aberdeen Proving Ground (APG; lat: 39.4°, lon: -76.3°) for the first DISCOVER-AQ deployment in July 2011 (Fig. S1). APG, a U.S. Army facility, is often influenced by transport of ozone precursors from the Baltimore-Washington Metro area. MARAF includes a 4-m flux tower, a Sigma Space MicroPulse Lidar (MPL), an acoustic Sonic Detection And Ranging (SODAR) with Radio Acoustic Sounding System (RASS) extension, surface trace gases (O_3, NO_x, SO_2, and CO), and a 3-wavelength Nephelometer. The surface instruments integrate with the measurements obtained by the tethered balloon. The tethered balloon system consists of a Vaisala TTS111 system that measures temperature, pressure, relative humidity, wind speed, and wind direction along with a 2Btechnologies Inc. trace gas analyzer for ozone. Semi-continuous profile measurements were taken in blocks of approximately 1.5-2.5 h depending on available platform battery power, where

typically four vertical profiles were measured per charge. The soundings coincided in time with the P-3B spirals to fill the gap from the lowest P-3B altitude to the surface. Profiles were also conducted between spirals to capture the temporal evolution of vertical variability throughout the day.

The MARAF site was set up 2.7 km SE of the Edgewood Maryland Department of the Environment monitoring site (MDE) as well as the Nittany Atmospheric Trailer and Integrated Validation Experiment (NATIVE; Martins et al., 2012) for optimal boundary layer sampling immediately on the coast of the bay (Fig. S2). MDE and NATIVE were collocated platforms for air quality and ground-based in-situ measurements. Chemical measurements included O_3, NO, NO_y, SO_2, and CO for NATIVE and O_3 for MDE.

2.3.2. Smith Point measurements (Table 1)

MARAF was deployed at Smith Point, TX (lat: 29.5°, lon:-94.8°) for the third deployment of DISCOVER-AQ in September 2013 (Fig. S3) alongside the NATIVE trailer. The tethered balloon operation was similar to the Edgewood deployment, but the 2B-Technologies Inc. NO_x analyzer and an NO_2 sonde developed by the Royal Netherlands Meteorological Institute (KNMI) were added for this deployment. The KNMI NO_2-sonde uses the NO_2 chemiluminescent reaction in a nearly specific to NO_2 aqueous luminol solution (Sluis et al., 2010). Similar surface instruments to the Maryland deployment were used such as the MPL, SODAR, Nephelometer, and trace gas suite; however, the flux tower experienced technical issues and was not used for this study. A Coastal Environmental WeatherPak 2000 was used in this deployment, which measured surface meteorological constituents such as: pressure, temperature, humidity, wind speed, and wind direction. Again for this campaign, the NATIVE mobile platform for air quality and ground-based insitu measurements were used which included surface chemical measurements of O_3, NO, NO_y, SO_2, and CO.

3. Bay and gulf breeze case studies

3.1. Bay breeze Edgewood, MD 29 July 2011

During the Baltimore/Washington DISCOVER-AQ campaign, five days exhibited a bay breeze and four days displayed evidence of bay breeze initiation, but were unable to persist due to a thunderstorm or gust front. During this deployment, ten days exceeded the EPA 8- h 2011 ozone standard of 75 ppbv at Edgewood - eight of ten were associated with a bay breeze or "interrupted" bay breeze (Stauffer et al., 2015). On 29 July 2011, surface (Fig. 1), and tethersonde observations indicated three bay breeze fronts at the Edgewood MARAF site. The morning of the 29 July featured weak synoptic forcing with a surface high pressure and an upper-level ridge in place over the Mid-Atlantic region, typically conducive for both ozone events and bay breezes. By 12 UTC (08:00 EDT; UTC-4), the synoptic wind pattern over the site was northwesterly (NW) at 850 mb (Fig. S4; NCEP Reanalysis data provided by the NOAA/OAR/ ESRL PSD, Boulder, Colorado, USA, from their website at http://www.esrl.noaa.gov/psd/).

The earliest bay breeze passage of the day (~13:30 EDT) was noted by both the surface observations and a tethersonde profile to be a shallow and brief, but intense boundary between the land environment and the high ozone air over the bay. Measurements made at the NATIVE trailer only 2.7 km NW of MARAF did not exhibit any effects of the first bay breeze event (Stauffer et al., 2015). A second bay breeze front passed through the site around 16:15 EDT

affecting surface concentrations for about an hour until a wind direction shift brought cleaner air to the research site. By 18:00 EDT, a third bay breeze passage was measured which was sustained until 20:00 EDT when a gust front pushed the ozone rich marine air off the coast bringing in cleaner continental air from the thunderstorm outflow. Continuous profiling by the tethersonde captured much of the variability observed on this day. Measurements at the NATIVE trailer were also affected by the second and third bay breeze passages with slightly smaller magnitude fluctuations (Stauffer et al., 2015).

July 29, 2011 was part of a multi-day ramp up of summertime air pollution as a consequence of the synoptic meteorological conditions. Around 08:30 EDT, a combination of stored ozone from the (nocturnal) residual layer mixing down to the surface during the growth of the mixed layer and photochemical production led to a rapid increase in surface ozone from 25 to 75 ppbv within 2 h (Fig. 1). This corresponds to negative vertical eddy momentum flux (downward transport) as measured by the flux tower, as well an end to directional wind shear and development of vertical speed shear (Fig. 2). Tethered balloon soundings between 08:20 and 09:07 EDT (Fig. 3) show enhanced ozone concentrations relative to the surface between 150 and 340m. The ascending profile (08:20e08:42 EDT) from the surface to 340 m shows increasing ozone concentration with altitude between 150 and 330 m. The descending profile (08:42e09:07 EDT) shows the downward transport of higher ozone concentrations from aloft to the layer below 200 m. The descending profile indicates a layer (260e340 m) of lower water vapor mixing ratio along with higher potential temperature and higher wind speeds than the ascent sounding. From 260 m to the surface, the descending profile shows fairly well mixed, enhanced water vapor mixing ratio, higher potential temperature, and higher wind speeds than the ascending profile. Back trajectory analysis calculated for the period 6 h prior to these profiles from the Air Resources Laboratory's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Hess, 2004) shows transport from central Pennsylvania at 1000 m and western Pennsylvania at 100 m. Back trajectories from the period 1 h before are from eastern Maryland at 1000 m and between Edgewood and Baltimore intersecting I-83 at 100 m. This suggests that some of the enhanced ozone concentration found in these profiles was due to transport from Pennsylvania cities and Baltimore, MD during early to mid morning, before the bay breeze.

Ozone concentrations at the surface fluctuated from 75 to 80 ppbv from 11:00 EDT until the first bay breeze passage (~13:30 EDT) that swept through the site in a shallow wedge (<100 m) bringing spikes in ozone and specific humidity, but a drop in temperature. The combined P-3B and tethersonde profile (Fig. 4) for the first spiral of the day demonstrates the shallow but intense bay breeze below the P-3B minimum flight altitude (Fig. 5; Table 2). The bay breeze also transported other trace gases during this passage: NOx increased from 6.5 to 8.8 ppbv, SO2 increased from 2.7 to 3.6 ppbv, and CO increased from 710 to 760 ppbv (Fig. 1). This bay breeze was too brief for the 30 min averaged SODAR wind measurements, however, the tethersonde anemometer measured wind direction in the bay breeze layer to be between 90 and 190°, but fluctuating between 80 and 280° near the surface (Fig. S5; wind direction light pink sounding). At 13:50 EDT, trace gas concentrations, specific humidity, and temperature returned to previous levels throughout the vertical profile (Fig. S5; green sounding) and at the surface (Fig. 5). The brevity of this intense yet shallow bay breeze demonstrates the steep, localized gradients at the bay breeze front and the significant impact on surface concentration.

A second bay breeze frontal passage occurred later in the day around 16:15 EDT (Fig. 5; Table 2). This was associated with a wind shift from NW to SSW with air coming from the

Baltimore area and passing over the bay. By 17:25 EDT the wind shifted direction again from SSW to SSE along with a change in surface concentrations. Although this air temporarily passed over the bay, specific humidity decreased to 14.6 g/kg, temperature increased to 38.2° C, and ozone decreased to 85.3 ppbv (Fig. 5). A combined P-3B and tethersonde profile was captured during this transition period between wind directional shifts. The tethersonde profile from 16:48 to 16:57 EDT captured the ozone and water vapor rich air mass from the bay breeze, whereas the P-3B spiraled down 25 min later (from 17:24 to 17:36 EDT) over the site measuring the air from the SSE with the previously detailed lower ozone and specific humidity concentrations along with slightly warmer temperatures (Fig. 6).

By 18:00 EDT, the bay breeze returned and remained for ~2 h (Fig. 5; Table 2). Around 20:00 EDT, the bay breeze was terminated by NW flow from a gust front heading southeastward from southern Pennsylvania/northern Maryland as shown by the Sterling, VA radar (KLWX) in Fig. S6. With the passage of the gust front, specific humidity decreased from 18.4 to 13.4 g/kg, temperature decreased from 36.4 to 35.6° C, and ozone concentration dropped from 88 to 67 ppbv (Fig. 5).

3.2. Bay and gulf breeze, Smith Point, TX 25 September 2013

The effects of local meteorology on this day resulted in the highest instantaneous measured ozone during all of the DISCOVERAQ deployments. Leading up to 25 September 2013, and for most of the DISCOVER-AQ Houston deployment, onshore flow dominated at Smith Point. The 25 September was not part of a ramp up pollution episode or heat wave common to high pollution case study events, but instead, the result of postfrontal and local wind shifts carrying polluted air masses. On this day, the local winds behind the front were northerly and pollution observed at Smith Point was largely, a function of the flow from Houston industrial area and chemical plants. The localized Houston pollution that accumulated over the bay and gulf was recirculated back over the research site at Smith Point by the gulf and bay breeze around 17:30 CDT (UTC-5) with concentrations of 175 ppbv of ozone observed at the surface at Smith Point.

3.2.1. Synoptic conditions, local winds, and air quality observation overview

In the early hours of 25 September 2013, a weak cold front stemming from a low over NW Arkansas moved SE over the Gulf of Mexico. High pressure filled in behind this front and a ridge was in place over much of the south-central United States by 12 UTC (07:00 CDT) featuring subsidence over southeast Texas (Fig. S7). At 10:00 CDT, SODAR (Fig. 7; colored background) detected a near-surface wind shift from WSW to N bringing higher concentrations of NO_y to Smith Point. Refineries and chemical plants in Baytown and Deer Park lie to the NW and NNW of Smith Point. While ozone was increasing since 7:00 CDT, an abrupt jump in ozone concentration was observed at the surface around 11:00 CDT, most likely due to mixing down of higher concentrations of ozone and precursors. From 12:00 to 15:00 CDT, surface winds were NNE while winds at 130 m-200 m were NNW and NW. The NNW and NW winds were associated with the transport of ozone and ozone precursors to Smith Point as shown by the tethered balloon profiles in Fig. S8. These profiles, which started at 13:14 CDT, exhibit peaks in ozone concentration of 220 ppbv and 200 ppbv with NO_y concentration of 18 ppbv between 100 and 200 m with low NO2 concentration.

Ozone continued to increase at the surface under northwesterly winds until easterly winds at 16:00 CDT brought a brief respite until 17:00 CDT. At this time, gulf and bay breezes brought the poorest air quality of the campaign - ozone concentrations at the surface soared from 70 ppbv to 175 ppbv. By 20:00 CDT, ozone concentrations retreated between 60 and 70 ppbv (Fig. 7).

3.2.2. Transport to Smith Point from major sources

During the first P-3B circuit of the day at 9:48 CDT, the aircraft flew over the largest petrochemical facility in the U.S. near Baytown (29.741, -95.010), and formaldehyde (CH_2O) concentrations rose dramatically (Fried et al. AQRP report, 2016) to 18-20 ppbv. CO concentrations were between 500 and 600 ppbv and Noy concentrations were between 45 and 50 ppbv (Fig. 8; concentrations located near the white square).

As the P-3B made its closest approach near Deer Park (29.703, -95.131) during the first circuit around 11:21 CDT, CH_2O concentrations were between 8 and 12 ppbv; CO concentrations were between 480 and 520 ppbv and NO_y concentrations between 55 and 60 ppbv (Fig. 8; concentrations near the black square).

During the second circuit (12:15-12:28 CDT), a polluted air parcel from the Baytown and Deer Park area was observed downwind, over the Galveston Bay and at Smith Point (Fig. 9). CH_2O concentrations were between 20 and 23 ppbv, CO concentrations were 300-400 ppbv, NO_y between 0 and 5 ppbv, and ozone 110-145 ppbv. The combined P-3B and tethersonde profile during this spiral shows an elevated layer of ozone between 400 m. Due to small differences in time and space between the P-3B and the tethersonde, the P-3B measured a NO_y plume at 100 m with an associated decrease in ozone where the balloon did not. This is likely due to NO titration from a local emission source (Fig. 10). The tethered balloon continued to profile between the second and third P-3B overpasses. In the next set of tethersonde balloon profiles from 12:56 to 13:59 CDT, the highest concentration of ozone was observed within the first 500 m at Smith Point due to photochemistry from significantly elevated precursors emitted upwind (Fig. 11).

Six-hour backward trajectories at 1 km horizontal resolution (WRF; Skamarock et al., 2008) were run at six initialization altitudes relevant to the tethersonde at Smith Point (2, 100, 200, 300, 400, and 500 m). The trajectory and wind directions agree with measured surface wind direction with variations in near-surface vertical wind shear observed by SODAR. The back trajectory from the first profile, which started at 12:56 CDT (Fig.12a), shows that air passed over the Deer Park plants between 200 and 300 m altitude. This corresponds to the layer of highest ozone concentration from the first sounding in Fig. 11 (red) in the layer between 125 m and 275 m. At 13:14 CDT, the sounding in Fig. 11 (green) shows ozone concentrations of 220 ppbv between 100 and 200maltitude, which correspond to air coming from the facilities near Baytown at 100 m and air from the Deer Park Plants at 200 m according to the WRF back trajectory (Fig. 12b). The sounding at 13:31 CDT Fig. 11 (dark blue) showed that ozone concentration retreated to mostly below 150 ppbv from 150 m to 500 m and increased to 180 ppbv in a shallow layer between 25 m and 75 m. By 13:49 CDT, the entire tethersonde profile retreated to ozone concentrations below 150 ppbv from the surface to 500 m (Fig. 11; purple sounding).

3.2.3. Recirculation from bay and gulf breeze

Northwesterly winds throughout the day transported pollutants offshore over the Galveston Bay and the Gulf of Mexico where secondary pollutants formed; these later returned to Smith Point in the strongest episode of the campaign. An intense, ozone-rich, shallow layer

only 200 m deep was observed in the vertical balloon soundings between 14:31-15:42 CDT due to the static stability of the air over the relatively cool surface of the bay. In this series of balloon soundings, the shallow marine boundary layer was diluted and warmed by mixing with free tropospheric air from aloft (Fig. 13a). Around 15:00 CDT, a negatively buoyant thermal that overshot its neutral level fell down back into the mixed layer bringing with it drier free tropospheric air. This was also associated with a wind directional change from NNE to NNW at the surface. A combination of this warmer, drier air that entrained into the boundary layer as well as the easterly wind shift observed around 16:00 CDT diluted the amount of ozone and water vapor observed within the first 200 m above the surface (Fig. 13b).

By 17:30 CDT, the gulf and bay breezes made their way over Smith Point (Fig. 14). At the surface, ozone concentration rose from 80 to 175 ppbv during the bay breeze passage (Fig. 7). Water vapor mixing ratio increased from 9 to 18 g/kg, temperature decreased from 36 to 32° C, and wind direction shifted from E to SW during the passage of the bay breeze between 17:28 to 18:00 CDT (Fig. 15). This stagnant, ozone-rich air at the edge of the gulf breeze and bay breeze front acted as a convergence zone for ascent of air with ozone concentrations of 120 ppbv from the surface to 100mand up to 150 ppbv at 100 m-320 m by 17:49 as observed by the tethersonde soundings. By 18:25 CDT, ozone concentrations of 120 ppbv were observed within the layer between 200 and 360 m while the surface concentration retreated to 90 ppbv (Fig. 13c).

4. Discussion

Microscale and mesoscale meteorological processes are essential to understanding and forecasting the dispersion of background and local pollution through growing boundary-layer/frontal mixing and bay/gulf breezes. Shown here are case studies where high ozone events were directly influenced by boundary-layer dynamics and recirculation of the thermally direct circulations. While the theme for coastal pollution recirculation is the same for each case e pollutants are transported out to the adjacent body of water where concentrations increase and are transported back over the land when the bay or gulf breeze forms, specific and important differences remain between these two cases.

The case study in Edgewood, MD (29 July 2011) demonstrates both vertical mixing and bay breeze phenomena. Entrainment shortly after sunrise led to an increase in surface ozone from around 25 to 75 ppbv within 2 h. Back trajectories show that ozone aloft originated over western and central Pennsylvania 6 h earlier, and from near Baltimore 1 h before. Later that day, the bay breeze reached the Edgewood site first as a shallow, short-lived (10 min) burst of ozone above 100 ppbv, reformed several hours later as a smaller bay breeze, and then reformed once again as a larger-scale incursion lasting roughly 2 h. Ozone concentration remained above 85 ppbv for 4 h after from the second and third bay breezes. In all bay breeze events, the air showed meteorological characteristics of having been over the Chesapeake Bay - lower temperatures and higher humidity. These air parcels also showed chemical signatures characteristic of reduced venting - higher concentrations of primary pollutants CO and NOx as well as ozone, where the latter could be the result of faster photochemistry over the cloud-free bay. These bay breezes were frequent, but not strong enough to penetrate inland to areas not directly influenced by the coast. Edgewood's coastal location, at a convergence zone with specific mesoscale dynamics, is a key factor for unique pollution episodes not seen at other MDE monitoring stations.

The case study at Smith Point, TX demonstrated a broad mid-to late day event with ozone in excess of 80 ppbv for 5 h when NW winds aloft brought pollution from the Port of Houston

area including petrochemical plants that mixed to the surface at Smith Point. Around 17:30 CDT, the winds shifted dramatically to the south/southwest bringing air from over the gulf and the Galveston Bay to the MARAF and NATIVE site with concentrations that exceeded 125 ppbv at the surface for over an hour.

For this case, light to calm winds throughout most of the afternoon allowed the pollution to stagnate and build up over Houston, Galveston Bay and the gulf where active photochemistry occurred for an extended period of time. Concentrations observed at Smith Point were mostly due to the transport of high pollution from the chemical plants and the Houston Metro area. A major shift in wind as the bay and gulf breeze developed in the early evening resulted in a huge impact on pollution at the surface and affecting a broad horizontal extent.

In both cases, the marine boundary layer was shallow, resulting in the buildup of ozone concentrations confined to a wedge close to the surface as it passed over land. Edgewood experienced an extremely shallow bay breeze <100 m deep with ozone surface concentrations of 113 ppbv and 75 ppbv around 100 m. Conditions measured at Smith Point were also representative of a shallow marine boundary layer, however, this was not part of the bay breeze passage that occurred later in the day. Smith Point, on a small peninsula in Galveston Bay, is susceptible to marine-like conditions without much forcing from specific bay breezes. The profiles that captured the conditions of the marine boundary layer over Smith Point measured a shallow layer <200 m deep with ozone surface concentrations 132 and 70 ppbv around 200 m. This further demonstrates the buildup of ozone concentration over the bay due to slower deposition rates, higher photolysis rates, and trapping of emissions over the bay, as discussed by Goldberg et al. (2014).

Differences between Edgewood and Smith Point arise from the locations relative to large bodies of water, types of emissions from urban centers, and regional buildup of background concentrations over several days vs. a quick burst of local emissions. Detailed case studies as described here are important for determining meteorological conditions and relevant scales (i.e., synoptic, mesoscale, and microscale) of pollution episodes. This knowledge can lead to better air quality prediction. Most regional atmospheric chemistry models, such as the Community Multiscale Air Quality model (CMAQ) in regulatory or forecasting operational mode, are run at 12-km resolution and have difficulty resolving some of the drivers of the largest pollution episodes such as vertical transport and horizontal gradients near coastal sites. Higher resolution (<4 km) is required to resolve bay/gulf breezes (Loughner et al., 2011). However, CMAQ run at 1 km for both Edgewood and Smith Point were unable to capture the high ozone transport due to the multiple bay breeze circulations from the Chesapeake Bay at Edgewood and the single, strong bay breeze from the Galveston Bay at Smith Point. Despite the inability to produce effects of the bay breeze at Smith Point in CMAQ largely due to the low emissions bias upwind of this location, the model was able to accurately represent the gulf breeze over the immediate Houston area during this day's event. This could be due to the much larger spatial extent along and across the gulf breeze frontal boundary. Thus, accurate representation of these phenomena can sometimes be difficult even at fine spatial resolution (1 km) depending on frequency, duration, biases in upwind emissions, and horizontal/vertical extent of the bay breeze event.

5. Conclusions

The tethersonde is a powerful tool for studying composition and circulation in the lowest few hundred meters of the atmosphere where air can be quickly mixed vertically, where

pollutants have the greatest impact on human health, and where aircraft measurements may be restricted.

We present two case studies that exemplify the importance of mesoscale and microscale meteorological processes on air quality. Large concentration gradients can exist both vertically and horizontally due to small-scale meteorological features that are difficult to accurately predict. In the absence of perfect emissions inventories, a detailed model characterization of the flow at high resolution may be the only way to successfully resolve high pollution events in locations subject to bay/gulf breezes. In each case, the driving mechanism for the observed pollution episode is the coupling of chemistry and small-scale meteorological features. These coupled processes include:

- Mixing down of polluted air from the residual layer air.
- A shallow marine boundary layer trapping emissions.
- Localized wind shifts.
- Recirculation of pollution from the meso-high set up over the bay/gulf and meso-low set up over the adjacent land.

The effects on air quality by thermally direct circulations are important to consider when analyzing data from monitoring stations susceptible to marine influences. While monitoring stations close to the bay or gulf will help characterize the marine effects, they may not be representative of the average concentrations over an adjacent metropolitan area. Many of the world's large cities are located near major bodies of water. For those cities with coastline configurations similar to those near Baltimore/Washington and Houston Metro, the results presented here may be helpful in understanding the circulation and causes of severe pollution events in those other cities.

Acknowledgements

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Figures

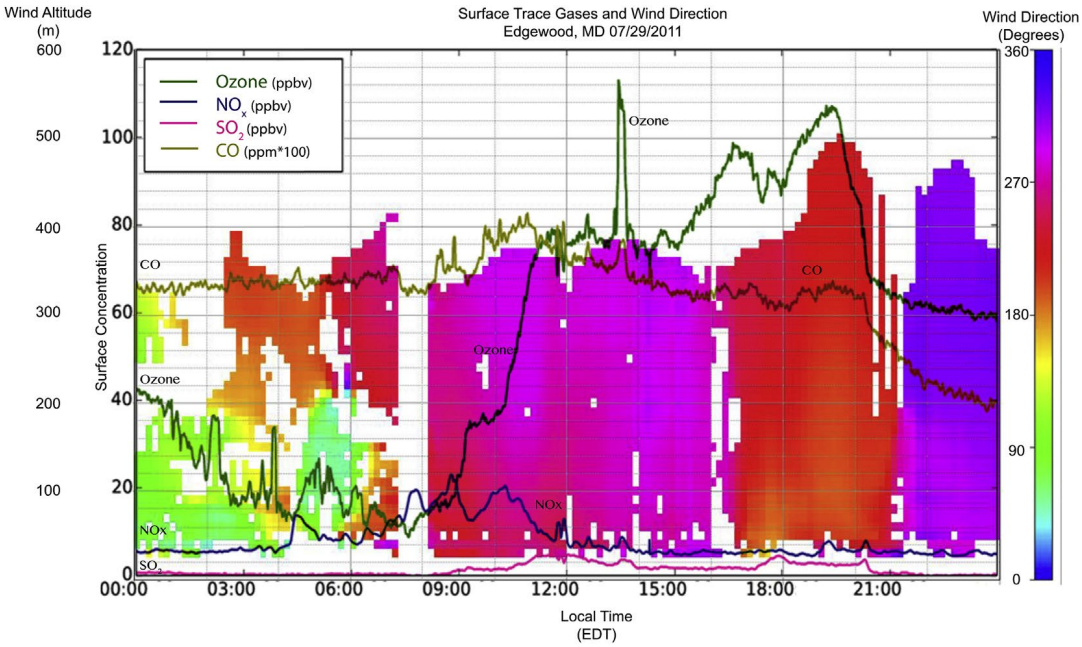


Fig. 1. 07/29/2011 Edgewood MARAF site wind direction with height derived from SODAR wind profiler (colors) and labeled surface trace gases: ozone (dark green), NO_x (blue), SO_2 (pink), and CO (olive green). Note WSW winds starting at 8:30 EDT as the nocturnal PBL broke up and concentrations of primary pollutants CO and NO_x increased. This is followed by inflow of more ozone rich air from over the Chesapeake Bay in a shallow (~100 m; see also Figs. 2 and 3) layer, shifting to generally SSW winds with sustained high O_3 concentrations by 16:30 EDT. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

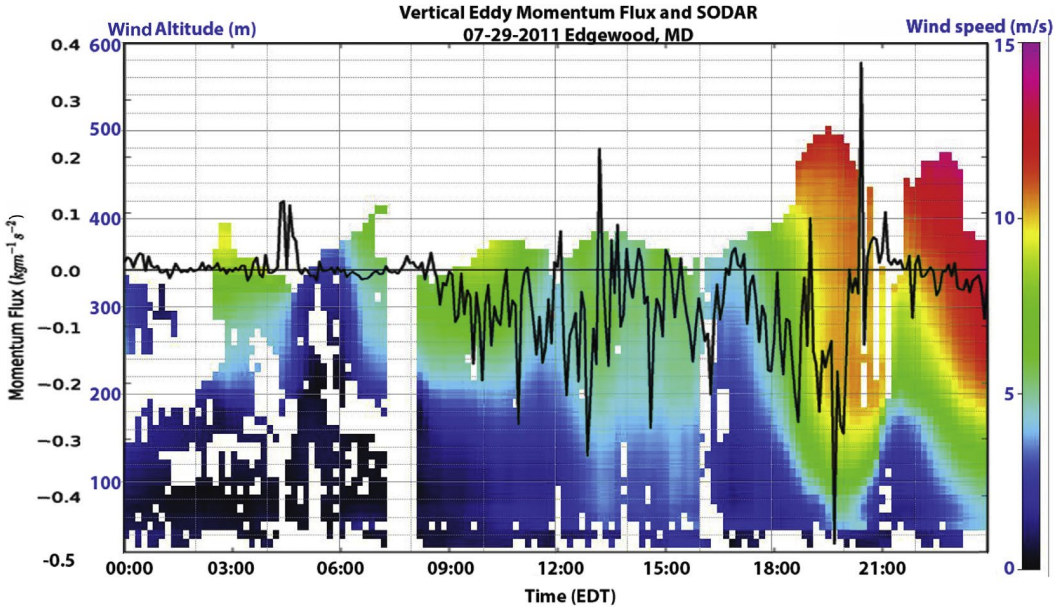


Fig. 2. 07/29/2011 Edgewood MARAF site wind speed with height derived from SODAR (colors) and 4-m vertical eddy momentum flux (black line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

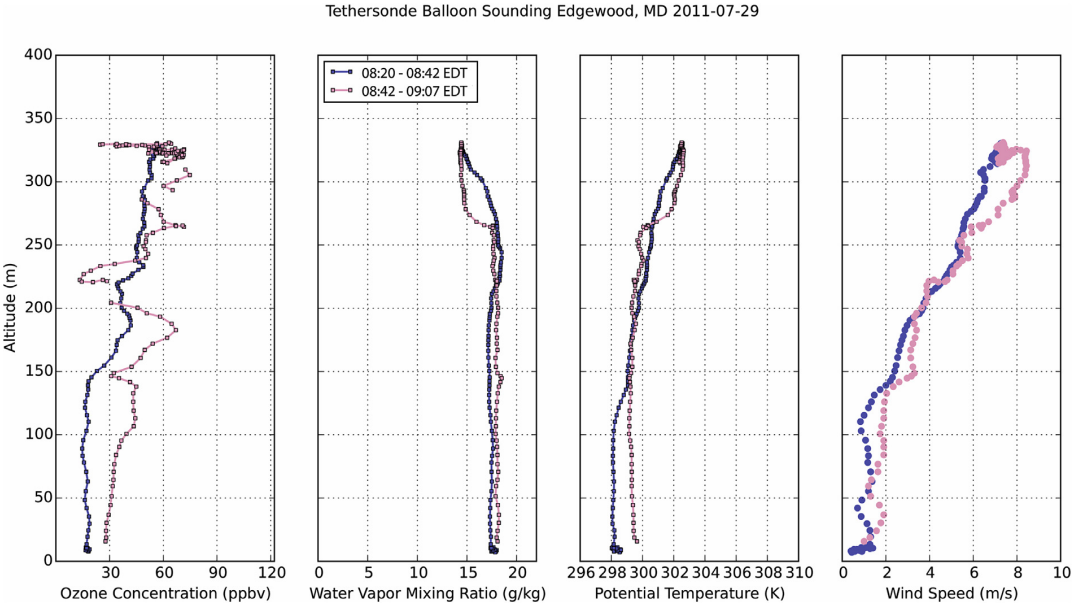


Fig. 3. 07/29/2011 Millersville tethersonde profiles of ozone concentration, water vapor mixing ratio, potential temperature, and wind speed from the surface to ~340 m. The first sounding (blue) is from the surface to maximum altitude (08:20-08:42 EDT) and the second sounding (pink) is down from maximum altitude (08:42-09:07 EDT). Profiles indicate higher ozone concentration and water vapor mixing ratio layer aloft during first sounding and mixing down (weaker vertical gradient) by the second sounding. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

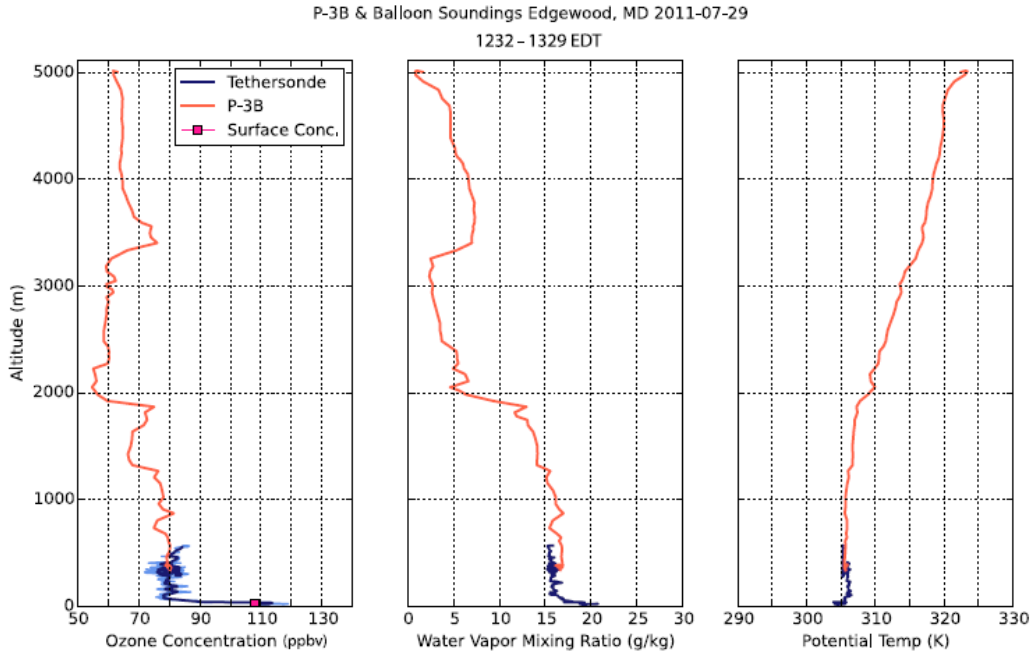


Fig. 4. 07/29/2011 Midday blended profile of the first circuit P-3B spiral over Edgewood (orange) and the corresponding Millersville tethersonde sounding (blue) of ozone concentration, water vapor mixing ratio, and potential temperature from the surface to ~5000 m; surface ozone concentration (pink dot; at maximum surface ozone concentration). The shallow bay breeze passage is observed in the tethersonde profile and the surface, but not by the P-3B due its extremely shallow depth. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

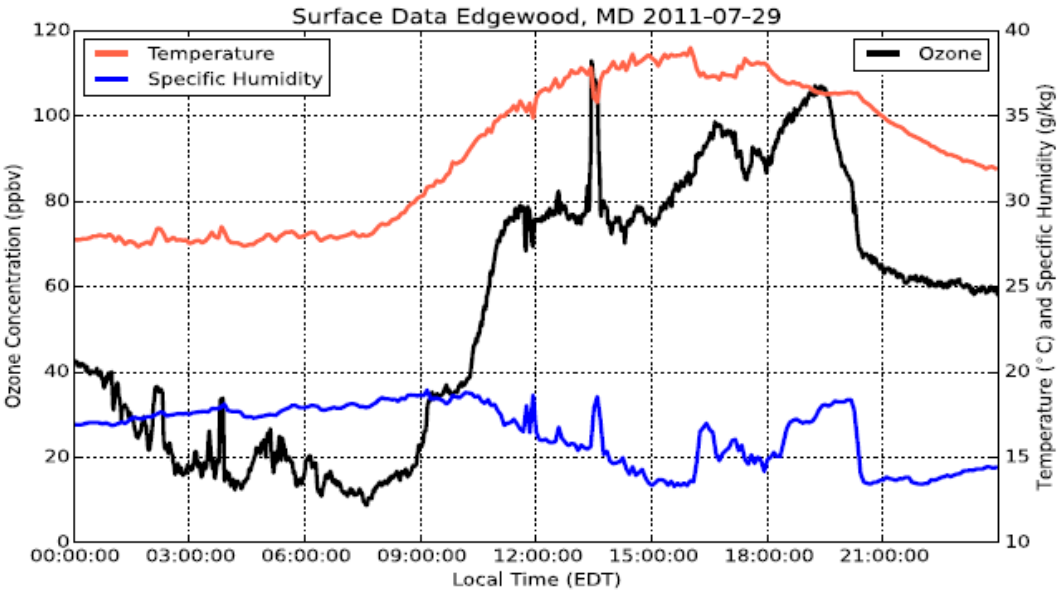


Fig. 5. 07/29/2011 MARAF surface ozone concentration (black line), 4-m temperature (orange line) and 4-m flux tower specific humidity (blue line). Spikes that positively correlate between ozone concentration and specific humidity and negatively correlate to temperature observed (13:30, 16:15, 18:00 EDT) indicate two small-scale bay breeze passages and then a larger scale passage from 16:00 to 19:00 EDT. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

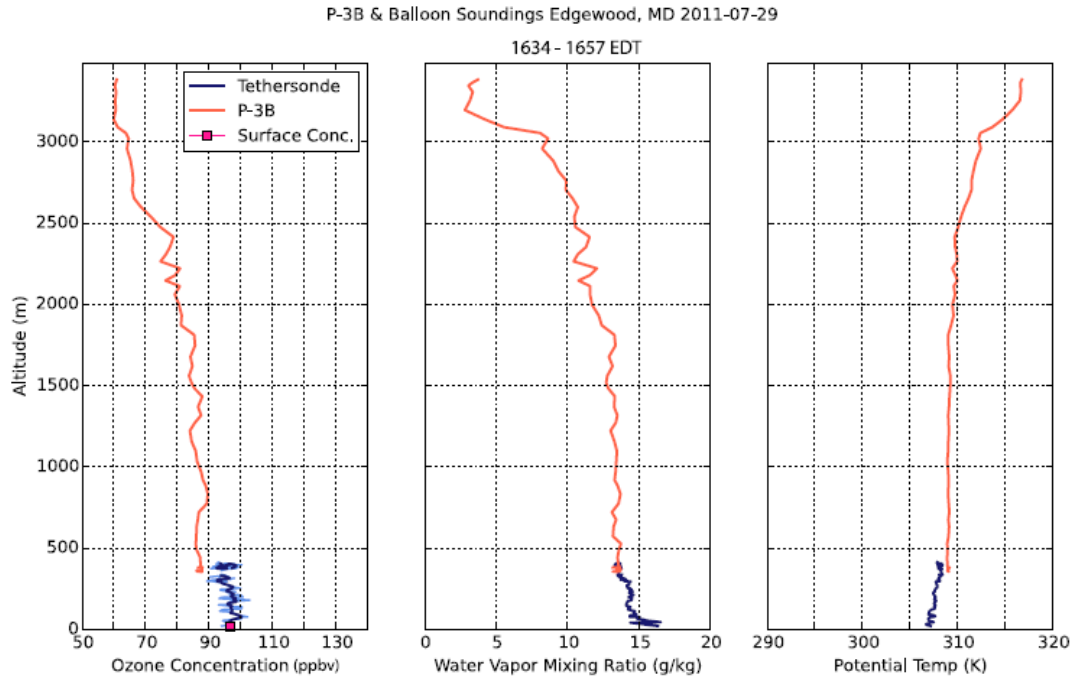


Fig. 6. 07/29/2011 Late afternoon blended profile of the third circuit P-3B spiral over Edgewood (orange) and the corresponding Millersville tethersonde sounding (blue) of ozone concentration, water vapor mixing ratio, and potential temperature from the surface to ~4000 m; surface ozone concentration (pink dot). The tethersonde profile was taken ~25 min before the P-3B spiral, resulting in somewhat greater disparity between platforms. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

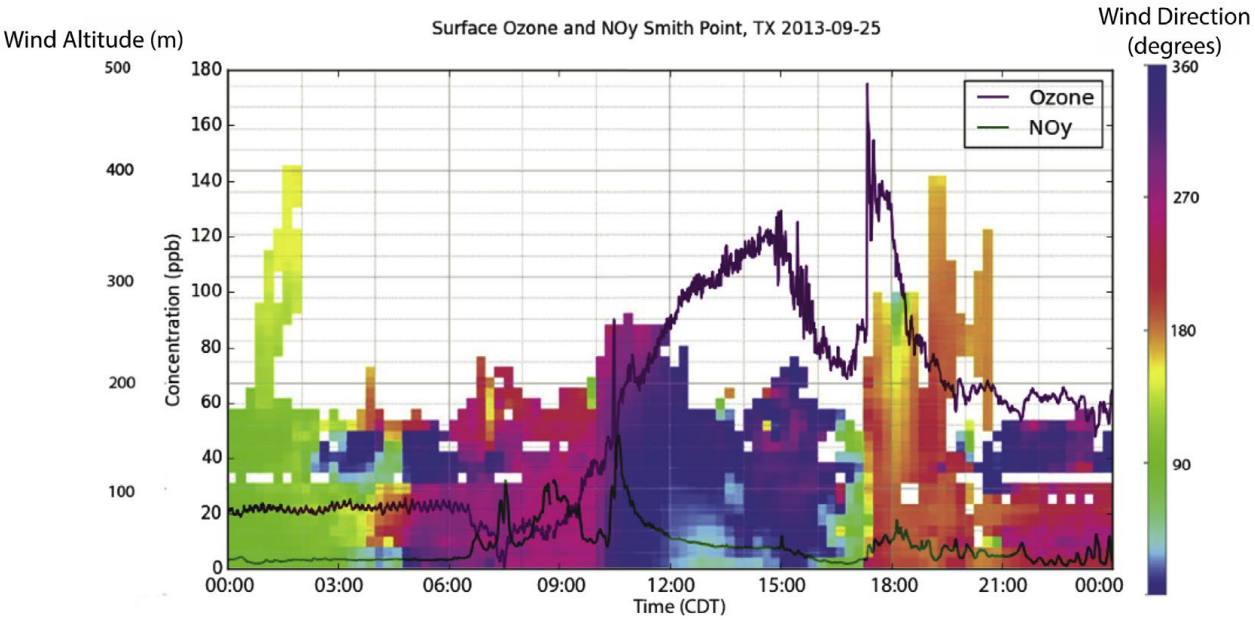


Fig. 7. 09/25/2013 Smith Point, TX MARAF site wind direction with height derived from SODAR (colors) and NATIVE surface trace gases: ozone (purple) and NO_y (green). Note: consistent buildup of ozone under NW winds was followed by a spike as winds shifted to SW around 17:00 EDT, which brought photochemically aged smog to the site. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

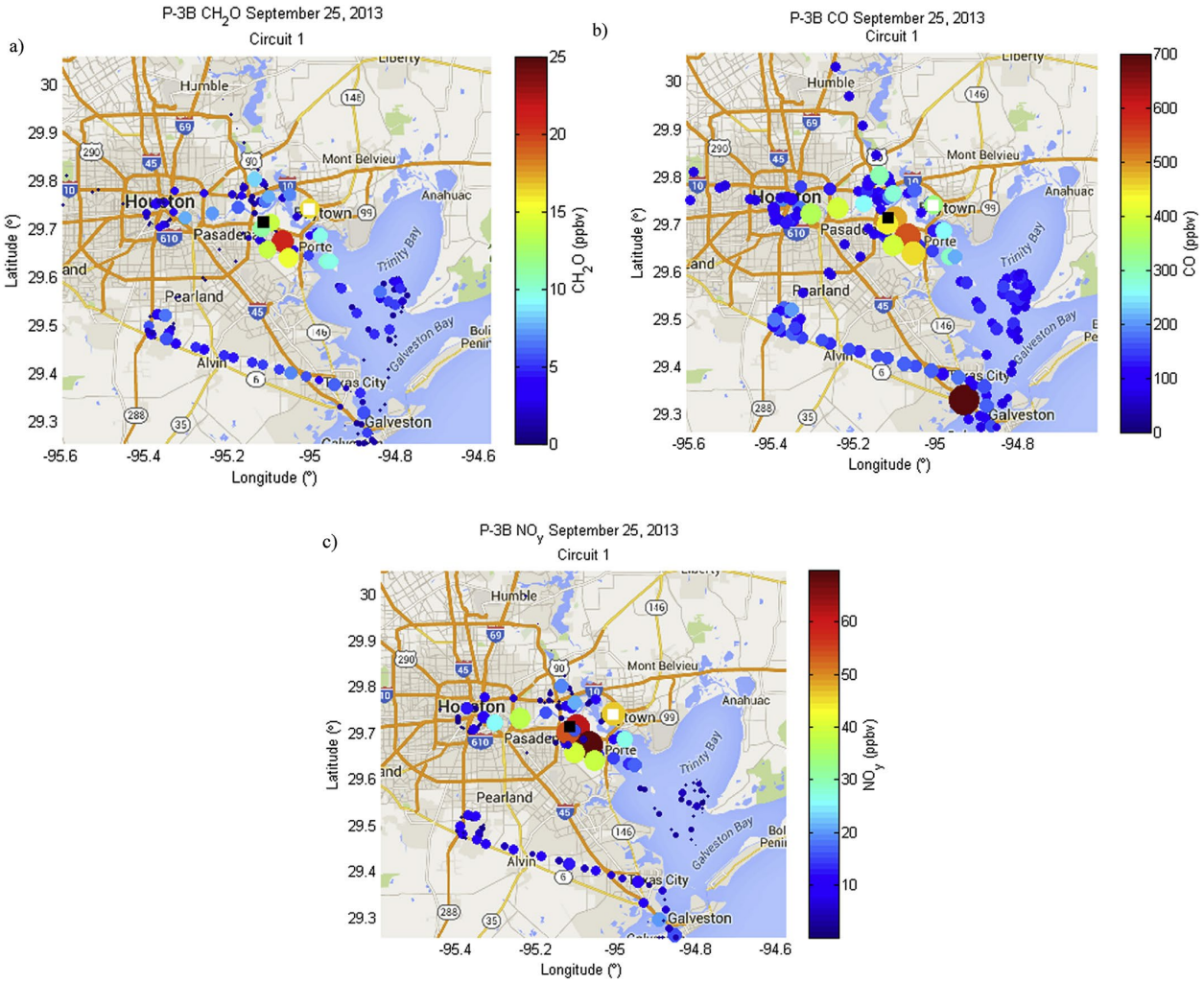


Fig. 8. 09/25/2013 (a) CH2O, (b) CO, and (c) NO_y concentrations measured on the P-3B flight track during the first circuit. Black square is location of Deer Park and white square is location of Facilities at Baytown (the size of the dot corresponds to the concentration).

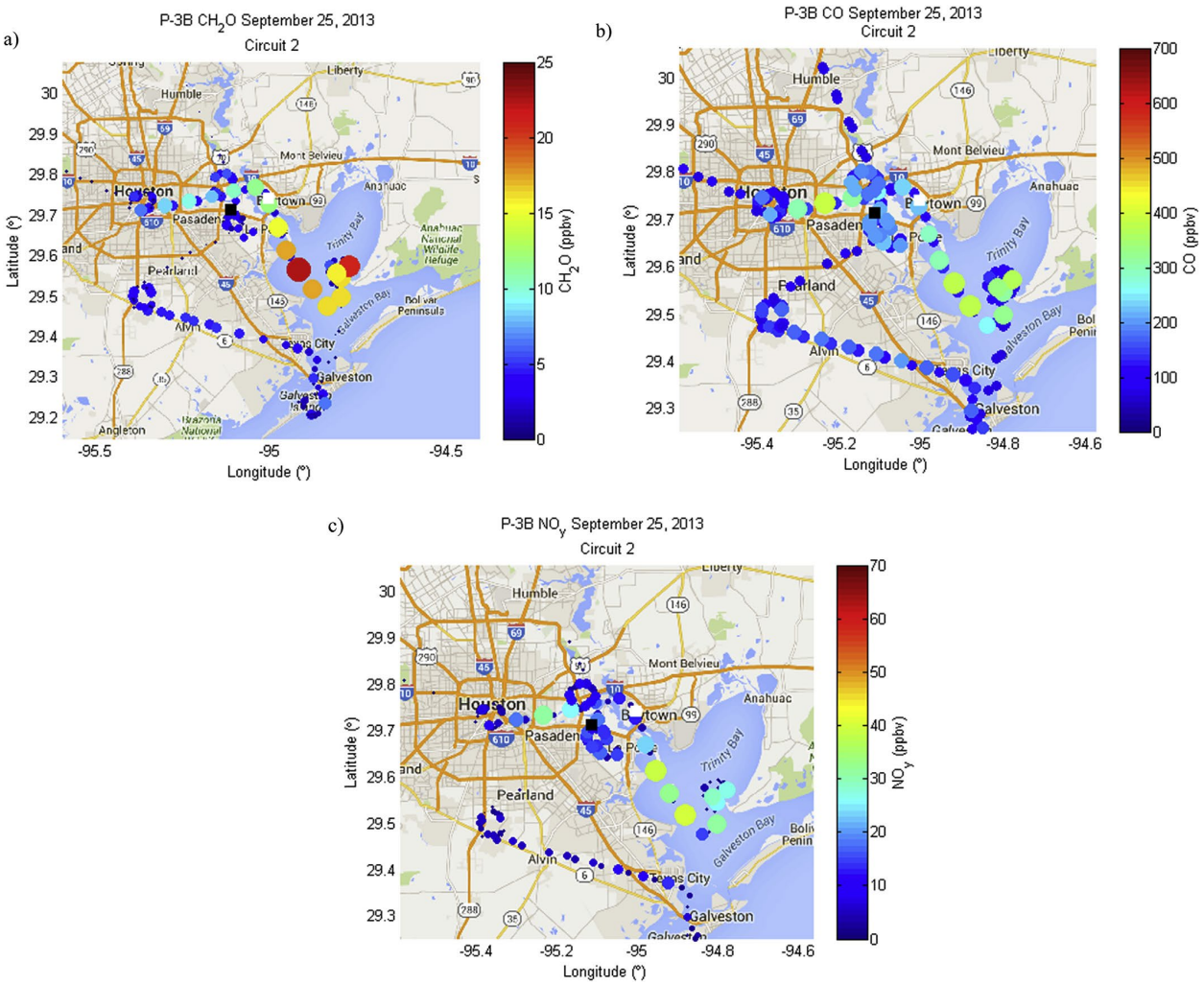


Fig. 9. 09/25/2013 (a) CH_2O, (b) CO, and (c) NO_y concentrations measured on the P-3B flight track during the second circuit.

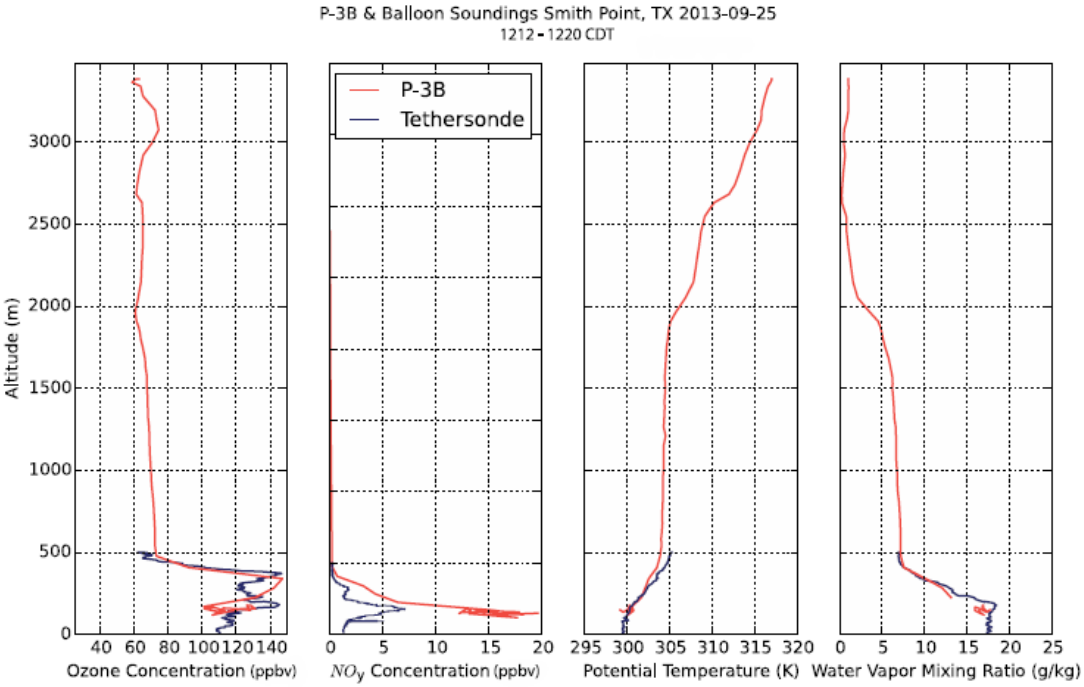


Fig. 10. 09/25/2013 Blended profile of the second circuit P-3B spiral over Smith Point (orange) and the corresponding Millersville tethersonde sounding (blue) of ozone concentration, NO_y concentration (NO_x with interferences from other reactive nitrogen species on tethersonde), water vapor mixing ratio, and potential temperature from the surface to ~4000 m. The P-3B measured a NO_y plume at ~100 m that the tethersonde did not due to temporal and spatial differences between the soundings. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

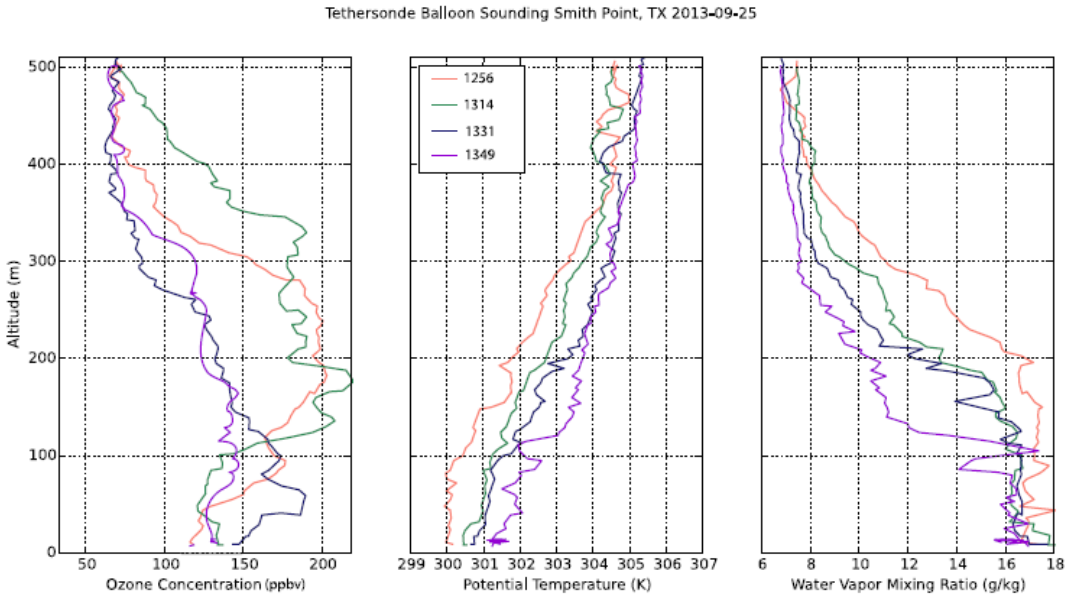
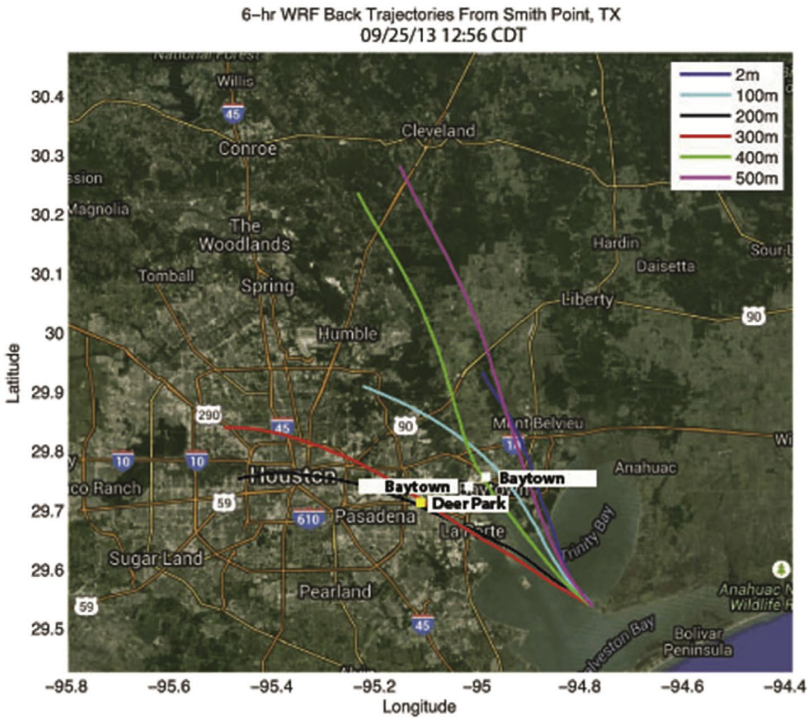


Fig. 11. 09/25/2013 Tethersonde soundings of ozone concentration, potential temperature, and water vapor mixing ratio at Smith Point from 12:56-13:49 CDT from the surface to 500 m.

a)



b)

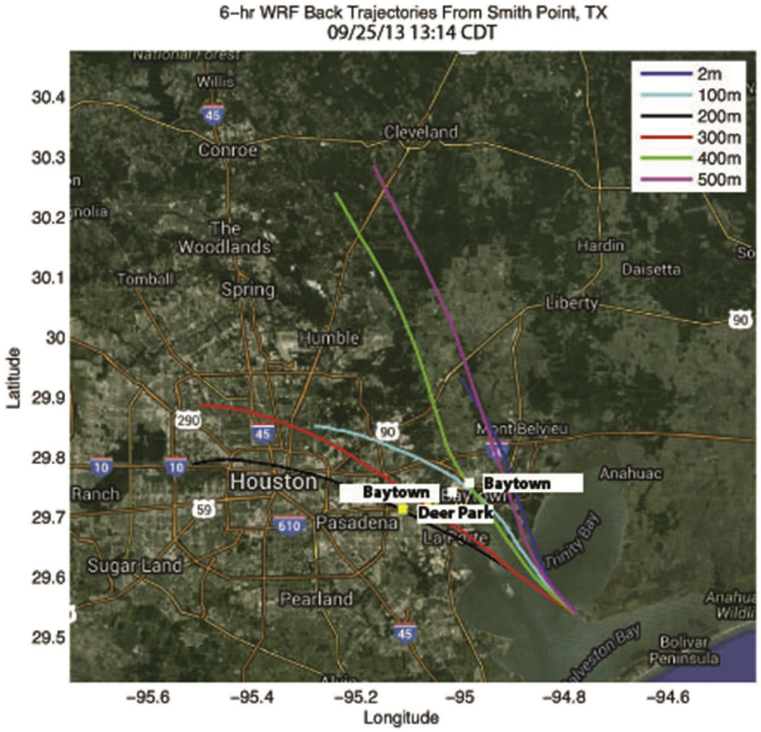
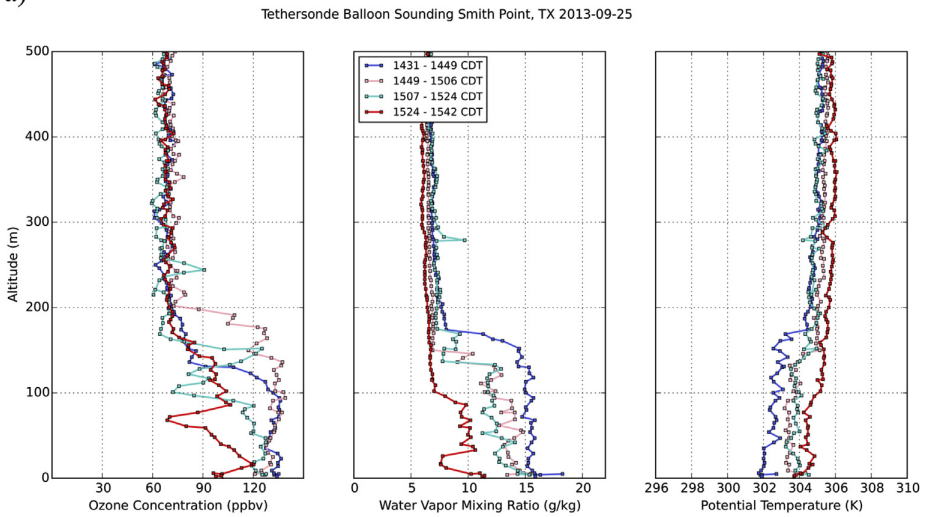
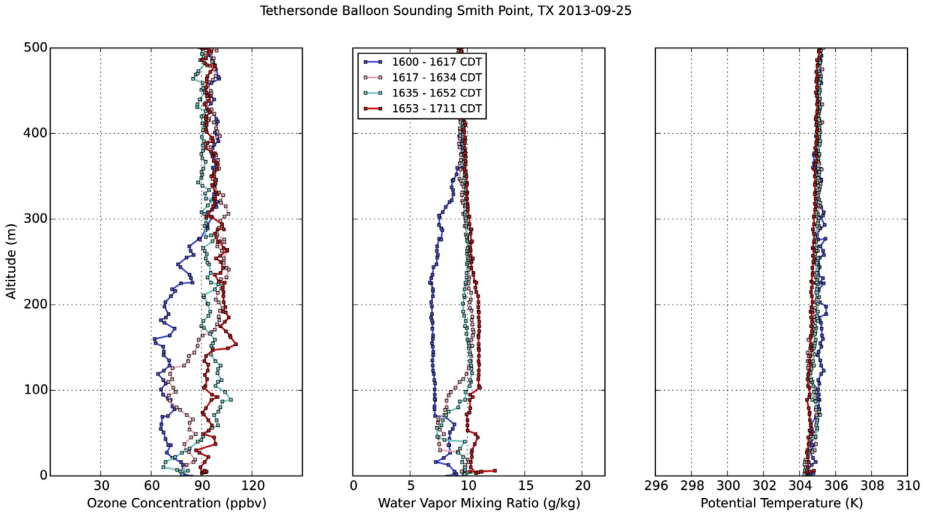


Fig. 12. Six-hour WRF back trajectories at six initialization altitudes (2 m, 100 m, 200 m, 300 m, 400 m, 500 m) from the location of the tethersonde at Smith Point. (a) Initialized at 12:56 CDT (b) Initialized at 13:14 CDT.

a)



b)



c)

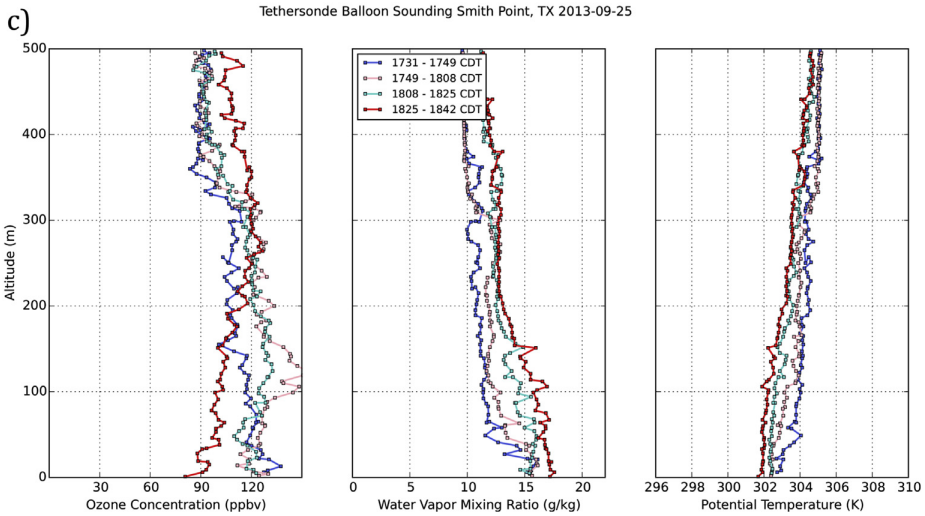


Fig. 13. 09/25/2013 Tethersonde soundings of ozone concentration, water vapor mixing ratio, and potential temperature at Smith Point from (a) 14:31-15:42 CDT, (b) 16:00-17:11 CDT, (c) 17:31-18:42 CDT from the surface to 500 m.

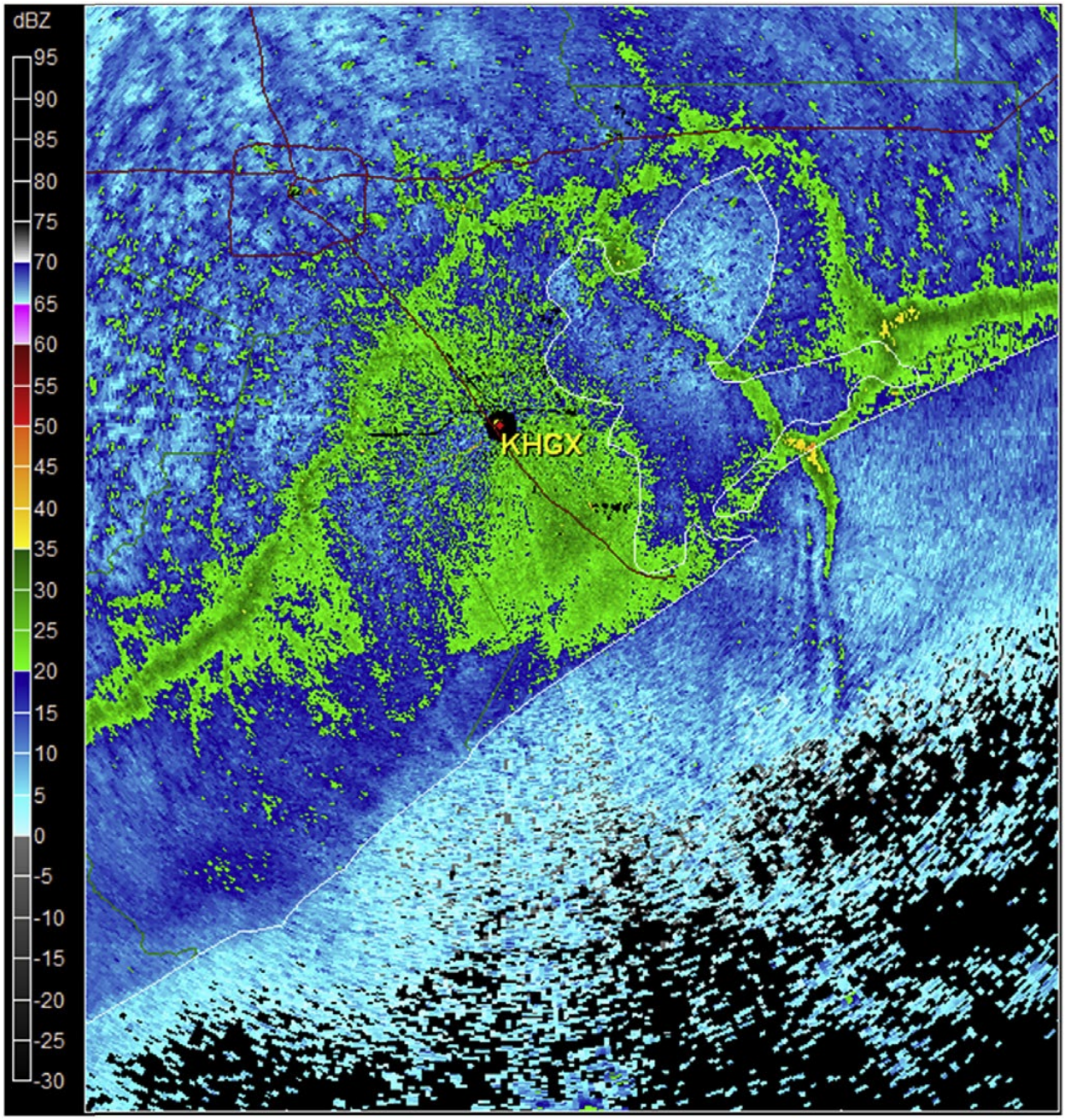


Fig. 14. 09/25/2013 Houston/Galveston, TX (KHGX) radar reflectivity in dBZ of the bay and Gulf breezes at 22:30 UTC (17:30 CDT) passing over Smith Point.

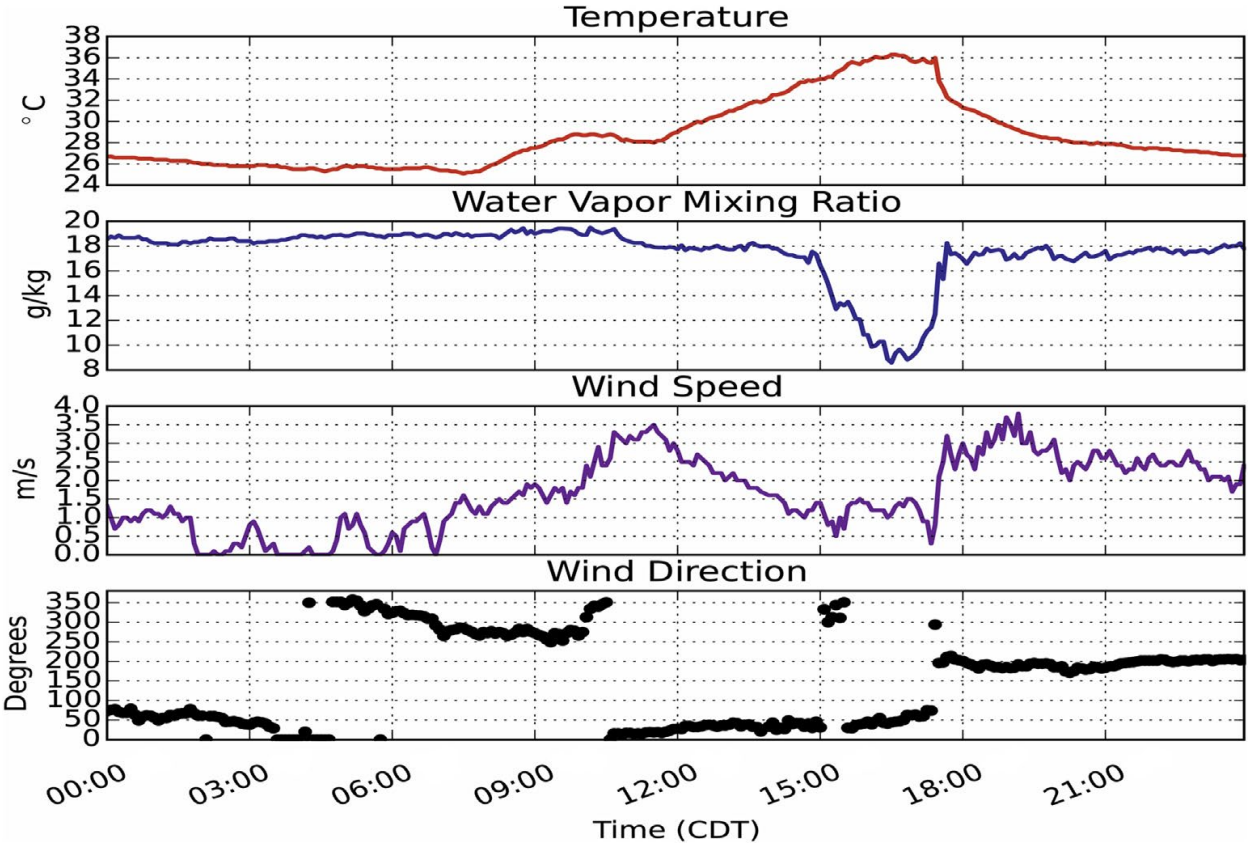


Fig. 15. 09/25/2013 surface WeatherPak observations of temperature, water vapor mixing ratio, wind speed, and wind direction from MARAF platform at Smith Point.

Tables

Table 1. Measurements with uncertainties/accuracies and deployment platform for Edgewood and Smith Point.

Instrument & Model	Measurement	Platform	Uncertainty/Accuracy
Vaisala, TTS111	Temperature, RH, Pressure	Tethered Balloon	±0.5 °C, ±5%, ±1.5 hPa
2B Technologies, 205	Ozone	Tethered Balloon	±2%
2B Technologies, 401/410	NO/NO_2	Tethered Balloon (Smith Point only)	±2%
KNMI NO2-sonde	NO_2	Tethered Balloon (Smith Point only)	N/A (TBD)
ScinTec, MFAS SODAR & RAE1 RASS	Vertically Resolved Wind Speed & Direction	MARAF	0.3-0.5 m/s, ± 3° (<2.0 m/s)
Flux Tower Instruments (denoted by *)	Near-surface Fluxes	MARAF	
CSI 3-D Sonic Anemometer, CSAT3*	u,v,w; Tv	MARAF	Ux,Uy: ±8 cm/s Uz: 4 cm/s Direction: ±0.7° at 1 m/s Tv: N/A
LI-COR H_2O/CO_2 Gas Analyzer, LI-7500*	H_2O/CO_2 concentration	MARAF	CO_2 ±1% H_2O ±2%
Vaisala Pressure Sensor, PTB220B*	Pressure (hPa)	MARAF	±0.25 hPa
Micromet Systems Net Radiometer, Q*7*	Net Radiation (Wm^{-2})	MARAF	- 6% @ 7 m/s for positive fluxes, 1% at 7 m/s for negative fluxes
Surface WeatherPak 2000	WxPak Pressure	MARAF	±1 hPa at 22 °C
	WxPak Compass	MARAF	< ±30°
	WxPak Wind Speed	MARAF	±0.3 m/s
	WxPak Wind Dir.	MARAF	±3°
	WxPak Humidity and Temperature	MARAF	±0.8%/±0.1 K at 23 °C
TECO Inc., 29C	Surface O_3	NATIVE	±2%
TECO Inc., 42C-Y	Surface NO/NO_y	NATIVE	±3%
NCAR 4 Channel Chemiluminescence	Ozone	P-3B	±5%
NCAR 4 Channel Chemiluminescence	$NO/NO_2/NO_y$	P-3B	10-15%
General Eastern, 1011B	Temperature	P-3B	±0.2 °C
Rosemount, MADT 2014	Pressure	P-3B	±0.25 hPa
DFGAS	CH_2O	P-3B	±4%

Table 2. Conditions during bay breezes observed on 7/29/2011 at Edgewood, MD.

	Time (EDT)	Specific humidity (g/kg)	Temperature (°C)	Ozone concentration (ppbv)
Bay breeze 1	13:30-13:50	15.6 to 18.5	37.8 to 35.8	77 to 113
Bay breeze 2	16:15-17:25	13.6 to 16.5	38.5 to 37.5	84 to 91
Bay breeze 3	18:00-20:00	15.0 to 18.0	37.6 to 36.3	87 to 107

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