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The California Baseline Ozone Transport Study (CABOTS)

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Abstract:	Ozone is one of the six 'criteria' pollutants identified by the U.S. Clean Air Act Amendment of 1970 as particularly harmful to human health. Concentrations have decreased markedly across the U.S. over the past 50 years in response to regulatory efforts, but continuing research on its deleterious effects have spurred further reductions in the legal threshold. The South Coast and San Joaquin Valley Air Basins of California remain the only two ‘extreme’ ozone non-attainment areas in the U.S. Further reductions of ozone in the West are complicated by significant background concentrations whose relative importance increases as domestic anthropogenic contributions decline and the national standards continue to be lowered. These background concentrations derive largely from uncontrollable sources including stratospheric intrusions, wildfires, and intercontinental transport. Taken together the exogenous sources complicate regulatory strategies and necessitate a much more precise understanding of the timing and magnitude of their contributions to regional air pollution. The California Baseline Ozone Transport Study was a field campaign coordinated across Northern/Central California during spring/summer 2016 aimed at observing daily variations in the ozone columns crossing the North American coastline, as well as the modification of the ozone layering downwind across the mountainous

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The California Baseline Ozone Transport Study (CABOTS)

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Capsule Summary: A coordinated field campaign across Northern and Central California during the spring and summer of 2016 measured daily fluctuations in the ozone column flowing onshore as well as its modification by regional emissions during transport over land, and helps to clarify the impacts that the variable chemical boundary conditions have on surface air quality in complex terrain.

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Abstract

Ozone is one of the six 'criteria' pollutants identified by the U.S. Clean Air Act Amendment of 1970 as particularly harmful to human health. Concentrations have decreased markedly across the U.S. over the past 50 years in response to regulatory efforts, but continuing research on its deleterious effects have spurred further reductions in the legal threshold. The South Coast and San Joaquin Valley Air Basins of California remain the only two ‘extreme’ ozone non-attainment areas in the U.S. Further reductions of ozone in the West are complicated by significant background concentrations whose relative importance increases as domestic anthropogenic contributions decline and the national standards continue to be lowered. These background concentrations derive largely from uncontrollable sources including stratospheric intrusions, wildfires, and intercontinental transport. Taken together the exogenous sources complicate regulatory strategies and necessitate a much more precise understanding of the timing and magnitude of their contributions to regional air pollution. The California Baseline Ozone Transport Study was a field campaign coordinated across Northern/Central California during spring/summer 2016 aimed at observing daily variations in the ozone columns crossing the North American coastline, as well as the modification of the ozone layering downwind across the mountainous topography of California to better understand the impacts of background ozone on surface air quality in complex terrain.

Air pollution is responsible for over 6 million premature deaths annually worldwide –twice as many as AIDS, malaria, and tuberculosis combined (Landrigan et al. 2018). It is estimated that air pollution directly contributes to more than 200,000 premature deaths per year in the U.S. (Caiazzo et al. 2013). Many of these deaths are linked to fine particulate matter (PM), but ozone (O_3) plays a central role in most air quality issues because it is the principal source of hydroxyl (OH) and nitrate (NO_3) radicals, the leading agents of atmospheric oxidation, which produce PM and other components of photochemical smog. Moreover, tropospheric ozone is an important greenhouse gas with a radiative forcing of 0.4 ($\pm$0.12) Wm^{-2}, just shy of methane (CH_4) (Myhre et al. 2013) and is a phytotoxic pollutant that impacts agricultural yields and tree growth (Lapina et al. 2016). Long-term exposure to O_3 has been implicated in the development of asthma in children (McConnell et al. 2002) and reduced cognitive performance in the elderly (Zhang et al. 2018).

Tropospheric ozone originates from both natural and anthropogenic sources and is photochemically produced by the auto-catalytic oxidation of carbon monoxide (CO) and volatile organic compounds (VOCs) in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$.) Natural sources include direct injection from the stratosphere and secondary production from emissions of non-anthropogenic origins such as biogenic VOCs (BVOC) emitted by vegetation (Sindelarova et al. 2014), and NO_x from unmanaged soils (Vinken et al. 2014) and lightning (Zhang et al. 2003). Wildfires (see sidebar) are another growing source of both NO_x and VOCs (Andreae and Merlet 2001; Jaffe and Wigder 2012). Because the principal loss of ozone requires photolysis followed by reaction with water vapor, and water vapor mixing ratios decrease dramatically with height due to the Clausius-Clapeyron relation, the photochemical lifetime of ozone is altitude

dependent and ranges from one week in the marine boundary layer (Conley et al. 2011) to one year in the upper troposphere (Kley et al. 1996). With a global average lifetime of about 3 weeks (Young et al. 2013), the ozone observed directly upwind of the U.S. West Coast can therefore include anthropogenic contributions transported from East Asia and Europe, and even ozone that originated in the U.S. but has circumnavigated the globe. This so-called 'baseline' ozone constitutes a significant fraction of the ambient concentrations measured in the Western U.S. Here we conform to the definition proposed by the Task Force on Hemispheric Transport of Air Pollution (HTAP) and adopted in the recent review by Jaffe et al. (2018): "Baseline ozone is defined as the observed ozone at a site when it is not influenced by recent, locally emitted or anthropogenically produced pollution (HTAP 2010)." 'Background' ozone, on the other hand (including, for example, North American Background, or Policy Relevant Background), is a model estimate of the O_3 abundances calculated when the anthropogenic precursor emissions from specific areas are omitted or "zeroed out". Throughout this work we will use the term 'baseline' due to the observational emphasis of the study.

The Western U.S. is particularly susceptible to transported ozone because of several physical factors. First, the baroclinicity induced by the North American coastline at the end of the North Pacific storm track, coupled with the planetary wave activity of the adjacent cordillera promotes cross-tropopause transport of stratospheric ozone in this region (Sprenger and Wernli 2003). This process peaks in the spring but can occur throughout the year (Škerlak et al. 2014). Second, synoptic-scale subsidence in the lee of the Pacific subtropical anticyclone draws both naturally occurring ozone and transported pollution (including ozone and its precursors such as peroxyacetylnitrate, PAN) from the middle and upper troposphere towards the surface (Hudman

et al. 2004). And third, the deep convective atmospheric boundary layers (ABLs) of the interior West provide stronger coupling between the free troposphere (FT) and the surface (Langford et al. 2017). This direct convective entrainment is strongest during the summer months above arid and/or high elevation surfaces but is weaker within California's Central Valley where the unusually shallow ABLs are deepest in the springtime (Bianco et al. 2011) and decrease markedly during summer as a result of mesoscale subsidence induced by the mountain-valley circulation (Trousdell et al., 2016) and irrigation practices (Li et al. 2016).

Several modeling studies (Brown-Steiner and Hess 2011; Liang et al. 2004) have shown that the trans-Pacific transport of anthropogenic ozone from Asia is fundamentally different in spring and summer. Episodic long-range transport from Asia increases in frequency with altitude, and peaks in the late spring (April-June) when the prevailing westerlies are strong and extratropical cyclone activity is highest (Lin et al. 2012a). During the summer, much of the Asian pollution is lofted into the upper troposphere (>5 km) by deep convection associated with the East Asian summer monsoon. This summertime ozone is then transported eastward by the prevailing westerly winds aloft, often remaining well above the Coast Range and the Sierra Nevada; however, some is brought downward in the quasi-isentropic flow equatorward and toward the elevated diabatic heating of the mountains.

Despite an estimated reduction in anthropogenic ozone enhancements above baseline levels by a factor of 5 since 1980 (Parrish et al. 2017), California and other western states must continue to achieve significant new emission controls on ozone precursors in order to attain compliance with the current National Ambient Air Quality Standard (NAAQS). In light of observations indicating rising emissions of ozone precursors from Asia (Sun et al. 2018; Verstraeten et al.

2015), more frequent wild fires (Westerling et al. 2006), and a possible increase in stratosphere-troposphere exchange (Stevenson et al. 2006), concern is mounting about the ability of California and other western states to meet the NAAQS because of a rising baseline component beyond their regional or national regulatory purview.

Numerous modeling (Jacob et al. 1999; Lin et al. 2012b; Pfister et al. 2011) and measurement (Cooper et al. 2011; Jaffe et al. 1999; Parrish et al. 2014; Yates et al. 2017) studies have examined the impact of baseline ozone on surface concentrations across California, but there remains a great deal of uncertainty about the contributions of stratospheric and other transported ozone to surface concentrations in the San Joaquin Valley (SJV), the southern part of the California Central Valley and one of only two 'extreme' ozone non-attainment areas remaining in the U.S. (U.S. EPA Green Book, https://www.epa.gov/green-book). Much of this uncertainty is thought to result from a lack of detailed information about the vertical distribution of ozone. Intermittent measurements by ozonesondes or aircraft do not provide the temporal coverage needed to easily evaluate how well models can determine the fraction of baseline ozone contributing to surface concentrations measured in the SJV, and this deficiency is by no means limited to California. The main objective of the California Baseline Ozone Transport Study (CABOTS) was to observe the daily changes in ozone layering at the coast (approximately upwind of California) and simultaneously the layering in the Central Valley (downwind of the major emission sources and coastal mountains) in order to produce an unprecedented data set that might serve to promote ensuing modeling studies. Some issues that the novel data set may help to illuminate include: i.) the fidelity with which air quality models represent ozone transport on synoptic, regional, and local scales in complex terrain; ii.) the relative contributions of stratosphere-troposphere

exchange, Asian emissions, and wildfires to the abundances and variability observed in the ozone profiles in and around California; and iii.) the extent to which exogenous ozone that has been transported from afar mixes down to surface sites affecting ozone NAAQS violations in the SJV. Because the project was sponsored primarily by the California Air Resources Board (CARB), a state agency, its initial scope is more limited than many federal scale projects in that it did not include observations of a large suite of photochemical reactants nor did it sponsor extensive modeling efforts. This work summarizes the measurements and unique meteorology of California that serve as the context for this novel data set and attempts to draw attention to its potential utility for future modeling studies of the impacts of long-range transport on global air quality issues.

THE CABOTS FIELD CAMPAIGN.

Geography

The San Joaquin Valley encompasses the southern two-thirds (~65,000 km^2) of the 700 km long Central Valley of California (Figure 1). Although it is one of the largest valleys in the world, the geometry and meteorology are not too dissimilar from other places such as the Po Valley in Italy, the San Luis Valley in Southern Colorado, the Latrobe Valley in Australia, and the Central Valley of Chile. The 50 – 100 km wide San Joaquin Valley is bounded by the Southern Coast Ranges with elevations of less than 1.5 km above mean sea level (asl) to the west, the Sierra Nevada (<4 km asl) to the east, and the San Emigdio and Tehachapi Mountains (<2.5 km asl) to the south. The flat valley floor rises gradually from sea level in the Sacramento–San Joaquin River Delta near Stockton to ~130 m asl at Bakersfield to the south (see Figure 1). The SJV is one of the most

important agricultural areas in the U.S. responsible for 12% of all U.S. agricultural production and home to about 10% of California's residents.

Measurement Suite

Measurements of ozone vertical profiles by ozonesondes, a ground-based lidar, and aircraft, together with surface ozone concentrations were made from May to August of 2016 (Table 1, Figure 2). The core measurements included near-daily ozonesonde profiles collected at two sites along the coast and an ozone lidar data set collected in Visalia, a city of nearly 140,000 residents located deep within the San Joaquin Valley, approximately 60 km southeast of Fresno. Figure 2 shows the locations of the two primary CABOTS ground sites: the ozonesonde launch site at the University of California, Davis Bodega Marine Laboratory (UCD BML) in Bodega Bay (BBY) and the lidar site at the Visalia Municipal Airport (VMA) on the eastern side of the SJV. The UCD BML (38.319°N, 123.072°W) is located approximately 80 km north of San Francisco and ozonesondes were launched daily there 6 days per week from the middle of May to the middle of August 2016 by researchers from San José State University (SJSU). Additionally, a second launch site was operated at the same periodicity for a subset of the project at Half Moon Bay (HMB, 37.504°N, 122.483°W) located 25 km to the south of San Francisco.

The National Oceanic and Atmospheric Administration (NOAA) TOPAZ (Tunable Optical Profiler for Aerosols and oZone) differential absorption lidar (DIAL) system (Alvarez et al. 2011) was operated daily during two 3-week long intensive operating periods: IOP1 (May 27-June 18) and IOP2 (July 18-August 7). TOPAZ uses a scanning mirror for measurements at different elevation angles to provide quasi-continuous ozone and aerosol backscatter profiles from 25 m

to about 6 km above ground level (≥8 km at night). A radar wind profiler and radio-acoustic sounding system (RASS) at the Visalia airport provided measurements of virtual temperature profiles up to 1-1.5 km and winds up to 4 km.

In addition to these core measurements, the University of California, Davis (UCD) and Scientific Aviation, Inc. team conducted aircraft measurements to characterize the evolution of ozone layering in the lower atmosphere (< 1,500 m) over the course of the entire diurnal cycle from Fresno to Bakersfield. Flights for this separate study investigating the effects of residual layer ozone and its mixing on surface exceedances overlapped with some of the CABOTS project. Additionally, supplemental funding was provided by the US EPA to study the afternoon ABL and photochemical ozone production rates in the valley between Fresno and Visalia, upwind of the TOPAZ lidar, on six additional days: July 27-29 and August 4-6 (Trousdell et al. 2019). Furthermore, the Alpha Jet Atmospheric eXperiment (AJAX) of the NASA Ames Research Center (Hamill et al. 2016) performs regular missions to measure ozone and greenhouse gases, such as ozone, carbon dioxide (CO_2), methane (CH_4), and formaldehyde (HCHO) over California and Nevada, and was coordinated to contribute to CABOTS. A comprehensive intercomparison of the TOPAZ lidar with both airborne platforms (Langford et al. 2019) found excellent agreement (within 5 ppbv on average) among the three measurements.

In addition to the usual ozone data collected at nearly 80 CARB (25 in the SJV Air Basin) routine surface sites in Central and Northern California that are available for the study (https://www.arb.ca.gov/aqmis2/aqdselect.php), the San Joaquin Valley Air Pollution Control District (SJVAPCD) had funded ozone measurements made by UC Davis at the Oliver Observing Station (http://tycho.mira.org/oosweather/), which is operated by the Monterey Institute for

Research in Astronomy (MIRA: http://www.mira.org/) on Chews Ridge. This solar and wind-powered mountain observatory is at an elevation of 1,550 m asl in the Los Padres National Forest 40 km southeast of Monterey but located about half that distance downwind from the Big Sur coastline to the southwest from whence the winds blow two-thirds of the time (Asher et al., 2018). Measurements of O_3, NO, NO_2 and NO_x collected at this site during CABOTS are available although much of it was heavily impacted by the Soberanes Fire during the second IOP, which burned along the northwest perimeter of the Oliver Observing Station from late July to early October.

Synoptic Setting

The SJV has a Mediterranean climate well-suited for agricultural production. The winters are usually cool and moist, but the summers are hot and dry with less than 5% of the annual precipitation falling between May and September (https://cdec.water.ca.gov/snow_rain.html). The principal ABL summer wind patterns are dominated by the thermally-driven mesoscale circulations of the coupled sea-land/valley-mountain flow. Onshore winds can usually only penetrate into California's Central Valley at breaks in the Coast Range because the marine ABL tends to be well below the height of the coastal mountains (Dorman et al., 2000), and mostly the winds feed in through the Carquinez Strait east of the San Francisco Bay Area (Figure 1). This inflow is modulated by the synoptic conditions as well as diurnal up-valley winds and by upslope/downslope flow along the flanks of the Sierra Nevada and Coastal Ranges (Bao et al. 2008; Zhong et al. 2004). The northwesterly inflow brings ozone and other pollutants from the San Francisco Bay area and Sacramento (Fast et al. 2012) up the valley towards Bakersfield, but also flushes the valley with clean marine air, particularly at night when up-valley winds can

strengthen into an elevated low-level jet (LLJ) (Caputi et al. 2019). Some of this flow exits the valley through the Tehachapi Pass 75 km southeast of Bakersfield, but much is blocked by the mountains and recirculated northward where it interacts with cross-valley drainage flows and the LLJ to form an early morning low-level counterclockwise circulation known as the Fresno eddy (Bao et al. 2008; Lin and Jao 1995), which is believed to foster the buildup of ozone in the SJV (Beaver and Palazoglu 2009).

The daytime upslope winds carry ozone and other pollutants from the SJV into the surrounding mountains (Panek et al. 2013) with the circulation closed by elevated compensation flows above the valley (Zardi and Whiteman 2013). Subsidence created by this mountain-valley circulation and the synoptically persistent North Pacific High (Trousdell et al. 2016), and to a lesser extent, evaporative cooling associated with widespread agricultural irrigation (Li et al. 2016), limits the growth of the convective mixed layers (ML) in the central SJV to ~1 km or less during summer (Bianco et al. 2011) with somewhat deeper mixed layers in the southern part of the valley where the converging up-valley flow is lifted by the weir of the Tehachapi Mountains (Trousdell et al. 2016). The sidewall venting creates elevated pollution layers above the ML that spread across the valley when the synoptic forcing is weak (Fast et al. 2012; Leukauf et al., 2016). Ozone remaining in the residual layer left after the ML decays can be re-entrained the following day or turbulently mixed into the stable nocturnal boundary layer by the LLJ and destroyed at the surface (Caputi et al. 2019). Above the average daytime ABL height (500 – 600 m), there exists an intermediate layer, composed of a mixture of free tropospheric inflow coming over the coast range and air lofted above the ABL by the sidewall slope flows. Because this region is a mixture of baseline FT air and surface-influenced ABL air, and because it is stably stratified yet

238 intermittently turbulent (Faloona 2018) we refer to it as a buffer layer (BuL) similar to that
239 described by Russell et al. (1998) over the ocean.

240 The synoptic and mesoscale subsidence associated with the Pacific High and mountain-valley
241 circulation also inhibits cloud formation during the summer, and the combination of high
242 temperatures and clear skies in July and August fosters rapid photochemical production of O_3
243 from NO_x and VOC precursors transported into the valley by the northwesterly inflow, emitted
244 by urban sources, or released by distributed transportation, agricultural, and petrochemical
245 sources within the SJV (Trousdell et al., 2019; Pusede et al. 2014). To some degree the Fresno
246 Eddy recirculates these pollutants within the Southern SJV to help create an ozone hotspot near
247 Fresno (Beaver and Palazoglu 2009), although the strong associated low-level jet may also induce
248 vertical mixing at night that depletes ozone (Caputi et al. 2019).

249 Synoptic charts of reanalysis data for both TOPAZ IOPs, as well as a June – August average for
250 2010-2015, are shown in Figure 3 (data from the National Center for Environmental Prediction's
251 North American Regional Reanalysis, NCEP/NARR: https://www.esrl.noaa.gov/psd/cgi-
252 bin/data/narr/plotday.pl). On the 900 hPa (≈ 1 km asl) geopotential height fields, the synoptic
253 patterns for both IOPs are seen to closely resemble the climatological pattern, with a thermal
254 low over the southern California/Nevada border and alongshore, equatorward winds over the
255 ocean. A mean up-valley (northwesterly) flow is present in the SJV due to the mountain-valley
256 circulation, but is geostrophically reinforced by the pressure gradient between the offshore
257 Pacific High and inland thermal low.

Figure 4 shows the monthly average tropospheric column ozone concentrations derived from the difference between the total column ozone measured by the NASA Aura Ozone Monitoring Instrument (OMI) and the stratospheric column measurements made by the Aura Microwave Limb Sounder (MLS) (Ziemke et al. 2006) during the three core months of the CABOTS project. Superimposed on the tropospheric averaged concentrations are the NCEP reanalysis winds at the 300 hPa isobaric level for each month considering that the main summertime trans-Pacific transport is centered aloft near 8 km (Brown-Steiner and Hess 2011). The patches of elevated ozone over the central Pacific in June and the faster winds may represent the trailing edge of springtime baroclinicity in the midlatitudes which is often accompanied by enhanced stratosphere-troposphere exchange. The seasonal progression of Figure 4 also shows the poleward advancement of the ITCZ with its depleted ozone and easterly winds that shunt the main Asian effluent northward throughout the summer season. Interannual variations in this advancement associated with the Pacific Decadal Oscillation (PDO) have been shown to markedly influence the long-term trends in ozone observed at the baseline site on Mauna Loa, Hawaii (Lin et al. 2014).

Some Examples of the CABOTS measurements

The curtain plot of Figure 5 shows the daily ozonesonde data from both the Bodega and Half Moon Bay sites. The daily data exhibits patterns similar to those seen in the Trinidad Head analyses of less frequent (weekly) soundings over a decade reported by Oltmans et al. (2008), with undulations of ozone concentrations in the mid-to-upper troposphere of 3-5 week periodicities. A comparable ozonesonde study by Cooper et al. (2011) coordinated with the CalNex campaign (Ryerson et al. 2013) reported the ozone percentile distributions observed in

near daily profiles above Trinidad Head (THD, 320 km north along the coast from Bodega Bay) for 5 weeks (May 10- June 19) during 2010. At 2 km the percentiles at 5%, 33%, 50%, 67%, and 95% were found to be 30, 44, 47, 51, and 62 ppbv (their Figure 10). Figure 5 shows three or four episodes of enhanced ozone in the upper troposphere at BBY and the depletion of marine ABL ozone, below about 500 m, as the summer progresses due to photochemical destruction in a more or less pristine environment. The observed day-to-day variability does appear to be larger during the three-month interval of CABOTS which shows the percentile values of 5%, 33%, 50%, 67%, and 95% at 2 km to be 36, 51, 57, 62, and 77 ppbv. The 10 ppbv greater median ozone at 2 km above BBY than at THD is consistent with the upward sloping meridional isopleths reported in Cooper et al. (2011). The larger interquartile range may be due to the longer interval of the CABOTS data set, but is indicative that day-to-day variability is important for accurately constraining the oxidant boundary conditions along the North American inflow. In order to take a preliminary look at how well a commonly used chemical transport model captures the ozone amounts and variability as a chemical boundary condition, a comparison of the observed mean and standard deviation profiles (black) with those generated in the Model for Ozone and Related Chemical Tracers (MOZART, Emmons et al., 2010) (red) is shown in Figure 6 for the 24 ozonesondes launched between mid-July and mid-August at the HMB site. While the model does capture the strong gradients at the top of the marine ABL well, there does seem to be a systematic 5-15 ppbv underestimate throughout most of the troposphere followed by overpredictions greater than 20 ppbv above 10 km (not shown). It is also important to note that in the region between 1-3 km, which likely influences the surface ozone levels in the interior Central Valley and western face of the Sierras the most, the observed daily standard deviation is

~60% greater than is captured in the model. Furthermore, in agreement with the findings presented in Figure 6 of frequent ozone concentrations well above 60-70 ppb (mean + 1 standard deviation) at 1.5 km altitudes and above, an analysis of the ozone time series collected at Chews Ridge from 2012-2014 determined that the ODV for this remote site located in a National Forest in the coastal mountains was 70.5 ppbv, technically in violation of the NAAQS.

Figure 7 shows the compiled time series from the TOPAZ ozone lidar at Visalia over the course of the two IOPs. The scalloped appearance of the daily curtain plots is caused by increased solar background radiation near local noon, which reduces the lidar maximum range. In general, the lidar data shows the rich textures of the air above the valley with frequent episodes of high ozone advected overhead in the upper troposphere (also seen in Figure 5), as well as synoptic accumulations of ozone in the ABL, but also in the buffer layer above up to 2-2.5 km. Generally, the features appear to slope downward over the course of several days as would be expected with isentropic (westerly) flow over the heated (and better mixed) continental atmosphere.

The TOPAZ measurements from IOP2 are also presented as diurnally-averaged profiles in Figure 8. Only the diurnal hours with more than 5 days of measurements are shown. The statistics are most robust for the hours between 1300 and 1900 PDT when more than 12 days of measurements are averaged. The corresponding mean winds from the co-located SJVAPCD profiler are superimposed on each plot; wind speeds less than 2 ms^{-1} are represented by crosses. The wind vectors show the horizontal wind direction with the top of the plot representing north and the right side east (arrows that point to the right in the figure therefore represent *westerly* winds.) The mean winds, primarily northwesterly throughout a deep layer (ABL + BuL) during daytime, begin westerly (upslope) in the late morning and grow stronger and more northerly in

the afternoon ABL as the up-valley thermal circulation prevails. The general pattern appears very similar to those shown in Zhong et al. (2004) and Bao et al. (2008) that were based on data collected at Visalia during the 2000 Central California Ozone Study. The solid black line in each plot represents the ABL height calculated from gradients in the RASS virtual temperature profiles. The dashed line in Figure 8 shows the ABL height calculated from direct measurements of the vertical velocity variance by an experimental NOAA ESRL Doppler lidar deployed next to TOPAZ during the last two weeks of the campaign. These two independent measurements are in very good agreement and are also consistent with the gradients observed in the TOPAZ UV backscatter profiles. All three methods indicate that the mean ABL height of ~500 m at Visalia corresponds fairly well to the reported average afternoon ABL heights of 550 m for the region between Visalia and Fresno (Trousdell et al., 2019) during the six EPA flights of the Scientific Aviation Mooney. The diurnally-averaged O_3 data for IOP2 shows depletion overnight in a shallow nocturnal boundary layer due to nitrate production and dry deposition (Caputi et al. 2019) followed by a well-mixed buildup of O_3 in the ABL during the day. The prevalence of elevated ozone (>70 ppb, orange colors) between 500 – 2,500 m in these average profiles of Figure 8 is evidence that ABL air is lofted into the buffer layer above due to daytime slope flow along the flanks of the valley during its up-valley progression towards the southeast. Further evidence of this slope venting is presented for other scalars in the discussion surrounding Figure 10 that follows. Note that this buffer layer lies above any residual layer left behind when the convective mixed layer decays in the evening. The accumulation of afternoon lofting is especially evident in Figure 8 at 21:00.

Another representation of the three-layered valley atmosphere is illustrated in Figure 9, which shows a terrain cross-section running from the coast near Chews Ridge perpendicular to

346 the valley axis into the Sierra Nevada (topography data from the NASA Shuttle Radar Tomography
347 Mission.) Winds from the Half Moon Bay ozonesondes are plotted along the coast, and those
348 averaged from the Visalia RASS from 12:00-16:00 July – August at the corresponding cross-valley
349 axis distance. Vector averaged winds are represented by the arrows (north oriented toward the
350 top of the page), and scalar average wind speeds are displayed by the arrow color. The red and
351 the blue lines are average potential temperature and ozone, respectively, measured by the
352 Mooney aircraft during the 6 EPA flights. These data show an average ABL height of ~600 m
353 wherein the afternoon ozone tends to be the highest, and then distinctly show an intermediate,
354 statically-stable BuL where the ozone concentrations are somewhere between ABL and free
355 tropospheric (above ~2,000 m) amounts. These data show the typical southwesterly flow at
356 Chews Ridge entering the valley over the coastal ridgeline, but as the air travels inland it is swept
357 up into the up-valley (northwesterly) wind of the SJV that dominates from the surface all the way
358 to the top of the buffer layer. Somewhere between 2,000 – 2,500 m the vector wind average is
359 almost the null vector, a near balance of up- and down-valley winds, with an average scalar wind
360 speed near 4-5 ms^{-1}, similar to the onshore flow at the same level. Thus, the presence of the high
361 southern Sierra Nevada downwind appears to block the flow's progression, but does not stagnate
362 as much as recirculate the air within the valley buffer layer. Higher up, in the free troposphere,
363 the obstruction of the mountains also impedes the flow, weakening the Coriolis force, but above
364 the reach of the mountain-valley thermal circulation air is pushed down the background pressure
365 gradient (Figure 3), which is directed more or less down-valley, driving southeasterly winds aloft.
366 Note that despite the well-defined thermodynamic capping inversion atop the valley boundary

367 layer, there is little wind shear across the inversion as the daytime up-valley flow persists
368 throughout the ABL and BuL.

369 A more instantaneous picture of the valley atmosphere's layering can be seen in Figure 10
370 showing NASA AJAX spiral/vertical profile at the coastal BBY site (dark purple) compared to the
371 inland profile flown near Visalia (cyan) only 30 minutes later on June 15. Because the AJAX flight
372 was scheduled for the morning, the ozonesonde was launched at BBY early to be nearly
373 coincident at 10:00 PDT. The five scalars shown (from left to right: CO_2, CH_4, O_3, H_2O, and HCHO)
374 clearly indicate the difference between the baseline composition of the air moving onshore at
375 the coast and the air heavily influenced by California's Central Valley. In this instance, the ABL at
376 Visalia appears to be around 800 m and the buffer layer seems to extend up to about 2,500 m.
377 The long chemical lifetime and strong surface sources of CH_4 from agriculture and oil production
378 in the SJV (Trousdell et al. 2016) clearly show the layering of an ABL with methane enhanced by
379 100-150 ppbv over the baseline observed offshore. The secondary enhancements of CH_4 at 1,500
380 m and 2,300 m clearly show the lofting of ABL valley air into the buffer layer that had occurred
381 most likely on the previous afternoon when western slope sidewall venting is most active (Fast
382 et al., 2012; Leukauf et al., 2016), injecting these two layers that wind up over Visalia on the
383 morning of June 15. These layers are more pronounced in CO_2 where they are characterized by
384 drawdown from the agricultural photosynthesis of the valley environment, far more prevalent
385 than where the particular CH_4 sources may have been. At the lowest levels, just above the Visalia
386 airport and CA-99, the CO_2 signal exceeds the marine baseline due to fossil fuel combustion.
387 HCHO also exhibits lofted plumes at 1,500 and 2,300 m over Visalia, similar to methane. Because
388 the midday lifetime of HCHO due to photolysis and reaction with OH is only about 3 hr (Choi et

al. 2010), yet should be very long in the dark away from the surface, the presence of significant amounts of HCHO (up to 1.5 ppbv) shows that the lofting into the buffer layer is accomplished on the time scale of several hours over the course of any day.

The June 15 vertical profile measurements can be put into a larger perspective. Figure 11 superimposes the AJAX CH_4 and O_3 profiles at Visalia from Figure 10 on the TOPAZ time-height aerosol and ozone curtain plots, respectively. The curtain plots show relatively elevated O_3 extending up to ~2,700 m in the BuL. Different origins for these layers is consistent with the abrupt wind shift around 1.5 km. The buffer layer is capped by very clean air with less than 30 ppbv of O_3 and low aerosol backscatter. The absence of turbulence in this layer is reflected by the rapid loss in the wind profiler return signals, and veering of the winds to westerly flow, consistent with the climatological pressure field presented in Figure 3, at the bottom of the clean layer (FT) can be seen. Above lies a 2 km thick layer with >80 ppbv of O_3 (and low aerosol) consistent with transported Asian pollution. The superimposed AJAX profile in Figure 11 shows that the aircraft sampled the very bottom of the transported pollution layer (not shown here, but exhibiting enhanced CO_2, HCHO, and CH_4) before descending into the clean layer.

Because CABOTS obtained many different vertical profiles of ozone across the state it is possible to look at how lower tropospheric ozone layers correlate across large distances. As Parrish et al. (2010) point out, ozonesonde measurements aloft may correlate with various surface sites downwind in a wide lateral swath not due to direct parcel trajectories, but because ozone laminae that are commonly found in the lower troposphere often extend for hundreds of kilometers in the horizontal (Liu et al. 2009). Figure 12 shows the correlations between the two coastal ozonesonde sites 100 km apart for the 22 days with simultaneous launches. The layers

seem to be spatially large enough to encompass both locations at altitudes between 1.5-2 km (near the altitude of the Chews Ridge observations), 4 km, and up near 6 km, with correlation coefficients of about 0.8 (the solid circles in Figure 12 represent the correlations with p-values less than 0.05.) The blue line of the left graph in Figure 12 shows the correlations between the BBY ozonesondes and the fixed Chews Ridge measurements at the same hour 260 km south along the coast (the correlation coefficient is calculated between the surface measurement and each altitude bin of the ozonesonde and lidar). Here the observations at the mountain top site are influenced by dry deposition to the forests and experience some influences of the continental sources, but there is still a correlation of nearly 0.4 with air sampled between 0.8-1.2 km above Bodega Bay. The correlation between the mountain site at Chews Ridge and the Visalia lidar inland shows a pronounced peak just below 2.5 km (green dashed line, right axis) and supports the idea that the Chews Ridge observatory may serve as a decent monitor of the inflow air over the coast range that dilutes the ABL air that is lofted into the buffer layer over the SJV. And finally, the measurements that were taken the farthest apart, 400 km between BBY and Visalia, show some modest correlations at 4.5 km and 1 km (red line, right panel.) The latter correlation near 0.5 corroborates the FLEXPART modeling results of Cooper et al. (2011) showing a strong influence of air parcels at 1 km above Pt. Reyes (~30 km south along the coast from BBY) and their receptor region covering the 'south central' SJV in dictating that flow above the marine ABL gets drawn into the sea/valley breeze circulation that affects the flow up to about 2 km in the valley.

Summary

The collective observations of 440 hours of O_3 lidar profiles over Visalia, over 100 coastal ozonesondes, and several targeted airborne surveys acquired during the 3-month CABOTS field experiment have the potential to greatly illuminate the meteorological mechanisms that determine the vertical distribution of O_3 above the central San Joaquin Valley. The high temporal resolution (daily for the coastal ozonesondes and daytime hourly for the lidar in the Central Valley) data set should be able to provide novel constraints on modeling efforts, ultimately producing new insights into the transport and mixing processes contributing to the high surface O_3 levels found in this and other large valleys around the world. The lidar, RASS, and airborne measurements all confirmed the unusually shallow convective mixed layers of the SJV and the formation of a persistent buffer layer above the ABL between about 0.5 and 2.5 km. Because of the surrounding topography the buffer layer overlying the well-mixed boundary layer is made up of partially stagnating air polluted by regional surface sources and a continuous dilution of incoming free tropospheric air that contains a highly variable amount of exogenous ozone usually present in discrete layers.

The TOPAZ lidar also frequently detected elevated O_3 layers between 4 and 6 km above the valley floor consistent with biomass burning, transport from Asia, or descent from the lower stratosphere. Many of these layers most likely passed over the Sierra Nevada into the Intermountain West where they may have been entrained by the much deeper convective mixed layers that form in that hot, dry higher elevation region (Langford et al. 2015; Langford et al. 2018).

The high frequency daily ozonesondes showed a dramatic gradient in ozone reaching the coast from a median of 23 ppbv near the ocean surface and rising rapidly on average 17 ppb km^{-}

454 [1] up to 2 km, consistent with rapid photochemical destruction in the summertime marine
455 boundary layer. Above that strong gradient the average rise is an order of magnitude gentler
456 (about 2 ppb km^{-1}), however the standard deviation increases aloft. The daily ozonesondes at
457 Bodega Bay discovered the appearance of ozone concentrations below 3 km exceeding 70 ppbv,
458 the 8 hr ambient air quality standard, on average once every 5 days. Initial comparisons with a
459 chemical transport model commonly used to provide the chemical boundary conditions at the
460 North American coast indicate a low bias of about 10 ppbv in the lowest 3 km, below which the
461 air flow damns up along the western edge of the Southern Sierra Nevada mountains and
462 potentially influences surface concentrations in California. The project data set was fortunate to
463 observe the coastal and valley air during several weeks of the Soberanes Fire, a large wildfire that
464 burned 130,000 acres in the coastal mountains over the course of nearly 3 months. The detailed
465 observations of ozone in the complex terrain of the western US is publicly available (at
466 https://www.esrl.noaa.gov/csd/groups/csd3/measurements/cabots/) and may serve to
467 challenge future modeling efforts that attempt to quantify the episodic trans-Pacific transport
468 and impact of distant ozone sources on ongoing surface air quality violations in the mountainous
469 west.

470

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SIDEBAR 1: Regulatory History of O_3 NAAQS and California standards

Exposure to ozone (O_3) can trigger a variety of health problems, including decreased lung function and pulmonary inflammation, especially for children, elderly, and those with pre-existing health conditions such as asthma (EPA 2013). To protect human health, the first ambient air quality standards (AAQS) in California were set by the Department of Public Health (DPH) in 1959, which set one-hour 0.15 parts-per-million (ppm) limit for photochemical oxidant levels in outdoor air. In 1967, the California Air Resources Board (CARB) was established and was granted authority to set future AAQS for the state. The federal Clean Air Act (CAA) of 1970 established the National Ambient Air Quality Standards (NAAQS), which set thresholds for criteria air pollutant levels in outdoor air (EPA 1970). Ground-level O_3 is categorized as one of the six criteria air pollutants regulated under NAAQS. Since then, California has continued to invest and lead a multitude of air quality and health research projects, including the evaluation of the peer-reviewed scientific literature on health effects of exposure. These activities led to the adoption of new O_3 standards, including the one-hour 0.09 ppm O_3 standard in 1988 when O_3 was identified as the primary link to respiratory health problems within the group of oxidants present in the atmosphere. This was also the year when the state legislature adopted the California CAA with strategies that include transportation emission control measures. Following the evaluation mandated by the Children's Environmental Health Protection Act of 1999, CARB and Office of Environmental Health Hazard Assessment (OEHHA) recommended a new 0.070 ppm O_3 standard for the maximum daily 8-hr ozone average (MDA8) in 2005 based on human health and environmental impact assessments on O_3. In October 2015, U.S. EPA followed suit and adopted the 0.070 ppm threshold for the 8-hour O_3 NAAQS. Compliance with the NAAQS is determined

514 by the “ozone design value” or ODV, the 3-year running average of the fourth highest MDA8
515 ozone concentration observed each year at a given site. The ODV is a metric of the high end of
516 the ozone distribution and roughly equivalent to the upper 98th percentile. Non-attainment
517 areas must develop a State Implementation Plan (SIP) that demonstrates how new controls will
518 reduce ground-level ozone to levels below the latest health-based standard. As a result of such
519 continued efforts to minimize the adverse effects of O_3, the maximum eight-hour O_3
520 concentration in the South Coast Air Basin (one of the most polluted regions in California), is now
521 a factor of three lower despite a threefold increase in the number of passenger cars on the road,
522 doubling of population, and economic growth of over two times.

523

524 **SIDEBAR 2: Do wildfires influence O_3, where and by how much?**

525 Fires generate smoke (fine particulates or PM2.5) and a variety of gaseous compounds,
526 including NO_x and VOCs, the photochemical precursors of O_3. The net production of O_3 by
527 wildfires is highly variable, however, with many contradictory observations reported in the
528 literature (Jaffe and Wigder 2012). The NO_x and VOC emission rates depend on many factors
529 including fuel type and combustion temperature, and the subsequent production of O_3 depends
530 on the plume injection height, smoke density, and cloud cover. The amount of O_3 within a given
531 smoke plume also varies with distance from the fire. Measurements made near active fires
532 sometimes show a decrease in O_3 relative to baseline concentrations because of titration by NO
533 (and possibly reduced NO_2 photolysis rates in the shade of the smoke plume), and measurements
534 made far downwind often show little additional O_3 production if the NO_x has been depleted or
535 sequestered into more stable compounds like peroxyacetyl nitrate (PAN). The >132,000 acre

Soberanes Fire was the largest wildfire in California during 2016. The National Interagency Fire Center estimated it to be at the time the most expensive wildland fire in U.S. history with suppression costs exceeding $262 million. The fire was started on the morning of July 22 by an illegal campfire in Garrapata State Park on the windward side of the coast range along California's Big Sur coast. It spread southeast from there over the next 10 weeks and by mid-August was burning along the perimeter of the Oliver Observing Station at Chews Ridge (Figure SB2.1). The fire burned more than 57,500 acres (40% of the final burn area) during the first two weeks, and smoke from the fire filled much of the central SJV by the last week of July when the highest surface ozone values of 2016 were recorded.

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Tables

Observational Overview Table:

Bodega Bay, Ozonesonde Site, (38.319N, -123.072W), San José State University		
Parameters Observed	**Method**	**Date Interval**
O_3	Electrochemical Concentration Cell (ECC)	6 days/week May 16 to August 16
T, RH	bead thermistor & capacitance hygrometer	
winds	GPS tracking	
Half Moon Bay, Ozonesonde Site, (37.505N, -122.484W), San José State University		
Parameters Observed	**Method**	**Date Interval**
O_3	Electrochemical Concentration Cell (ECC)	6 days/week July 24 to August 17
T, RH	bead thermistor & capacitance hygrometer	
winds	GPS tracking	
Chews Ridge, Mountain Inflow Monitoring Site, (36.306N, -121.567W), University of California Davis/Monterey Institute for Research in Astronomy		
Parameters Observed	**Method**	**Date Interval**
O_3	UV-Absorption (2B-Tech)	May 1 to August 31
NO/NO_x	Chemiluminescence/Photolysis (TECO w Mo convertor)	
T, RH, Wind speed/direction	Davis Instruments	
Mooney Aircraft, University of California Davis/Scientific Aviation, Inc.		
Parameters Observed	**Method**	**Date Interval**
O_3	UV-Absorption (2B-Tech)	June 2-4, June 28, July 24-26, July 27-29, August 4-6, August 12-18
NO/NO_x	Chemiluminescence/Photolysis (EcoPhysics/Air Quality Design, Inc.)	
$CH_4/CO_2/H_2O$	Picarro CaRDS	
T, RH	Vaisala Instruments	
Horizontal Winds	In-house dual GPS (Hemisphere)	
AJAX, NASA Ames Research Center		
Parameters Observed	**Method**	**Date Interval**
O_3	UV-Absorption (2B-Tech model 205)	May 12, May 19, June 3, June 15, June 23, July 6,
$CH_4/CO_2/H_2O$	IR Cavity Ringdown Spectroscopy (Picarro model G2301-m)	
3-D winds, P, T	NASA Ames MMM package	

		July 21, & July 28 2016
HCHO	non-resonant laser-induced fluorescence	May 12, June 15, June 23, July 21, July 28
TOPAZ ozone lidar, NOAA Earth System Research Laboratory		
Parameters Observed	**Method**	**Date Interval**
O_3, Aerosol backscatter profiles	Differential Absorption Lidar (DIAL)	May 27 – June 18, July 18- August 7
O_3	UV-Absorption (2B-Tech model 205)	May 27 – August 8
T, p, RH, wind speed and direction	Airmar 150WX weather station	May 27 – August 8

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Figures

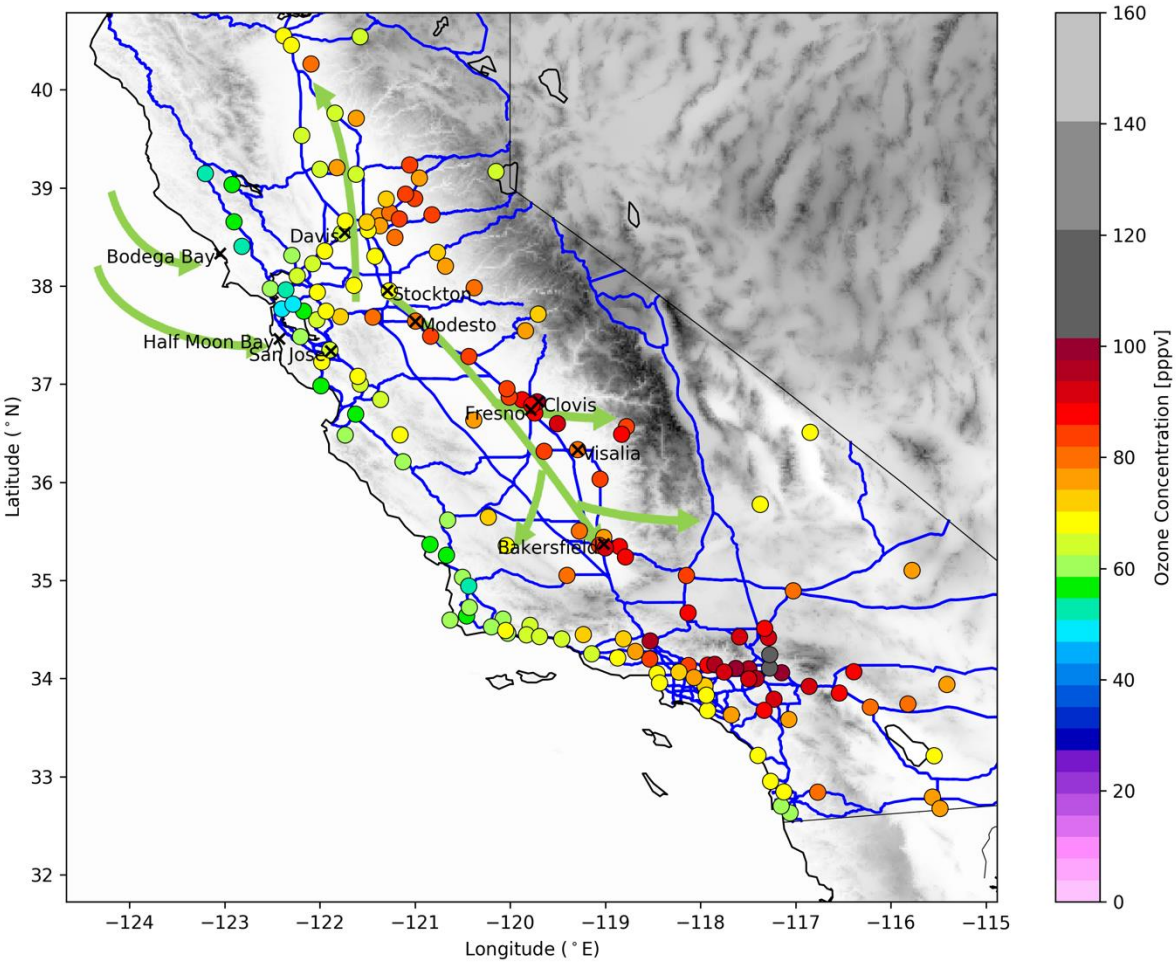


Figure 1. 2016 ozone design values (ODV) across the state of California from the CARB surface air quality network. The ODV is defined as the three year running mean of each year's fourth highest maximum daily 8-hr average (MDA8) ozone concentration. Thin blue lines represent major highways, and thick green arrows show the typical daytime air flow near the surface during the warm season.

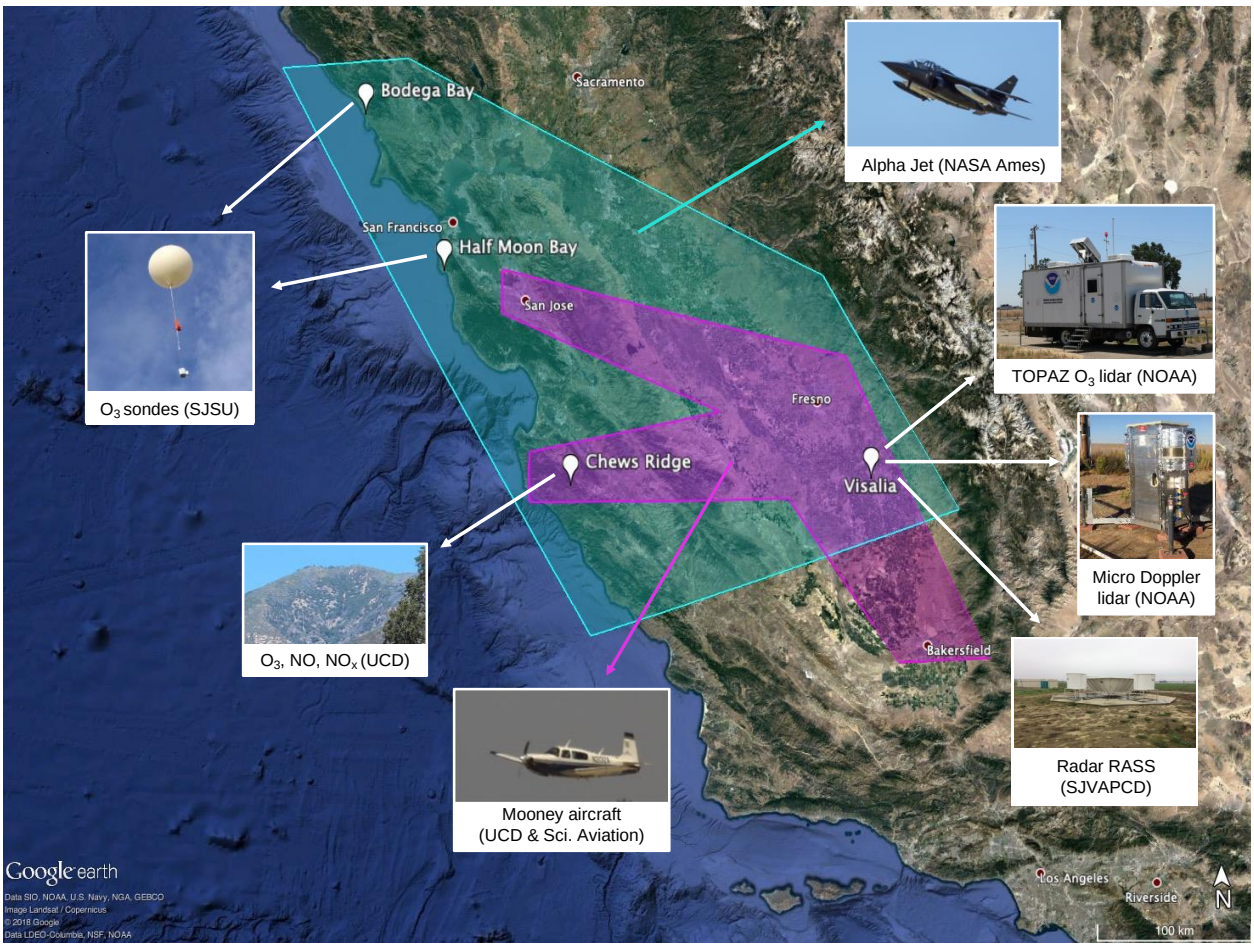


Figure 2. Overview of CABOTS study domain and measurement platforms ranging from daily ozonesondes launched at the two coastal sites (Bodega Bay & Half Moon Bay) to the NOAA TOPAZ lidar in Visalia. The green and purple polygons represent the approximate domains surveyed by the NASA Alpha jet and Scientific Aviation, Inc. Mooney aircraft, respectively.

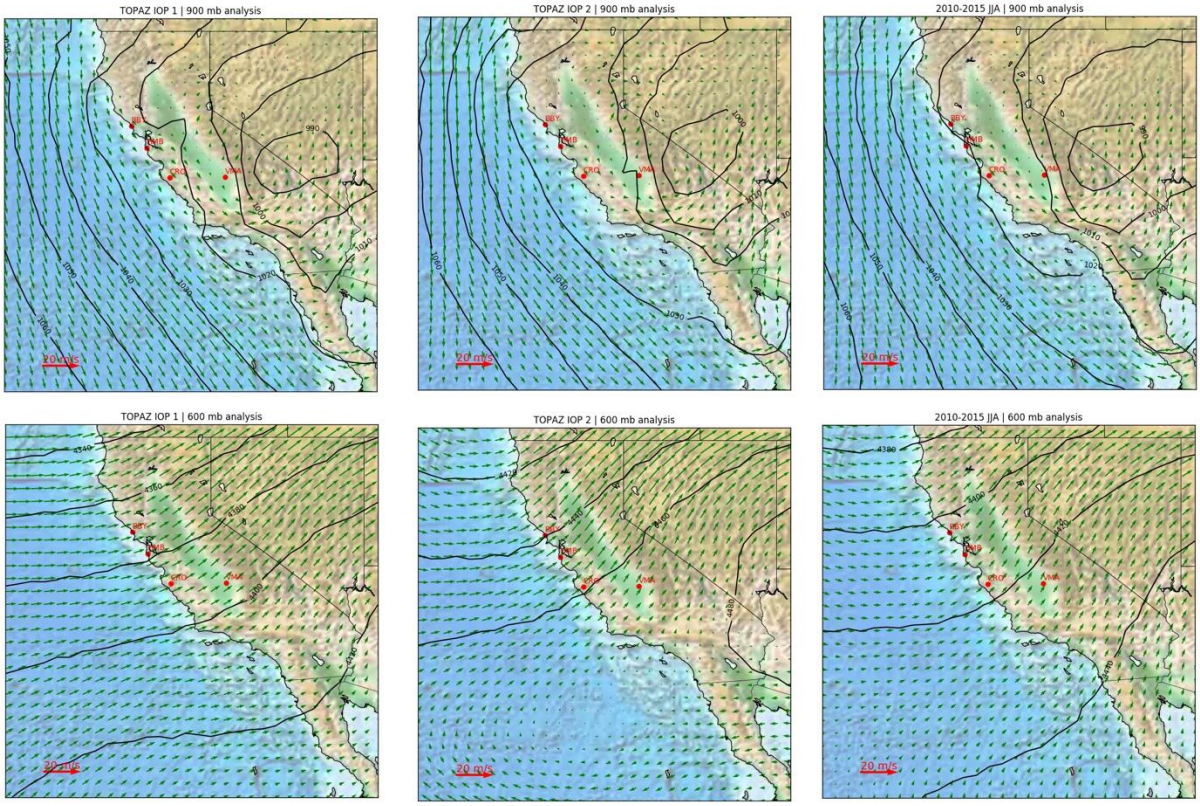
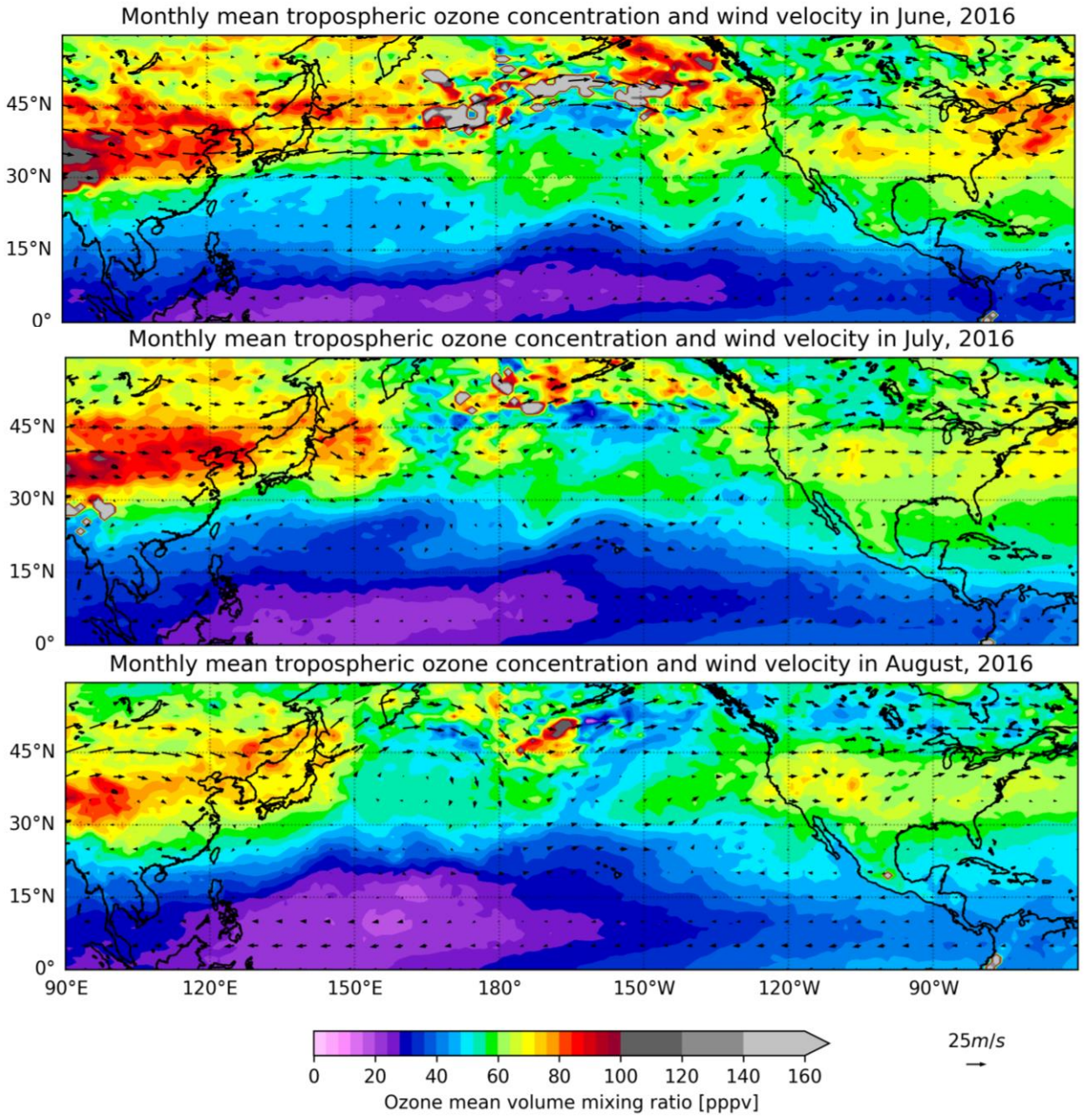


Figure 3. Geopotential heights for CABOTS at 900 hPa (top row) and 600 hPa (bottom row) for IOP1 (left), IOP2 (middle), and climatological average for June – August 2010-2015 (right). The two pressures are near the altitudes of maximum correlation found between O_3 observations at Bodega Bay (BBY) and Visalia Municipal Airport (VMA).



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779 Figure 4. NASA Goddard tropospheric column O_3 product from Aura OMI/MLS and the NCEP

780 Reanalysis winds at 300 hPa for the summer months of the CABOTS project.

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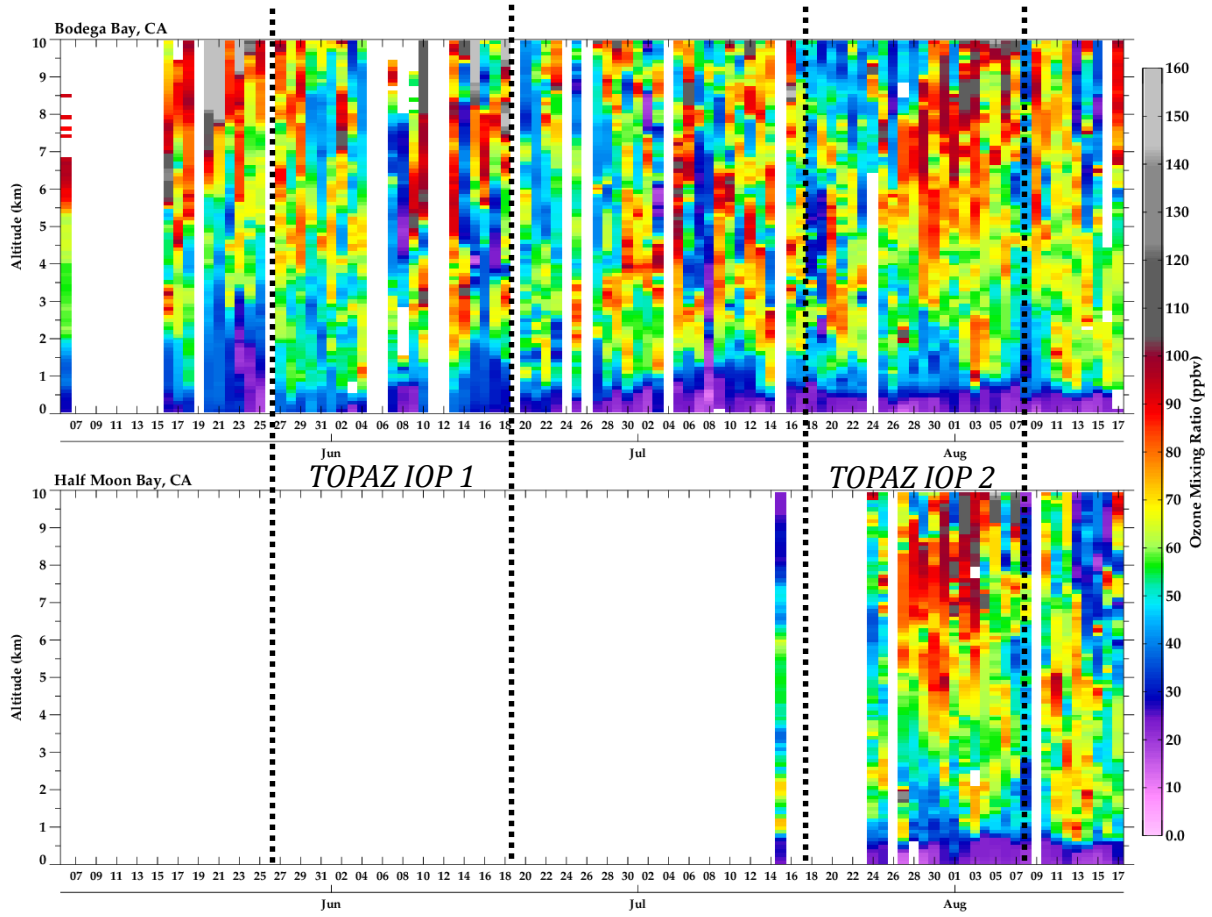

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784 Figure 5. Ozonesonde profiles measured at Bodega Bay and Half Moon Bay during CABOTS

785 (vertical dashed lines indicate TOPAZ IOPs).

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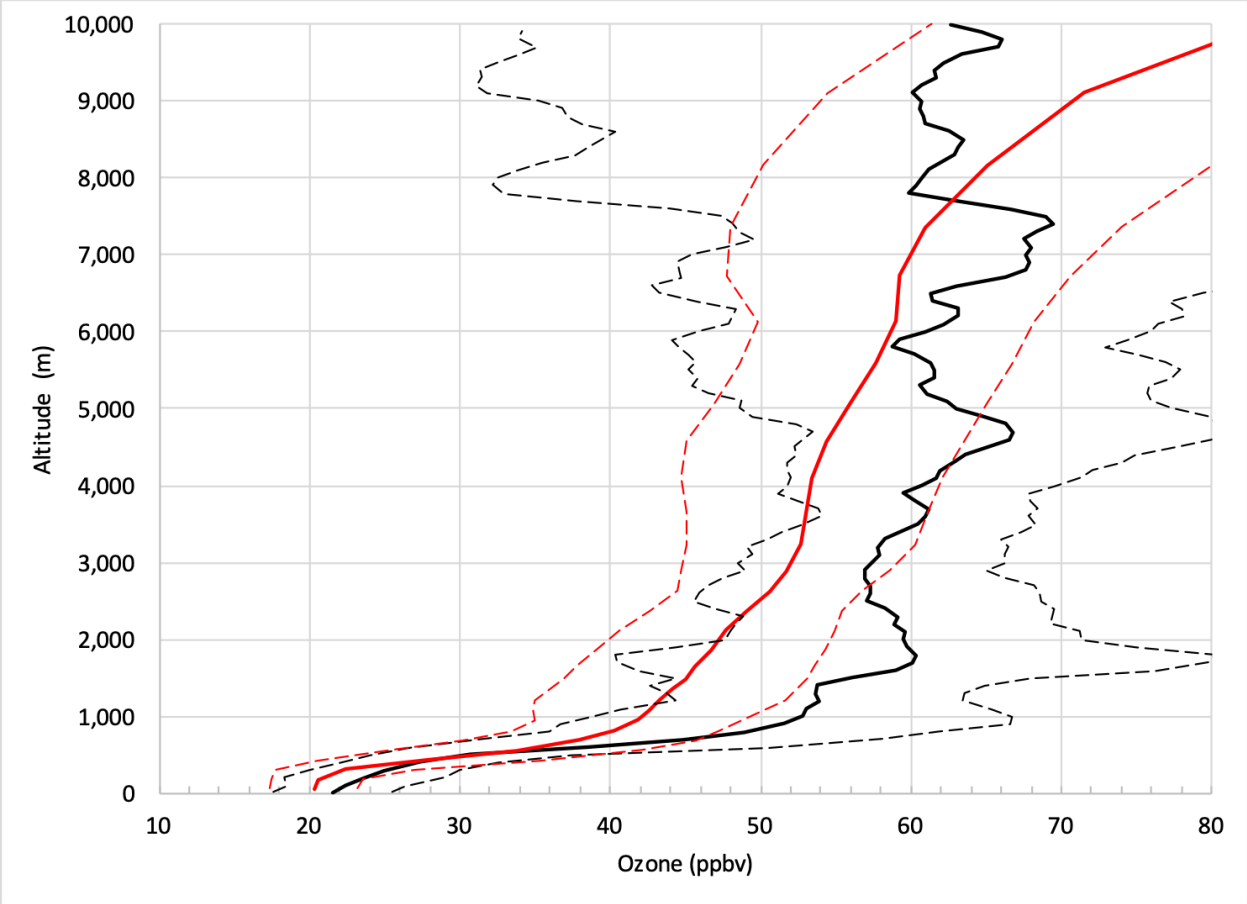

Figure 6. Mean (solid line) and ±1 standard deviation (dashed lines) of the observed ozone (black) and the model ozone from MOZART (red) above the Half Moon Bay site. The comparison is for 24 days between mid-July and mid-August 2016.

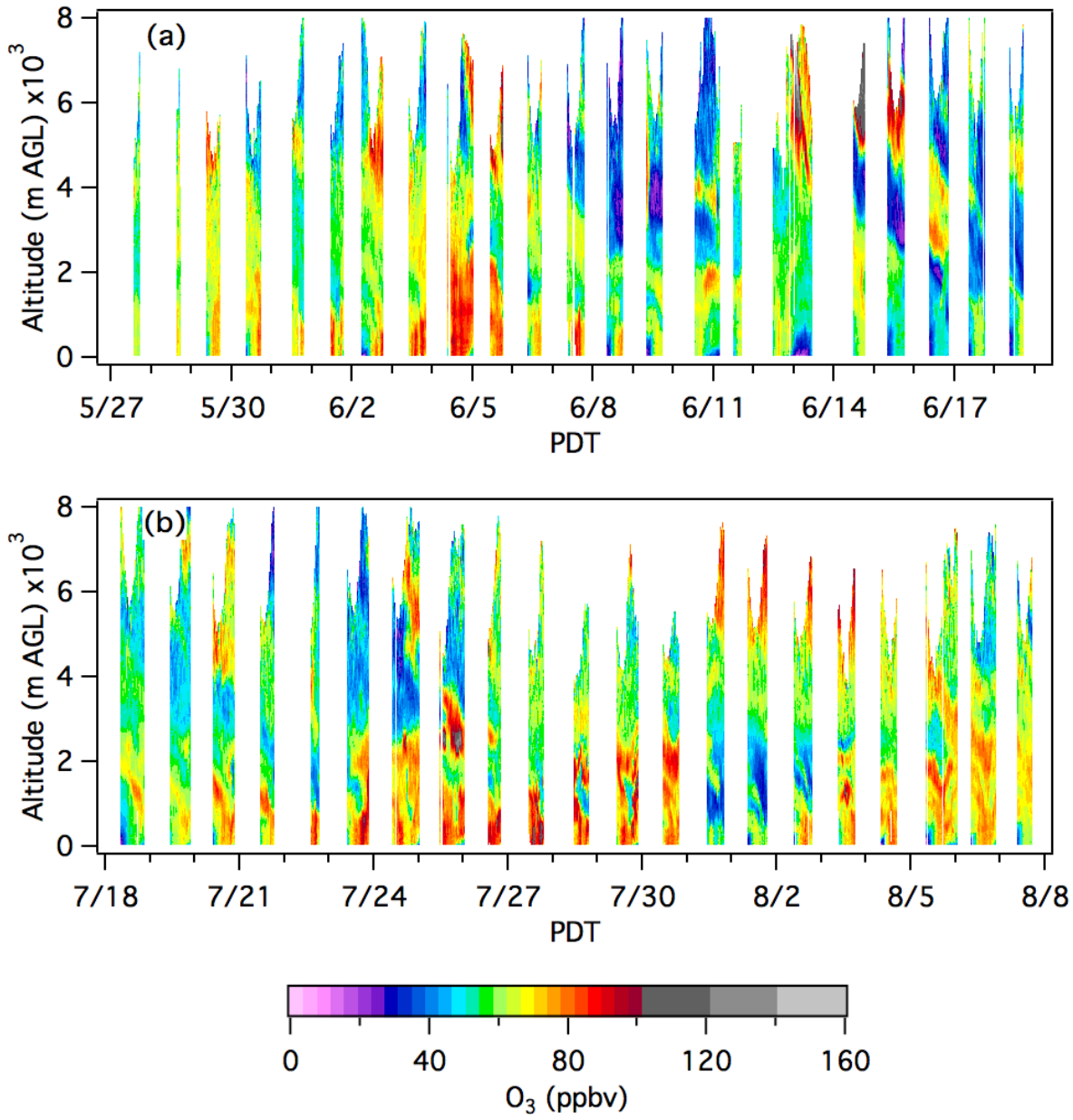

Figure 7. TOPAZ O_3 lidar profiles for (a) IOP1 and (b) IOP2.

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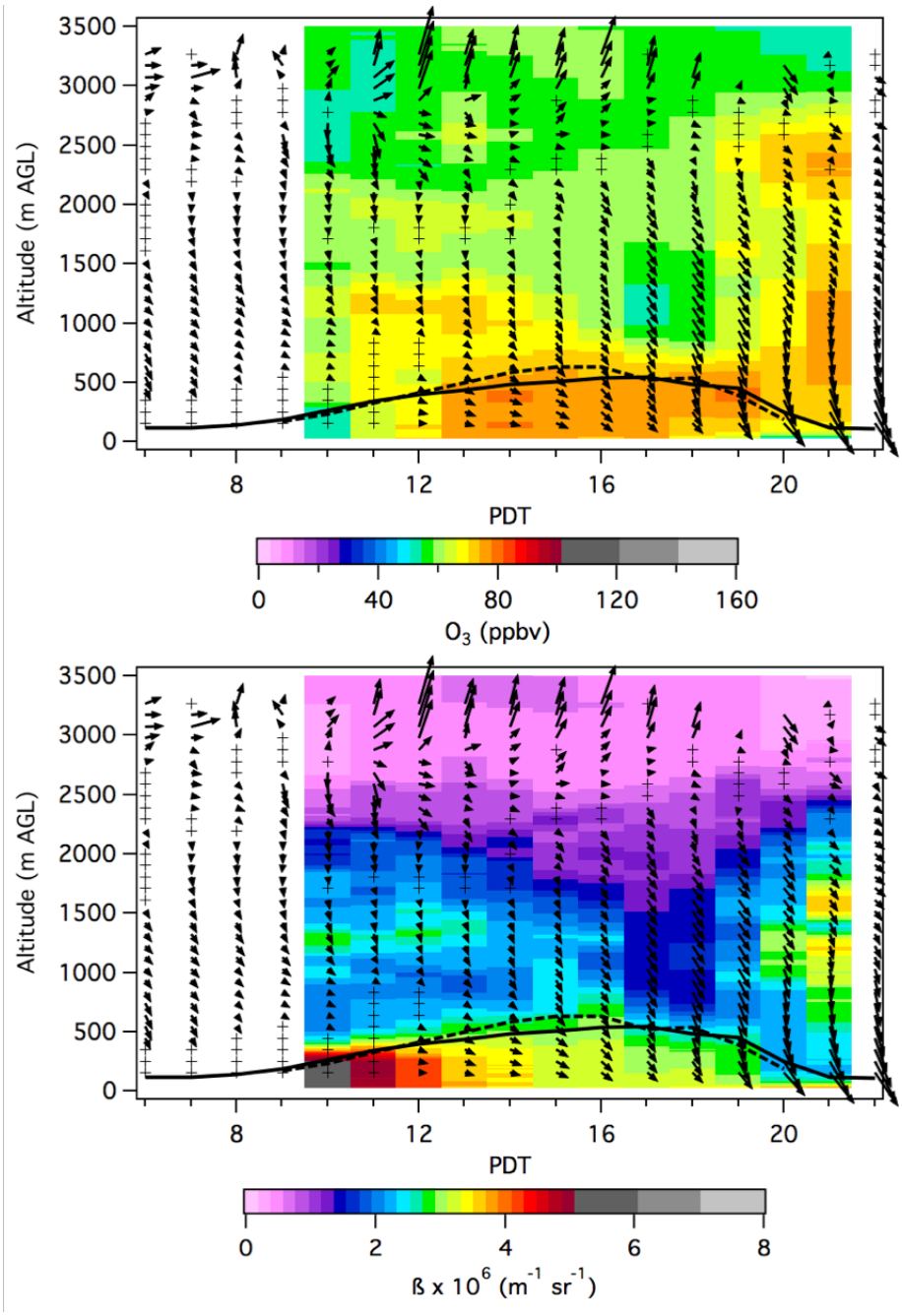

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805 Figure 8. Mean diurnal profiles of TOPAZ ozone (top) and backscatter (bottom), along with winds

806 from the co-located SJVAPCD radar profiler and RASS during the second CABOTS IOP. The black

807 lines show the ABL height inferred from the RASS temperature profiles (solid) and a co-located
808 NOAA Doppler lidar (dashed).

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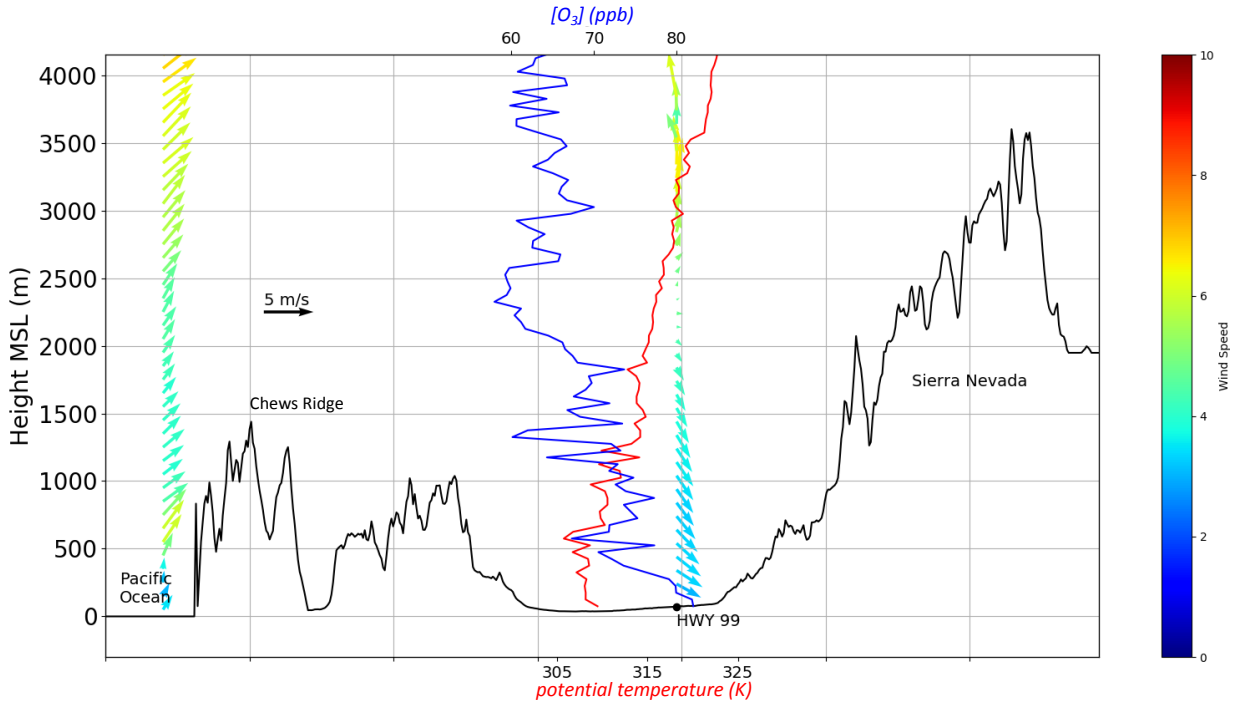

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811 Figure 9. Cross section of SJV through the Chews Ridge site passing across CA Hwy 99 between
812 Madera & Merced (approximately 100 km north of Visalia). Vector average winds as a function
813 of height and mean scalar wind speed (colors), shown with respect to north directed straight up,
814 from the coastal wind profile observed by sondes at Half Moon Bay (approximately 100 km north
815 of Chews Ridge), and at Visalia by the RASS for 12:00-16:00 PST July-August 2016. Ozone (blue)
816 and potential temperature (red) are averages from the afternoon Mooney aircraft flights profiling
817 throughout the region.

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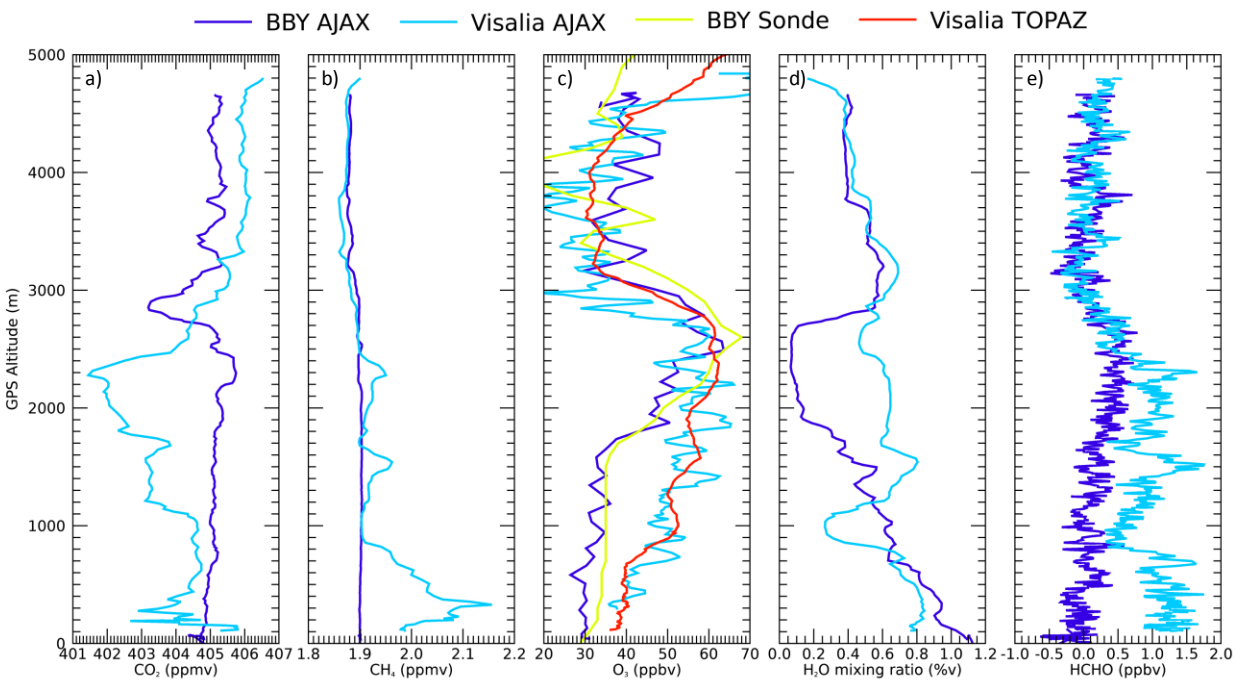

Figure 10. June 15, 2016 AJAX profiles taken at BBY and VMA (dark and light blue, respectively) of (a) CO_2, (b) methane, (c) ozone, (d) water vapor, and (e) formaldehyde. Aircraft profiles were measured at 10:00 PDT at Bodega Bay and half an hour later at Visalia. The profiles of ozone measured by the ozonesondes at Bodega Bay (yellow) and from the TOPAZ lidar at Visalia (red) are superimposed in (c) for comparison.

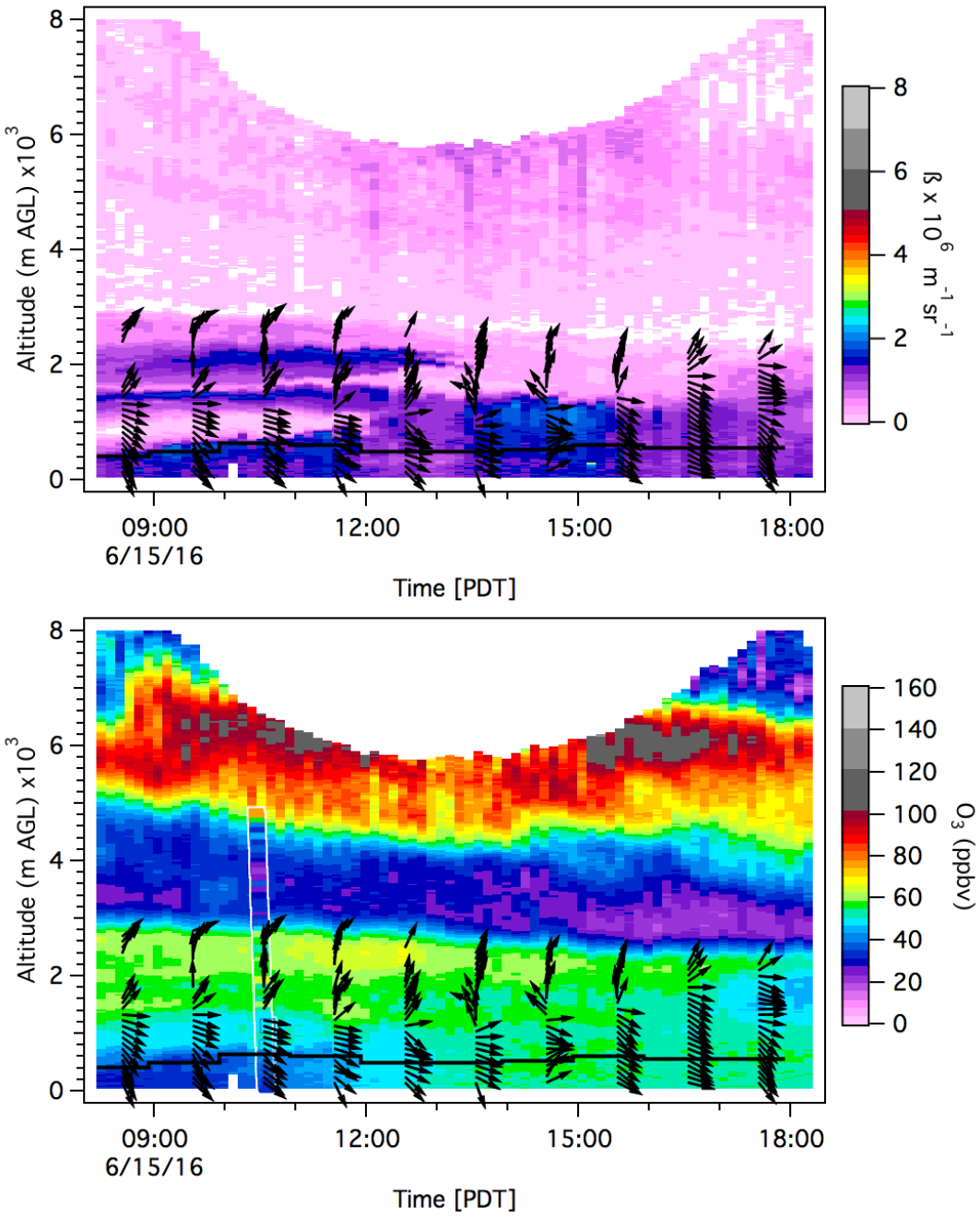

Figure 11. Time-height curtain plots of the TOPAZ aerosol backscatter (top) and ozone (bottom) above the VMA with the co-located profiler winds and RASS boundary layer height. The AJAX ozone profile above the VMA is superimposed on the ozone plot (~10:30-40 PDT).

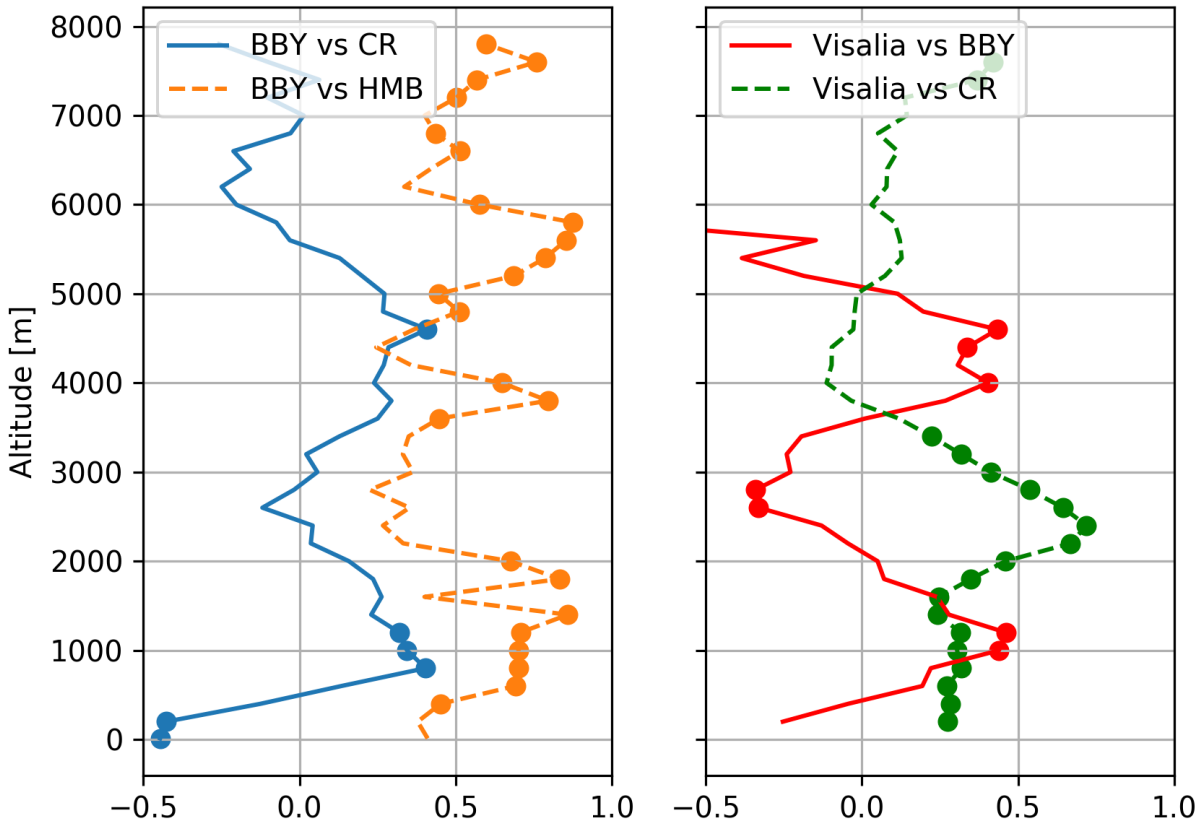

Figure 12. (Left) Correlation coefficients of Bodega Bay ozonesondes observations with coincident Chews Ridge surface ozone at 1.5 km (blue line), and Half Moon Bay ozonesonde measurements (orange dashed line). (Right) Correlations of TOPAZ lidar data with Bodega Bay ozonesondes (red line) and Chews Ridge surface (green line). Circles represent correlations with p-values < 0.05.

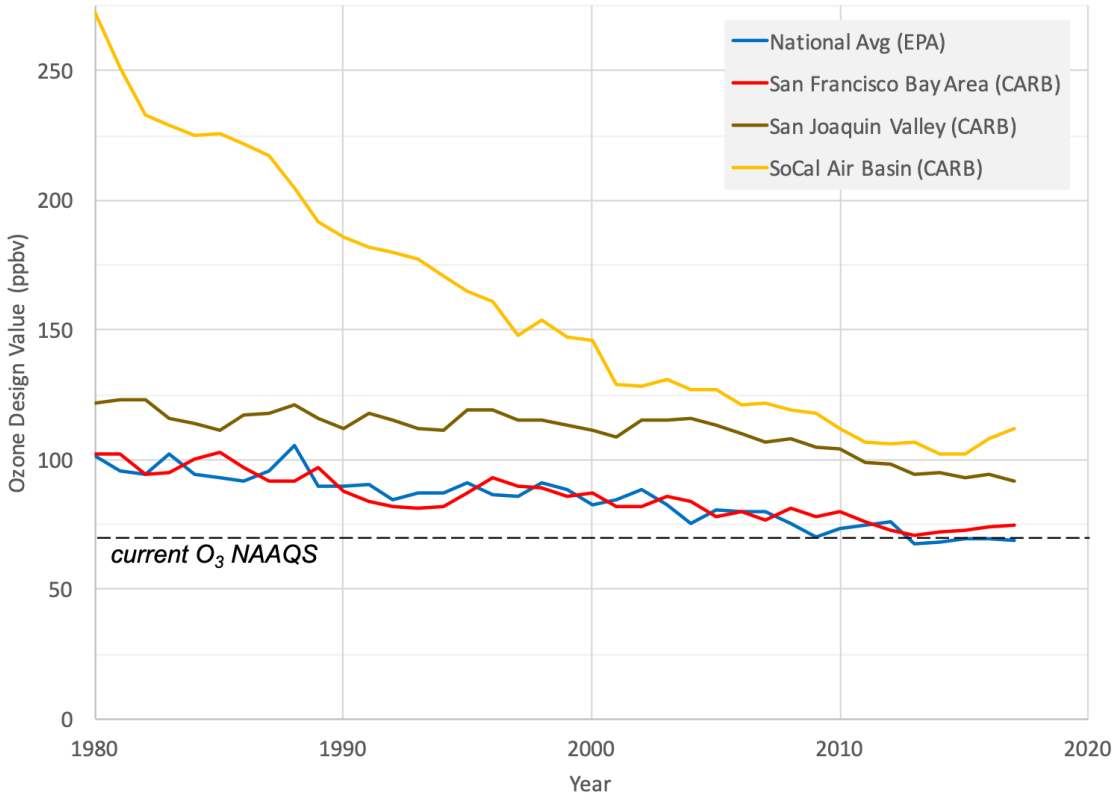

Figure SB1. The decadal trends in ozone design values from three different air basins in California compared to the national average and the current National Ambient Air Quality Standard (NAAQS.)

Figure SB2. Three views of the Soberanes Fire taken during CABOTS. (top left) View from Scientific Aviation, Inc. Mooney aircraft looking northward from Chews Ridge. (bottom left) View towards the southwest from the Oliver Observing Station on Chews Ridge (photo credit: Chris Reed, observatory caretaker). (right) MODIS satellite image of the fire from July 26, 2016 (https://earthobservatory.nasa.gov/images/88483/wildfire-along-the-california-coast).

Final responses to second round of reviews for “The California Baseline Ozone Transport Study (CABOTS)“ submission to BAMS

2nd Review of "The California Baseline Ozone Transport Study (CABOTS)" by Faloona et al.
MS Number: BAMS-D-18-0302

Summary:
I originally recommended that this paper be rejected, largely based on my question whether BAMS is the appropriate journal for this paper. Since my recommendation was not followed, I assume that the editor does find the paper appropriate. Further, the paper has been shortened and the discussion improved. I recommend publication after the following minor issues are addressed.

Minor issues:
1) Lines 156-157 mention “... near-daily ozonesonde profiles collected at one site on the coast and ozone ....” but the figure under discussion shows two sites. I suggest that the discussion in this sentence be clarified.

Changed to, “The core measurements included near-daily ozonesonde profiles collected at two sites along the coast and an ozone lidar data set collected in Visalia, a city of nearly 140,000 residents located deep within the San Joaquin Valley, approximately 60 km southeast of Fresno.”

2) Line 233: “comprised” would be better replaced by “composed”.

Changed to “composed”.

3) Lines 297-300: In the discussion of Figure 6, the authors may wish to note that the observations show only a small mean vertical ozone gradient between 2 and 10 km (<1 ppb/km) while the model shows a significantly stronger gradient (~3.3 ppb/km). The authors may also wish to note that the median of the observations approaches 60 ppb in the 1.5 to 3 km altitude range, which is only 10 ppb below the current NAAQS. The + 1 std. dev is close to the NAAQS, but the NAAQS represents above a + 2 std. dev. value. Hence, if the air between 1.5 and 3 km were transported to the surface of the Central Valley without dilution, O3 above the NAAQS would occasionally be observed at surface sites.

Thank you for the additional comments on the model, but based on some critical comments about the MOZART model that we received from another technical reviewer of this manuscript, we feel that it would not be helpful to include more specific analysis of the model results here. Moreover, it is our opinion that transport of air aloft into the boundary layer without mixing is not a physically relevant scenario, not to mention that in order to violate the NAAQS it would have to persist for at least 8 hours. But considering your points more carefully, we have decided to include a sentence to draw attention to the fact that elevated surface *sites may experience these higher ozone levels even in the absence of contributions from local sources. Consequently,*

we have added a sentence to that paragraph that reads, "Furthermore, in agreement with the findings presented in Figure 6 of frequent ozone concentrations well above 60-70 ppb (mean + 1 standard deviation) at 1.5 km altitudes and above, an analysis of the ozone time series collected at Chews Ridge from 2012-2014 determined that the ODV for this remote site located in a National Forest in the coastal mountains was 70.5 ppbv, technically in violation of the NAAQS."

4) Lines 314-315: This description is confusing, at least to this chemist. Northwesterly winds have vectors that point to the bottom and right, so perhaps the top represents south, and the right side represents west? Please clarify.

It is meteorological convention to name the wind based on the direction from which it blows. This is opposite to the oceanographic convention of naming a current by the direction it is moving toward, so confusion in the Earth Sciences is easy. The wind vectors under consideration are pointing to the right and then bottom/right, so they are blowing from the west(erly) and northwest(erly) directions. We have added the following clause for absolute clarity, "The wind vectors show the horizontal wind direction with the top of the plot representing north and the right side east (arrows that point to the right in the figure therefore represent westerly *winds.)*

5) Line 329: O_3 depletion overnight in a shallow nocturnal boundary layer does not necessarily require nitrate production. NO_2 formation, surface deposition, and downslope flow of cleaner air may be enough to explain this?

Yes, line 329 states that O_3 depletion is occurring predominantly due to nitrate production ***and*** *dry deposition, both of which were estimated in the Caputi et al. (2019) reference wherein we attempted to quantify all these terms overnight (including advection) based on a simplified odd oxygen (O_3+NO_2) budget so as not to include titration. While not covered in this work, we measured ~2 ppb NO and ~7 ppb NO_2 in the afternoons in this region, so we expect the significant lowering of O_3 near the surface in the dark to be due mostly to reaction with NO_2, not simply titration. We find the average loss of O_x overnight due to nitrate production to be -2.7 ppb/h, compared with -1.2 ppb/h due to dry deposition and -0.2 ppb/h for horizontal advection in addition to +2.8 ppb/h from vertical mixing.) We were hoping that readers interested in the overnight ozone chemistry would read our further analysis presented in Caputi et al. (2019).*

6) Line 330-333: The prevalence of elevated ozone (>70 ppb, orange colors) between 500 – 2,500 m in these average profiles of Figure 8 is cited as evidence that ABL air is lofted into the buffer layer above due to daytime slope flow along the eastern flank of the valley during its up-valley progression to the southeast. Is this really correct? Figure 6 shows that mean ozone coming ashore between 500 and 2500 m can average 60 ppb. Very little additional O_3 from ABL air would be required to reach 70 ppb. Please discuss how persuasive is the evidence provided by these relatively small enhancements in O_3.

First, the average of the profile in the layer between 500 – 2,500 m in Figure 6 is much closer to 50 ppbv, which leads to enhancements of ozone within the valley at those altitudes of over 20 ppb, which is indeed significant. Second, enhancements over the baseline in the much deeper

buffer layer are proportionally larger than a mere additive ppb, given that the ABL has only about one-third of the mass of the buffer layer. Third, we go to greater lengths to support this assertion in the discussion surrounding figure 10, which clearly shows discrete, elevated layers of other corroborating scalars above the valley within the BuL. And finally, this effect is well established in the literature, including references that we have cited (Leukauf et al., 2016; Fast et al., 2012) and others we did not have the space to cite (Rotach et al., 2015; Serafin et al., 2018). Nevertheless, we have added a sentence to draw attention to the continuing analysis in the manuscript, "Further evidence of this slope venting is presented for other scalars in the discussion surrounding Figure 10 that follows."

7) Line 341-342: (Same comment as 4 above.)

Hopefully the added comment in lines 314-315 help clarify.

8) Line 392-393: Low aerosol and high ozone might indicate stratospheric source rather than Asian? Please clarify.

*The sentence on lines 392-393 states that the low aerosol and high ozone are **consistent** with transported Asian pollution. Looking back at the original submission's Figure 13, it illustrates the AJAX flight data from June 15, the same day being discussed here in the current Figure 11. That figure, which was eliminated at the suggestion of reviewers, showed that the layer near 5 km above Visalia showed signs of elevated levels of CO_2, HCHO, and CH_4, thus providing further evidence that its origin is not the stratosphere. We have added a statement to the following sentence to make this clear in the new draft, "The superimposed AJAX profile in Figure 11 shows that the aircraft sampled the very bottom of the transported pollution layer (not shown here, but exhibiting enhanced CO_2, HCHO, and CH_4) before descending into the clean layer."*

9) Line 411: "... just below 1 km ..." is better described as "... 0.8 to 1.2 km ..."

Changed.