Spatial Variation of Vertical Ozone Distribution over California

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LIST of ACRONYMS and CHEMICAL FORMULAE

3-D: three Dimensional
AJAX: Alpha Jet Atmospheric eXperiment
BBY: Bodega Bay
CAA: Clean Air Act
CABOTS: California Baseline Ozone Transport Study
CalNex: California Nexus of Air Quality Climate Experiment
CARES: Carbonaceous Aerosol and Radiative Effects Study
CARB: California Air Resources Board
CH4: methane
CMAQ: Community Multiscale Air Quality Model
CO2: carbon dioxide
ECMWF: European Centre for Medium-Range Weather Forecasts
EPA: Environmental Protection Agency
ESRL: Earth System Research Laboratory
FLEXPART: FLEXible PARTicle dispersion model
GPS: Global Positioning System
GSFC: Goddard Space Flight Center
HMB: Half Moon BayNAAQS: National Ambient Air Quality Standard
H2O: water
HCHO: formaldehyde
HYSPLIT: Hybrid Single-Particle Lagrangian Integrated Trajectory
ICARTT: International Consortium for Atmospheric Research on Transport and Transformation
KVIS: Visalia Municipal Airport
LPDM: Lagrangian Particle Dispersion Model
LT: Lower Troposphere
MDA8: Maximum Daily 8 hour Average ozone
MERRA: Modern Era Retrospective analysis for Research and Applications
MMS: Meteorological Measurement System
MT: Mid-Troposphere
NASA: National Aeronautics and Space Administration
NIST: National Institute of Standards and Technology
NOAA: National Oceanic and Atmospheric Administration
O3: ozone
ODV: Ozone Design Value
PBL: Planetary Boundary Layer
PFA: PerFluoroAlkoxy polymer
PPBV: Parts Per Billion by Volume
PPM: Parts Per Million
Pt Sur: Point Sur
PTFE: PolyTetraFluoroEthylene
PV: Potential Vorticity
SI: Stratospheric Intrusion

SJSU: San Jose State University
SJV: San Joaquin Valley
STE: Stratosphere Troposphere Exchange
THD: Trinidad Head
TOPAZ: Tunable Optical Profiler for Aerosol and oZone lidar
US: United States
UT/LS: Upper Troposphere /Lower Stratosphere
UTC: Coordinated Universal Time
UV: UltraViolet
WMO: World Meteorological Organization
WRF-Chem: Weather Research and Forecasting model coupled with Chemistry
WRF-STILT: Weather Research and Forecasting Stochastic Time Inverted Lagrangian Transport

Abstract

The National Ambient Air Quality Standard (NAAQS) for ozone (O_3) was lowered by the United States Environmental Protection Agency (U.S. EPA) in 2015 from 75 ppbv to 70 ppbv. Meeting this stricter air standard in regions like the San Joaquin Valley (SJV) is challenging due to complex topography, meteorology, and local emissions that affect the ground-level O_3. There is also the possibility that trans-Pacific baseline O_3 is contributing to the total observed O_3 in the SJV, where the term "baseline ozone" is defined as the O_3 observed at a site when it is not influenced by recent, locally emitted or produced anthropogenic pollution (Cooper et al., 2011). The significance of baseline O_3 in the SJV is unquantified, and the California Baseline Ozone Transport Study (CABOTS) was conducted by the California Air Resources Board during the spring and summer of 2016 to address this knowledge gap.

The AJAX airborne science project based out of NASA Ames Research Center performed eight science flights in coordination with the CABOTS campaign. Many of these flights included a series of vertical profiles (~ 0 – 5 km) distributed roughly along either a north/south or east/west transect. Some flights also connected the fixed-location measurements at Visalia (TOPAZ ozone lidar, operated by NOAA) and Bodega Bay (ozonesondes launched by San Jose State University). AJAX measured O_3, CO_2, CH_4, H_2O, HCHO, and 3-D winds on each flight. Under this contract, analysis of data collected by AJAX over this period investigated the horizontal and vertical scales of correlation between air sampled offshore and in the SJV. Vertical profiles, time series, back trajectories, tracer-tracer correlations, and comparisons with the fixed-location measurements were utilized to identify the sources of O_3 features observed during these flights and to assess the overall impact of baseline O_3 in the SJV.

Executive Summary

The 2016 California Baseline Ozone Transport Study (CABOTS) was designed by the California Air Resources Board (CARB) to investigate questions of the content and daily variability of the vertical distribution of ozone as air masses enter the State from the Pacific and the extent to which long-range transported ozone mixes down to surface sites in the San Joaquin Valley (SJV). CABOTS was conducted during the spring and summer, the time of year known to favor meteorological conditions which lead to both long-range transport of pollution and stratosphere-troposphere exchange of ozone-rich air. In coordination with the CABOTS campaign, the NASA Alpha Jet Atmospheric eXperiment (AJAX) team, an unfunded collaborator, conducted eight flights of an airborne trace gas payload measuring ozone (O_3), carbon dioxide (CO_2), methane (CH_4), water (H_2O), formaldehyde (HCHO), and 3-dimensional winds. Many of these flights included a series of vertical profiles (~ 0 – 6 km) distributed roughly along either a north/south or east/west transect. Whenever possible, flights also sampled at two fixed-location profile sites operated by other CABOTS team members: Visalia (TOPAZ ozone lidar, operated by NOAA) and Bodega Bay (ozonesondes launched by San Jose State University).

This project completed the calibration and the quality assessment of the *in situ* trace gas data collected by the NASA AJAX group during CABOTS. Datasets including time and aircraft position have been delivered electronically to CARB as comma-separated-value text files. Also under this contract, analysis of data collected by AJAX investigated the horizontal and vertical scales of correlation between air sampled offshore and in the SJV. Vertical profiles, time series, back trajectories, tracer-tracer correlations, and correlations with various fixed-location measurements were utilized to identify the sources of O_3 features observed during these flights and to assess the overall impact of baseline O_3 in the SJV. Surface level ozone values throughout California were examined for evidence of influences common to those affecting the ozone sampled aloft. No statistically significant correlations were found, indicating that the variance in surface O_3 on flight days cannot be attributed to the same factors that influence the variance of O_3 sampled at altitudes up to 6 km by AJAX.

Co-located complete profiles (>6 – 0 km) of AJAX and ozonesonde data show a high degree of correlation with no significant bias. The good agreement between these two different

techniques when sampling essentially the same air lends confidence to comparisons of the two data sets when collected farther away from each other. At longer separation distances, sonde profiles at Bodega Bay (BBY) typically correlated well, but not statistically significantly, with profiles collected by AJAX ~240 km down the coast at Point Sur. This suggests that broadly similar ozone vertical profiles can be presumed to exist at unsampled near-shore locations along the northern and central California coast, but a strict predictive capability is unlikely.

The source of ozone coming into California (36 – 42° N, 118 – 125° W) during the spring and summer of 2016 has been assessed using trajectory modeling and other methods. Three categories of influence were identified: stratospheric intrusion, long-range transport of ozone, and local influence. In general, the upper troposphere and lower stratosphere (UT/LS) was a source of ozone to California in May and June, with a decreasing influence as the campaign progressed into summer. Long range transport from ozone source regions in Asia and the western Pacific was also commonly observed on flight days in May and June. Flights later in the campaign demonstrated local influences, including emissions from the Soberanes and other wildfires in late July.

1. Introduction

In the U.S., the Clean Air Act (CAA) requires that the EPA establish National Ambient Air Quality Standards (NAAQS) for O_3. In October 2015, the U.S. EPA revised the NAAQS for O_3 from 75 ppbv to 70 ppbv (EPA, 2015). A location is in violation of the NAAQS when the 3-year average of the fourth highest daily maximum 8 h average O_3 (MDA8), called the "O_3 design value (ODV)" exceeds a 70 ppbv threshold. The continual lowering of the NAAQS threshold combined with regulatory efforts aimed at decreasing local emissions increases the relative importance of understanding baseline O_3. In particular, understanding the contribution of aloft O_3 to NAAQS violations is critical.

To accurately quantify and understand the impact of baseline O_3, many studies have utilized balloon launches, ground-based lidars, and aircraft to measure incoming ozone. Ozonesondes launched along the California coast as well as from inland locations have been used to compare airflow and to find any correlation between offshore and onshore air (Parrish et al., 2010; Cooper et al., 2011). These studies have observed that that airflow above 6 km has minor impact on surface air quality, while airflow between 0-3 km can impact the surface. It was determined that O_3 precursors at these altitudes can be linked to East Asia (Cooper at al., 2010). Ground-based lidars (Langford et al., 2017) have also been used to measure baseline O_3 at high and low elevations to better understand the spring O_3 spikes caused by East Asian air pollution. Aircraft-borne lidar measurements (Langford et al., 2012) made by the National Oceanic and Atmospheric Administration's Earth System Research Laboratory (NOAA ESRL) during the CalNex/CARES campaigns in June of 2010 frequently found layers of elevated O_3 between 2 and 4 km above the northern SJV which could be attributed to descent of pollution from the free troposphere in dry airstreams or tropopause folds. Other assessments included the use of an aircraft (Ryoo et al., 2017; Yates et al., 2017) to connect ground-level and free-tropospheric O_3. Modeling of backward air trajectories were then utilized to identify the origin of the air masses. Several modeling studies (Brown-Steiner and Hess, 2011; Cooper et al., 2004; Lin et al., 2012) have shown that the transport of O_3 across the Pacific from Asia is fundamentally different in spring and summer.

Despite this variety of previous work, there remains a significant level of uncertainty about the contributions of stratospheric and transported O_3 to surface concentrations in the SJV. Much of this uncertainty results from a lack of information about the spatial and temporal variability of the vertical distribution of O_3. Intermittent measurements by ozonesondes or aircraft do not provide the temporal coverage needed to evaluate how well models such as Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) or the Community Multiscale Air Quality Model (CMAQ) can determine the fraction of background O_3 in the surface concentrations measured in the SJV, especially during the spring and summer transport regimes. To address this need, CABOTS produced a three-month dataset of aloft O_3 measurements, collected with sondes and a ground-based lidar, as well as airborne data from the UC Davis team. The AJAX team from NASA Ames Research Center was able to coordinate eight research flights during the CABOTS period.

2. NASA Ames Research Center Contributions to CABOTS

CABOTS, a joint effort between the California Air Resources Board (CARB), NOAA, and San Jose State University (SJSU), was conducted during the spring and summer of 2016 to quantify baseline O_3 in the SJV and elsewhere in California. During CABOTS, the NASA AJAX team, an unfunded collaborator, conducted eight flights linking ozonesonde launches at the coast to lidar measurements in Visalia (see Figure 1 and Table 1). AJAX measured O_3, CO_2, CH_4, H_2O and 3-D winds on each flight. <u>The objective of this project was to calibrate, finalize, and analyze the atmospheric trace gases and related data collected by the NASA AJAX group during the CABOTS of 2016.</u>

The AJAX flight paths were pre-designed based on historical meteorological data of dominant wind flow patterns at 500-mb height contours. For each flight, an analysis of the meteorological forecast allowed the selection of a pre-determined AJAX flight plan to best support the CABOTS intensive. Five different synoptic meteorological patterns were defined based on the 1800 UTC 500-hPa weather maps and the associated general wind patterns in central California. The five patterns were further grouped into three different transport regimes with assigned AJAX flight plans for each regime: offshore ridge; onshore high with offshore trough and zonal flow; and onshore high or offshore trough. Vertical profiles were collected with an ascent/descent rate of 1500 ft/min.

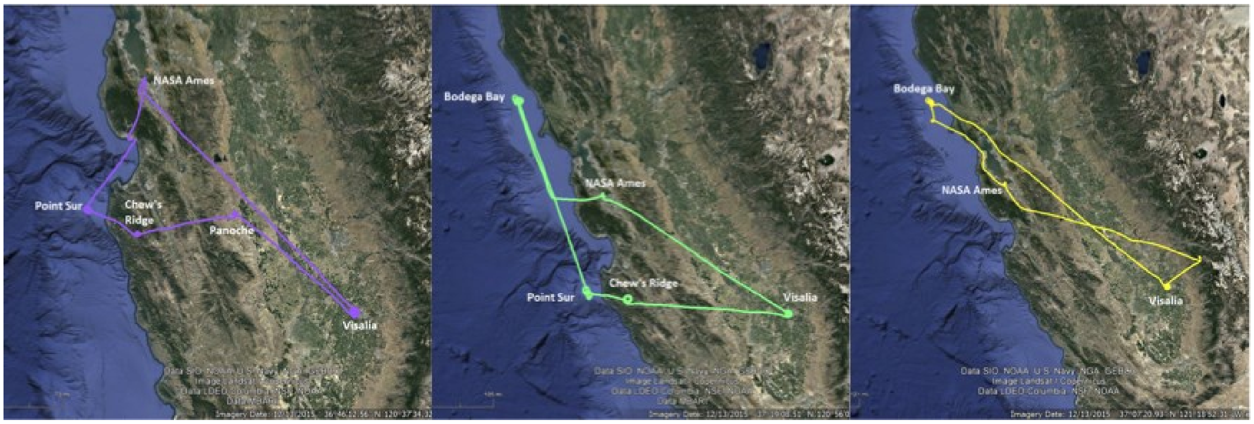

Figure 1. Flight tracks for three AJAX flights during CABOTS. Left: Flight 194, a west-to-east transect with vertical profiles at Point Sur, Chews Ridge, Panoche, and Visalia. Middle: Flight 195, north-to-south and west-to-east transects with profiles at Bodega Bay, Point Sur, Chews Ridge, and Visalia. Right: Flight 191, a north-to-south transect with profiles at Bodega Bay and Visalia.

Table 1. An overview of AJAX flights during the 2016 CABOTS intensive.

Flight Number	Date	Profile Locations	Other Measurements	Peak 1 & 8-hr O_3 in SJV (ppb)
188	May 12	Farallones, CORSE, SHOEY		96 / 82
189	May 19	Trinidad Head		73 / 67
190	June 3	Point Sur, Chews Ridge, Panoche, Visalia	Ozonesonde and TOPAZ	106 / 87
191	June 15	Bodega Bay and Visalia	Ozonesonde and TOPAZ	62 / 59
192	June 21	Point Sur, Chews Ridge, Panoche, Visalia	Ozonesondes	94 / 81
194	July 6	Point Sur, Chews Ridge, Panoche, Visalia	Ozonesonde	83 / 73
195	July 21	Bodega Bay, Point Sur, Chews Ridge, Visalia	Ozonesonde and TOPAZ	103 / 86
196	July 28	Point Sur, BLANC	Ozonesonde and TOPAZ	116 / 92

3. AJAX Data Set

This project completed the calibration and the quality assessment of the *in situ* trace gas data collected by the NASA AJAX group during CABOTS. Finalizing the data collected during eight AJAX flights required applying measured calibration factors to O_3, CO_2, and CH_4 observations to align them with the appropriate international scales (NOAA and World Meteorological Organization (WMO)). Calibration results are included in the metadata provided with each data set. Quality assessment screening removed data points with instrument temperature or pressure anomalies. All data are reported against Coordinated Universal Time (UTC); ambient temperature and aircraft position (latitude, longitude, GPS altitude) are also included.

Measurements of O_3 mixing ratios were made using a commercial O_3 monitor (2B Technologies Inc., model 205) based on ultraviolet (UV) absorption techniques and modified for flight. The dual-beam instrument uses two detection cells to measure simultaneously the UV light intensity differences between O_3-scrubbed air and unscrubbed air to give precise measurements of O_3. The instrument has been modified by upgrading the pressure sensor and pump to allow measurements at high altitudes, including a lamp heater to improve the stability of the UV source, and the addition of heaters, temperature controllers and vibration isolators to control the monitor's physical environment. The air intake is through Teflon tubing (perfluoroalkoxy polymer, PFA) with a backward-facing inlet positioned on the underside of the instrument wing pod. Air is delivered through a 5 µm polytetrafluoroethylene (PTFE) membrane filter to remove particles prior to analysis. The ozone monitor has undergone thorough testing in the laboratory and in a temperature and pressure chamber (that simulates flight conditions) to determine its linearity (performance is linear over 0-300 ppb), precision (typically 1.2 ppb), zero-offset variability (1.4 ppb), repeatability (0.6 ppb) and dependence on pressure (2.2 ppb). The overall accuracy (root mean square of all 1-sigma errors) of the O_3 instrument is determined to be 3 ppb.

A commercial cavity ring-down instrument (Picarro Inc., model G2301-m) forms the basis of the flight instrument which measured CO_2, CH_4, and H_2O mixing ratios. CO_2 and CH_4 mixing ratios are derived from their absorption at selected spectral lines, and simultaneous H_2O measurements are used to correct for water interferences and to provide dry-air-equivalent mixing ratios. The original instrument has been physically reconfigured to fit the space available on the aircraft, and the original thermal management scheme was modified by improving air circulation and by adding an insulating blanket. A 0.5-µm-pore filter was placed after the stainless steel inlet to protect the optics inside the cavity from deposition of particles. The in-flight measurements of CO_2, CH_4 and H_2O are continuously logged at a frequency of ∼3 Hz and post-processed as 10-point averages reported at ∼3 sec intervals. Linearity of the instrument is checked using at least three Scott-Marrin standard gases covering a range of ~70 ppm in CO_2 and ≥10 ppm in CH_4. For CABOTS, five secondary standards were used to confirm instrument linearity (cyl # LL119173, LL106490, LL106469, LL104109, LL123405; Reports of Analysis available from PI). It is important that the optical cavity (sample) pressure of the instrument is maintained at 185 hPa; deviations from this value cause a reduction of instrument precision. Thus, when the recorded point-to-point change in cavity pressure is more than 2-σ (standard deviation), the associated measurements at the point of deviation and for a total of 3 seconds after the deviation are discarded. The calculated 1σ overall uncertainties are 0.14 ppm for CO_2 and 2.2 ppb for CH_4, as calculated from the sum in quadrature of precision (1σ over 3 min; 0.03 ppm for CO_2 and 0.3 ppb for CH_4), repeatability (0.03 ppm for CO_2 and 0.4 ppb for CH_4), variance in flight due to cavity pressure fluctuations (0.08 ppm for CO_2 and 0.8 ppb for CH_4), uncertainty due to water corrections (0.1 ppm for CO_2; 2.0 ppb for CH_4), and pressure dependence of measurements based on environmental chamber studies (0.04 ppm for CO_2 and 0.1 ppb for CH_4).

The Meteorological Measurement System (MMS) is a NASA–Ames-developed airborne instrument that provides calibrated, science quality, *in situ* measurements of static pressure, static temperature, and wind in three dimensions. This instrument has been integrated into a number of NASA aircraft including the ER-2, the DC-8, the Global Hawk unmanned aircraft, and now the Alpha Jet, where the MMS system is mounted in the nose of the instrumented pod. The MMS instrument consists of three major systems: an air motion sensing system, an inertial navigation system, and a data acquisition system. The measured parameters are GPS positions, velocities, accelerations, pitch, roll, yaw, heading, angle of attack, angle of sideslip, dynamic total pressure, and total temperature. The primary products of MMS are pressure (precision of

±0.3 mb with accuracy of 0.5%), temperature (±0.3 K, 0.2%), horizontal wind (±1 m s^{-1}, 3.3%), and vertical wind (±0.3 m s^{-1}) (Gaines et al., 1992). The derived parameters are potential temperature, true airspeed, turbulence dissipation rate, and Reynolds number.

Finalized data sets containing O_3, CO_2, CH_4, and water vapor mixing ratios have been provided for each of the eight research flights listed in Table 1. O_3 data have been post-processed with the results of an eight-point calibration (ranging from 0 to 300 ppbv) performed before and after each flight using a NIST-traceable calibration source (2B Technologies, model 306) referenced to the WMO scale. Calibration settings for the O_3 monitor were left at manufacturer default settings during flight, and corrections to account for linearity offset and zero offset have been applied during data processing. Data have been averaged to 10 seconds and the inlet delay (lag time) has been subtracted; uncertainty estimates has been provided and are expected to be on the order of 3 ppbv. Quality assurance methods focus primarily on instrument pump performance; instrument temperature is a secondary diagnostic which has been evaluated. Data have been discarded if either characteristic is out of specification.

Similarly, CO_2 and CH_4 data have been post-processed to the WMO Mole Fraction Scale, version X2007 and NOAA2004 Scale, respectively, by comparison to two standards prepared by the NOAA ESRL's Global Monitoring Division (cyl #CC114990 with 416.267μmol CO_2/mol and 1985.69 nmol CH_4/mol; cyl #CA06327 with 393.319 μmol CO_2/mol and 1843.62 nmol CH_4/mol). Water vapor corrections determined by Chen et al. (2010) have been applied to calculate the dry mixing ratios of CO_2 and CH_4 during this study. The most important quality assurance step evaluates instrument optical cavity pressure and temperature for nominal conditions. The inlet lag time has been accounted for, and data has been averaged to 3 seconds. Uncertainty estimates have been provided.

In addition, formaldehyde (HCHO) was measured using the technique of non-resonant laser induced fluorescence (St. Clair et al., 2017). These data were not a primary focus of the CABOTS effort, but for completeness the HCHO data recorded have been included in the data set delivered.

Trace gas datasets including time and aircraft position have been delivered electronically to CARB as comma-separated-value text files. Meteorological data (temperature, pressure and 3-dimensional winds) and formaldehyde data are provided at 1 Hz in ICARTT-compliant text files.

4. NASA Modeling Efforts Conducted Under This Contract

Previous work from our research group has shown that integrated analyses using models, reanalysis, and diagnostic tools allow high O_3 values detected by *in situ* measurements to be attributed to multiple source processes (Ryoo et al., 2017). Backward air trajectories created using NOAA's Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model provided our first-level understanding of the origin of air masses sampled by AJAX during each flight. More thorough analyses began with back trajectories calculated using the National Aeronautics and Space Administration Goddard Space Flight Center (NASA GSFC) back trajectory model and the Weather Research and Forecasting Stochastic Time Inverted Lagrangian Transport (WRF-STILT) model. The NASA GSFC back trajectory model can be used for determining the origins (source regions) of air masses by tracing the position of mean wind trajectories. Mean wind trajectories may be less accurate in the planetary boundary layer (PBL) when the variability in observations is associated with a heterogeneous surface structure (i.e. mountains) and the surface condition is highly turbulent. The WRF-STILT model, one of the Lagrangian Particle Dispersion Models (LPDM), can overcome this limitation by simulating both advection and turbulence, and capturing the small scale heterogeneous structures in the PBL. The WRF model provides meteorology input data to drive the STILT model. To represent air that would have been sampled across the majority of the State on flight days, back trajectories were begun from starting locations evenly distributed over 36 – 42°N and 118 –125°W, from the surface to ~ 3km. As in previous studies (Ryoo et al., 2017), we used those trajectories to assess what fraction of the air masses were previously influenced by the mid troposphere, the lower troposphere, and the upper troposphere/ lower stratosphere. In addition, footprint and surface influence analyses enable us to estimate the potential future surface impacts of ozone sampled below 1 km.

Additional indicators that were explored include potential vorticity (PV) and O_3 from Modern Era Retrospective analysis for Research and Applications 2 (MERRA-2) and ERA-interim reanalysis data. ERA-interim is known as one of the latest and the best global atmospheric reanalyses produced by the European Centre for Medium-Range Weather Forecasts (ECMWF). MERRA-2 O_3 fields can be expected to provide a realistic estimate of mixing ratios within stratospheric intrusion layers, but will weakly represent observations in air masses dominated by surface emissions (Wargan et al., 2015; Ott et al., 2016). Thus, comparison of AJAX data to the ozone data product from MERRA-2 and ERA-interim gives additional indications of source altitudes for O_3 features observed in the AJAX data.

5. Analysis of Data and Model Results Performed Under This Contract

In this report, we first summarize the AJAX airborne measurements and compare to *in situ* surface monitors, NOAA ERSL TOPAZ and SJSU ozonesonde measurements. Data were accessed from: https://www.esrl.noaa.gov/csd/groups/csd3/measurements/cabots/. AJAX flights and comparisons were designed to expand the scope of the CABOTS data and provide additional data points between multiple stationary measurement locations both horizontally and vertically. Following comparisons to other data sets, the AJAX team performed model analyses using forward and backward trajectories. For each AJAX flight the following analyses were performed:

- Quantitative analysis with ozonesondes/lidar
- Quantitative analysis with surface data
- NASA GSFC back trajectories
- WRF-STILT footprint and surface influence

As an example of how AJAX data were put into context with other measurements, two different types of map plots are shown in Figure 2 to give an overview of O_3 in California during AJAX flight times. For 21 June 2016, Figure 2 highlights the differences between surface sites at the coast and those more polluted inland sites; an east-west O_3 gradient is evident. Such plots and time series plots of the underlying AJAX data are provided in Appendix A for each flight day.

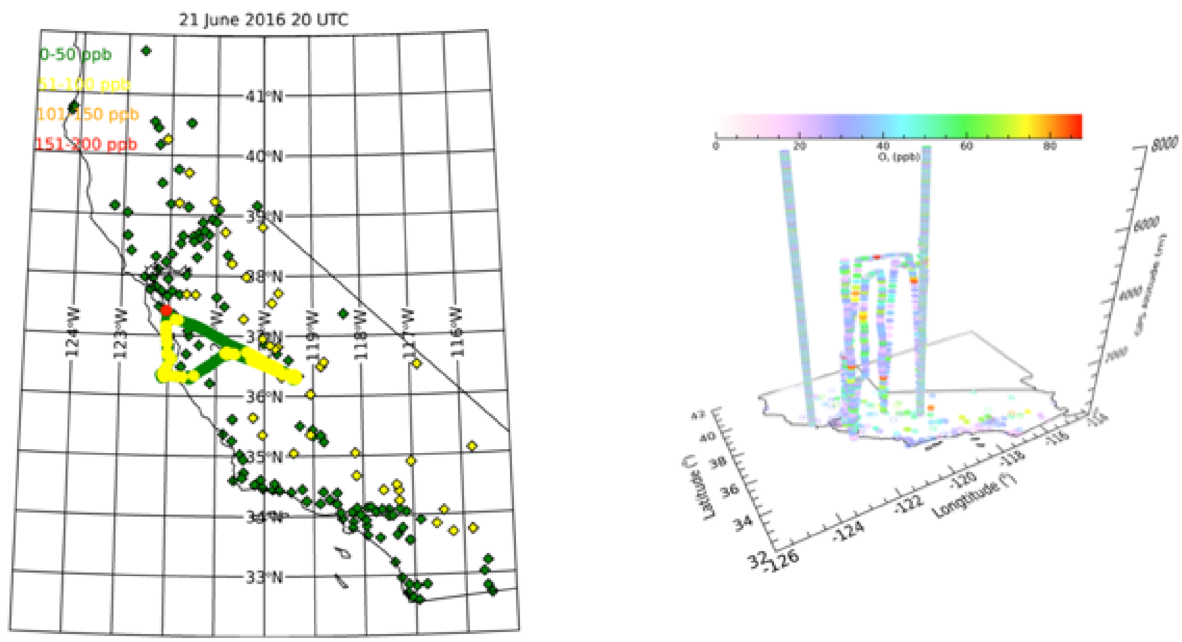

Figure 2. O_3 maps from surface sites, AJAX, ozone sonde and TOPAZ lidar for Flight 192 on 21 June 2016. Left: AJAX flight measurements and the corresponding 1-hr surface O_3 measurements at the median hour of the AJAX flight are shown. The color bins are given in the legend. Right: Using a common, continuous color scale, hourly surface O3 (hour closest to mid-way through AJAX flight), ozone sonde, lidar (closest in time to AJAX VIS profile) and AJAX data are plotted.

5.1. Quantitative Analysis with Ozonesondes/Lidar

Correlations were performed for every AJAX flight profile where ozonesonde and TOPAZ lidar profiles were also available. Data were co-plotted and averaged into 300 m altitude bins to perform a quantitative analysis. Because the airborne ozone data are averaged to 10 sec to reduce instrument noise, bins smaller than 300 m contained fewer than 5 data points, despite the similar rate of ascent/descent between the Alpha jet and the sondes. To ensure these layers were not too narrow, binning to 500 m was tested and no appreciable difference was seen. For TOPAZ lidar measurements, the profile used was the one closest in time to the AJAX Visalia profile (if available) and for ozonesondes, data were only correlated if the AJAX flight and ozonesonde were collected on the same date. Correlations between co-located profiles were examined to determine the degree of agreement between two datasets at the same location (see Figure 3), with significant correlation of observations made by two different techniques lending confidence to the accuracy of both. Correlations between measurements made at similar altitudes but different locations shed light on the spatial extent of uniform chemical composition and the relevance of measurements made at one location to understanding the ozone profile at locations elsewhere along the coast.

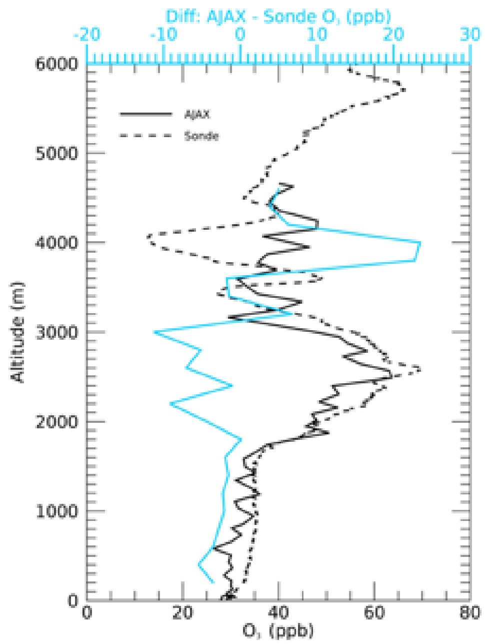

Figure 3. Bodega Bay O_3 profiles by ozone sonde (dash, black) and AJAX (solid, black) and the difference (AJAX – Sonde O_3) (blue) on 15 June 2016.

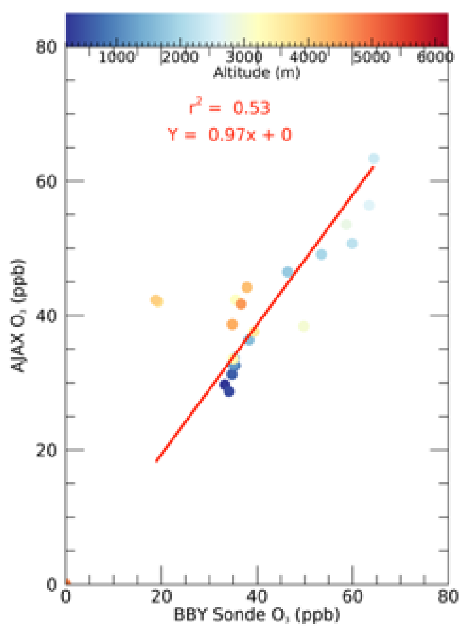

Figure 4. 300 m average AJAX O_3 (y) vs. ozone sonde O_3 (x) colored by altitude.

The two profiles in Figure 3 taken at a similar location and time show highly correlated results (slope = 0.97, r^2 = 0.53). Plots for additional days are documented in Appendix A, and Table 2 summarizes the results of all O_3 profile comparisons. Statistically significant correlations (r^2 > 0.5, bold face type in Table 2) are found for coincident profiles. Two instances with co-located and complete profiles (>6 – 0 km) of AJAX and ozonesonde data at THD (19 May) and BBY (15 June, Fig. 3) show a high degree of correlation (slopes = 1.01 and 0.97) with no significant bias. The good agreement between these two different techniques when sampling essentially the same air lends confidence to comparisons of the two data sets when collected farther away from each other.

The third coincident data set reveals how differences can be dependent on altitude. On 21 July, AJAX profiled over BBY only between 2.5 and 5.5 km (21 July). When compared with the BBY ozonesonde below 4.5 km, the average O_3 difference (AJAX-Sonde) is only -10 ppb, yet above 4.5 km the difference increases to -30 ppb. This divergence of ozone values with altitude is likely due to differences in sampling locations; AJAX profiles in a vertical column, whereas ozonesondes move with the wind and thus lateral displacement is greater at higher altitudes.

When comparing non-co-located profiles, correlations become less statistically-significant. On 28 July the AJAX profile at Pt Sur can be compared to a sonde launched by SJSU from Half Moon Bay (HMB), ~130 km to the north-northwest of the aircraft sampling location. This comparison shows a high degree of correlation (slope = 1.04, r^2 = 0.2), as anticipated because both sites are located south of the Golden Gate gap, although the correlation is not as strong as those for profiles which are essentially co-located.

At longer separation distances, sonde profiles at Bodega Bay (BBY) typically correlate well, but not statistically significantly, with profiles collected by AJAX ~240 km down the coast at Point Sur (Pt Sur).

Table 2. Dates, locations, and correlation results of offshore profiles by AJAX and ozone sondes during CABOTS. Boldface type indicates strong correlations. (BBY = Bodega Bay, HMB = Half Moon Bay, Pt Sur = Point Sur, THD = Trinidad Head)

Date	AJAX profile location	Ozonesonde location	Slope (ozonesonde (x) vs AJAX (y))	r^2
19 May 2016	**THD**	**THD**	**1.01**	**0.97**
3 June 2016	Pt Sur	BBY	0.87	0.01
15 June 2016	**BBY**	**BBY**	**0.97**	**0.53**
21 June 2016	Pt Sur	BBY	0.80	0.06
6 July 2016	Pt Sur	BBY	0.80	0.01
21 July 2016	**BBY (2.5-5.5 km)**	**BBY**	**0.76**	**0.52**
28 July 2016	Pt Sur	BBY	0.80	0.01
28 July 2016	Pt Sur	HMB	1.04	0.20

In comparisons between AJAX and TOPAZ profiles at Visalia, the mean slope of lidar (y) vs. AJAX (x) is 1.07 (data combined from 3 flights on 3 & 15 June and 21 July); differences lie within the combined uncertainties of the instruments and within 10 % accuracy standard set by ECCC ozonesondes. Further analysis is presented by Langford et al. (2019). Additional comparisons of AJAX vertical profile measurements with ozonesonde and TOPAZ results can be seen in Figure 10 of Faloona et al. (2020).

5.2. Quantitative Analysis with Surface Data

For each AJAX flight, surface sites were sorted based on distance from a selected (offshore when available) profile, as shown in Figure 5. Correlations were then performed between the mean AJAX O_3 measured within the "boundary layer" (<1 km), the "lower troposphere" (<2 km) and the "free troposphere" (3-6 km) and the mean O_3 measured at surface sites at "near" (<200 km), "mid" (200-500 km) and "far" (<500 km) distances from the selected airborne vertical profile. For each surface site, O_3 at the hour closest to the median time of the AJAX flight was correlated with the mean AJAX O_3 within altitude bins. The correlation was repeated replacing AJAX vertical profile O_3 with profiles from ozonesondes or lidar taken on the same days and times as the AJAX flights. No statistically significant correlations were observed, indicating that the variance in surface O_3 is not attributed to factors that influence variance of O_3 at altitude. An example of results is shown in Figure 6.

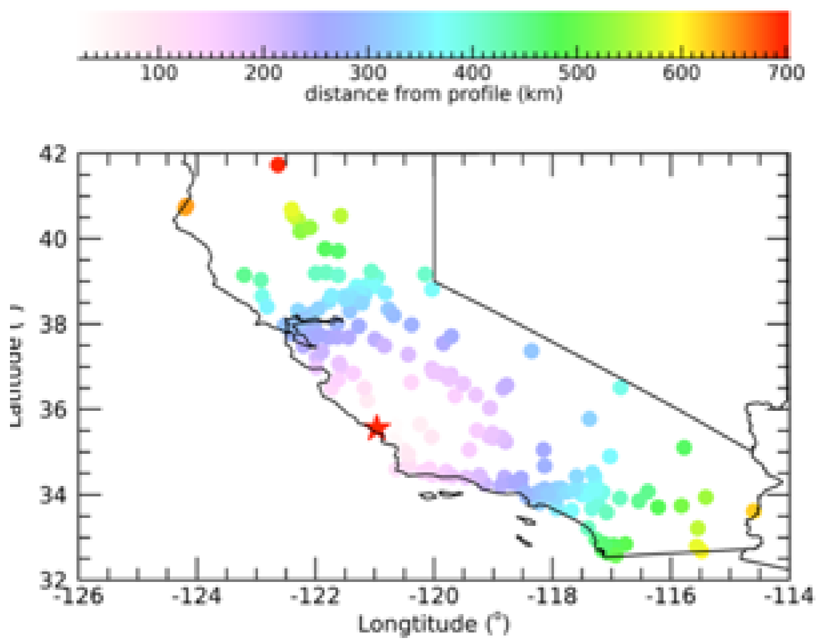

Figure 5. Map of surface sites colored by distance from AJAX offshore profile (star).

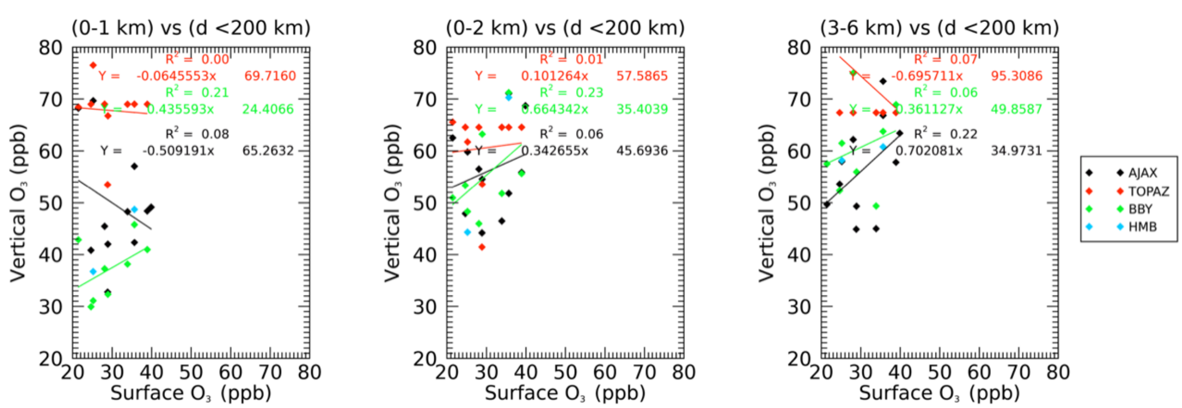

Figure 6. Correlating mean AJAX O_3 on each flight day within the boundary layer (left), lower troposphere (middle) and free troposphere (right) with mean surface hourly O_3 for surface sites near (<200 km) to AJAX.

5.3 Trajectory Model and Surface Influence Analyses

Using the trajectory modeling and other methods described above, three categories of influence were identified: stratospheric intrusion, long-range transport of ozone, and local influence. One example is given in detail here and the complete set of output results is included in Appendix B.

Long-range transport was the dominant influence on airborne data collected on 15 June 2016. Back trajectories (Figure 7) show there was a strong low-pressure system developing, and air parcels were transported from Northern Asia across the Pacific before reaching California. MERRA-2 reanalysis (Figure 8, pink ovals) further supports this identification of a strong cyclone developing over Asia 12 -7 days before June 15, 2016, leading to convective uplift of lower tropospheric pollution into the mid-troposphere, where it was subsequently transported eastward. The MERRA-2 fields plotted in Figure 9 show high values of potential vorticity in the upper troposphere coupled with low amounts of water vapor, indicating that the dynamic mechanism responsible for bringing this polluted air down to lower altitudes included an element of stratosphere-troposphere exchange (STE).

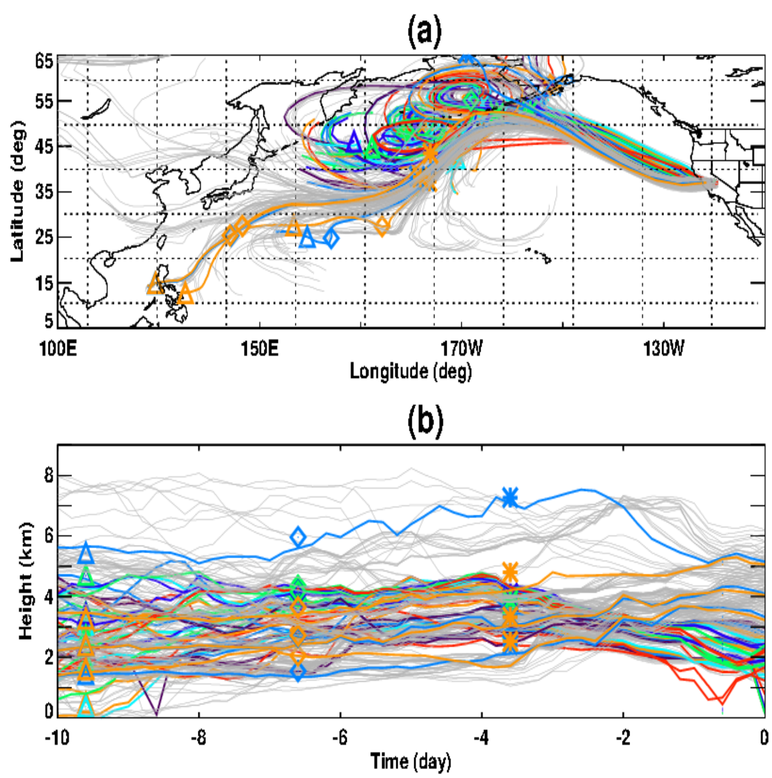

Figure 7. (a) Spatial and (b) temporal evolution of 12-day NASA GSFC back trajectories starting from California on 15 June 18Z, 2016. To improve readbiity, only trajectories starting within 36°N-38°N and 125°W-118°W are plotted here.

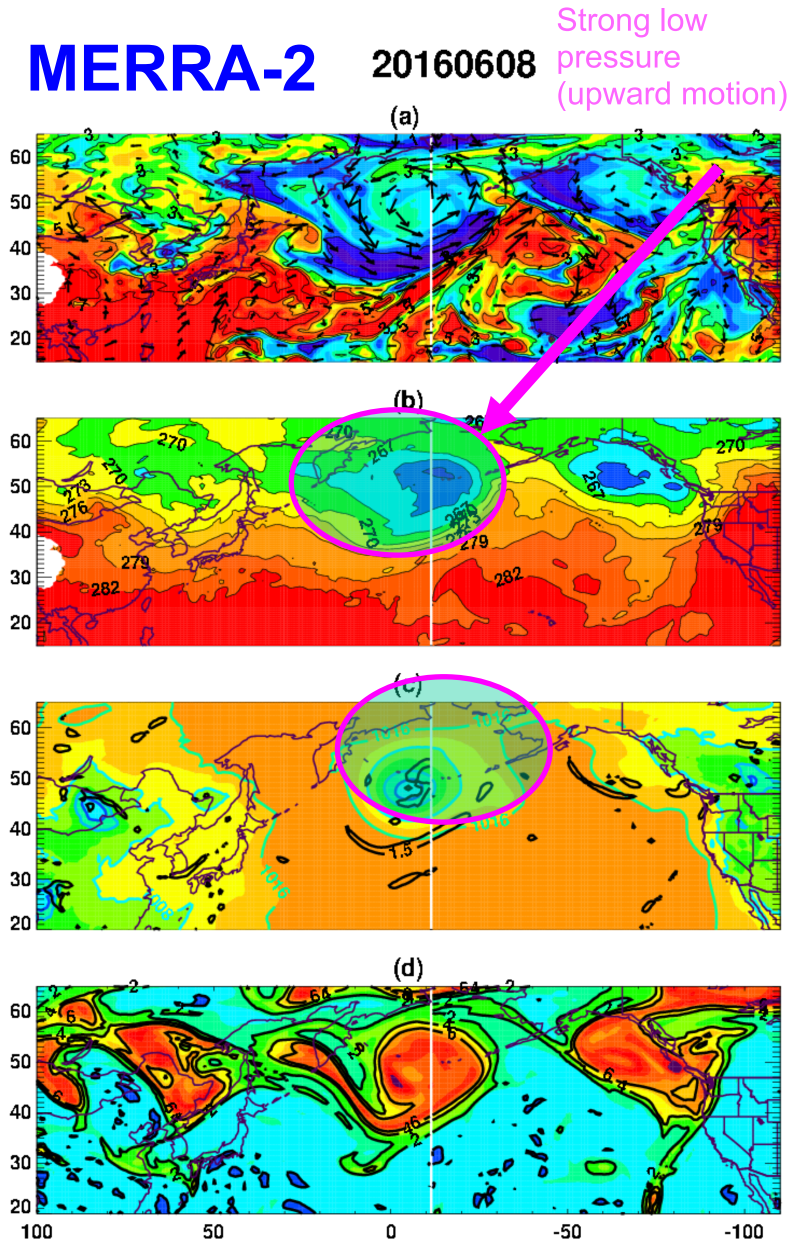

Figure 8. MERRA-2 fields on 8 June 2016, one week before the AJAX flight on 15 June, showing upward motion due to a strong low pressure system. (a) q (water vapor) overlaid with wind at 700 hPa, (b) temperature at 700 hPa, (c) sea level pressure, and (d) Potential Vorticity (PV) at 250 hPa.

This air (polluted, but dry from the inclusion of stratospheric air) was sampled by AJAX at altitudes near 5 km over Visalia (Figure 10, green profiles). The enhanced amounts of CO_2 and CH_4 identify this parcel as polluted, and thus not solely resuting from the intrusion of ozone-rich but otherwise clean stratospheric air. The horizontal extent of this region of dry, ozone-rich air can be seen in Figure 11, where the purple rectangle indicates an area with O_3 > 80 ppb and H_2O < 0.2%v, ~70-90 km wide at an altitude of 5-5.25 km.

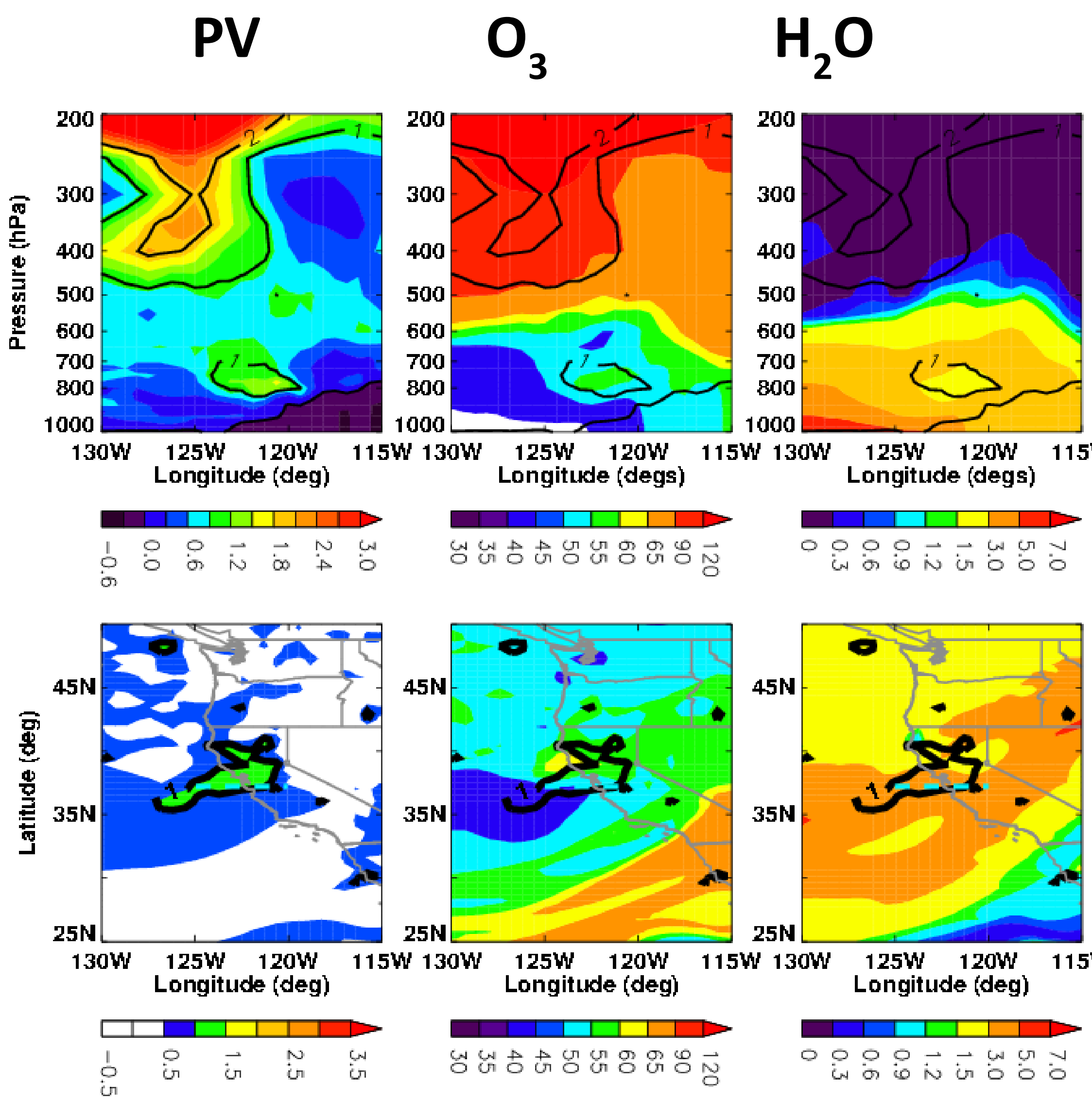

Figure 9. For 15 June 2016, high MERRA-2 O_3 from 200 to 400 hPa (middle column, in ppbv) is closely linked to the high PV in the upper troposphere and lower stratosphere (left, in PVU), containing CO transported from Asia (not shown, but observed in FLEXPART model results provided by NOAA). Very low H_2O supports this as well (right, in g/kg). The top row shows values averaged over 35-39° N, while the bottom row provides values on the 700 hPa surface.

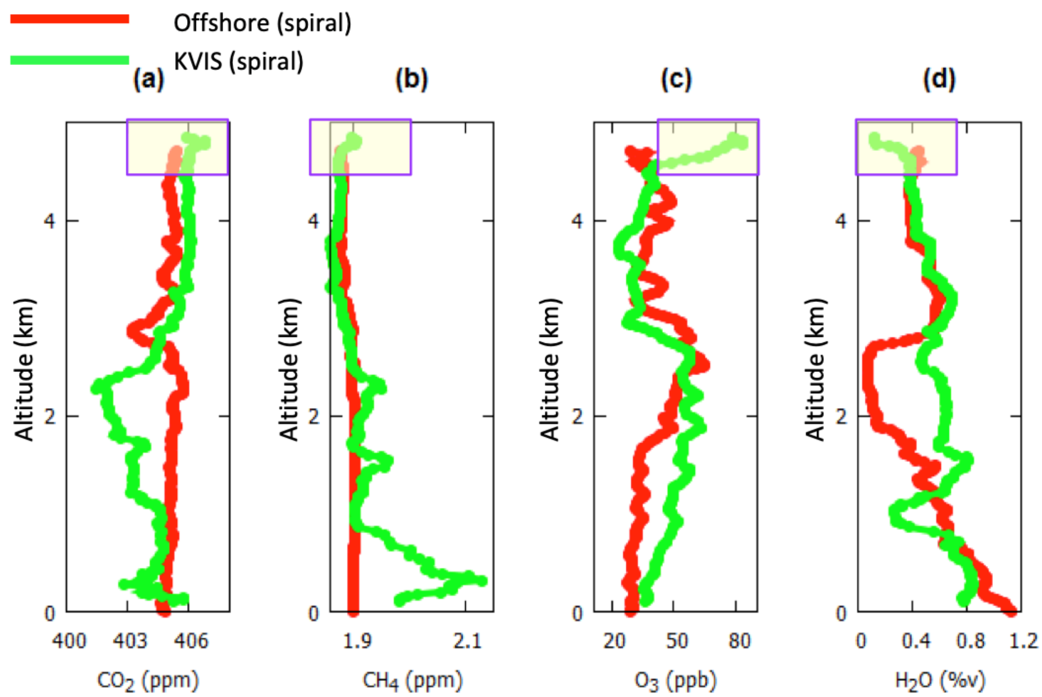

Figure 10. Vertical profiles of (a) carbon dioxide, (b) methane, (c) ozone, and (d) water vapor collected in spiral profiles on 15 June 2016 over Visalia (KVIS, green) and offshore (Bodega Bay, red). Note the layer of increased CO_2, ozone and methane (and reduced water) at the very top. This feature can also be seen in Figure 11.

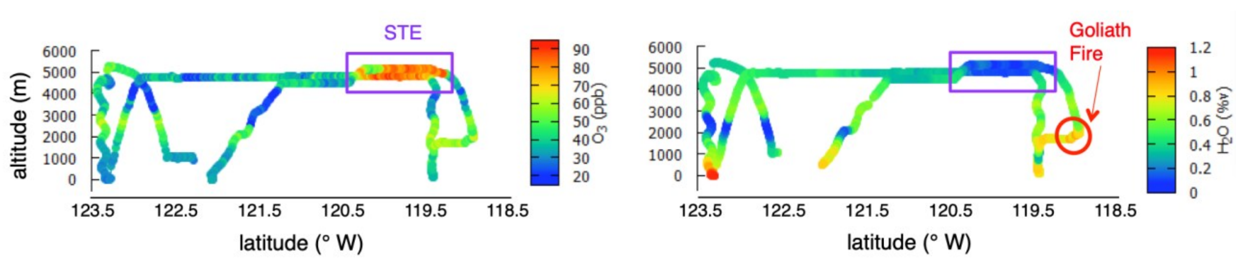

Figure 11. Flight tracks of (left) ozone and (right) water vapor for the 15 June flight containing the vertical profiles shown in Figure 10. The offshore spiral is on the left edge of these plots. The Goliath fire was sampled after the ascent out of Visalia, and the purple box indicates where a stratosphere-troposphere exchange (STE) event carrying Asian pollution was intersected on both the southbound and northbound legs.

The influence of this polluted air on surface sites downwind was examined with WRF-STILT forward trajectories and footprint analysis. Figure 12 highlights the regions where air that was sampled at 1000 m impacted the surface within 2 days subsequent to sampling.

This footprint technique was also applied to the data collected on 28 July 2016, several days after the ignition of the Soberanes wildfire. The reader is encouraged to consult the extended analysis of forward trajectory and footprint plots given in Appendix B for that day.

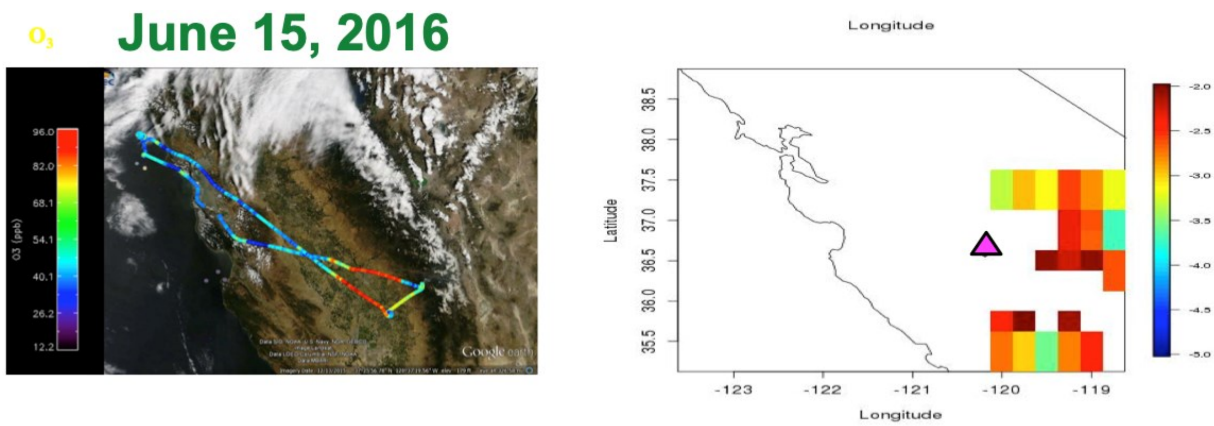

Figure 12. Left: Flight track for AJAX Flight #191 on 15 June 2016, colored by ozone amount; compare to left panel in Fig. 11. Right: forward (+2 days) impact on surface ozone from air parcels released at 1000 m at the location (pink triangle) and time of the AJAX observation. The scale is the logarithm (base 10) of the footprint. The red colors indicate the large surface impact on concentration change at the receptor for a unit surface flux persisting over a given time interval [mol-1 m2 s].

Using these tools to explore all the *in-situ* data collected by AJAX during the CABOTS campaign, the analysis of Ryoo et al. (2017) was used to characterize the source regions for the ozone sampled. One example is explained in detail here, and all analyses are collected in Appendix B.

The position (latitude, longitude, altitude) of calculated back trajectories ~10 days before an AJAX flight was used to identify dominant influence mechanisms: local (if air masses were east of 130°W (equivalent to 230°E) and altitude was 6 km or lower), long range transport (west of 160°E and 6 km or lower), or stratospheric influence (all longitudes if altitude was > 6 km). Trajectories which ended below 6 km over the Pacific Ocean (160°E – 230°E) were categorized as "unassigned" for the purpose of this analysis.

The results of this categorization for the 12 May flight are shown in Figure 13. The solid bars in panel (a) indicate the percentage of trajectories in each of the three source categories; in this instance, the largest number of the back trajectories run were located at an altitude > 6 km (blue solid bar in Fig. 13a) ten days before sampling by AJAX. From MERRA-2 reanalysis data, the ozone values can be sampled at the trajectory origins, and those values are displayed as probability distribution functions in panel (b), using the same color-coding. Lastly, the hatched bars in panel (a) show the relative proportion of ozone delivered to California from the three origins. Because the UT/LS air parcels contained much higher values of ozone than the lower altitudes, the percentage of ozone delivered (hatched blue) significantly exceeds the simple percentage of trajectories (solid blue).

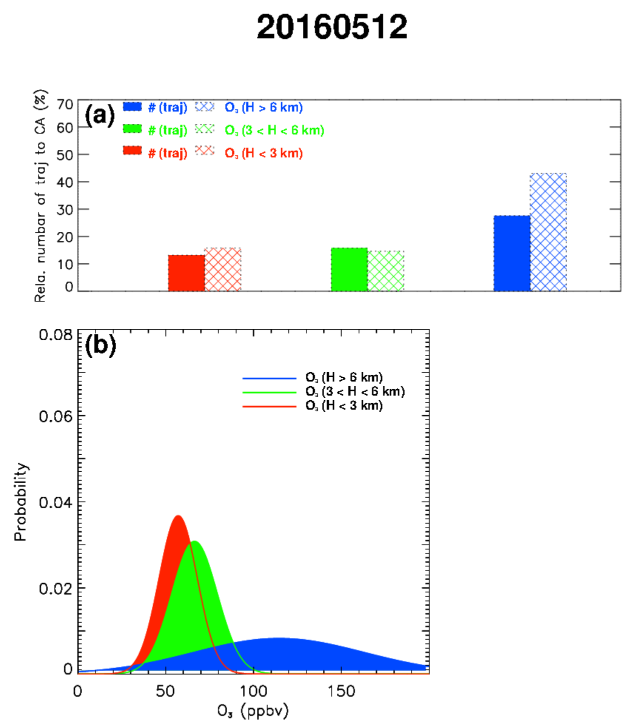

Figure 13. (a) The percentage of back trajectories (solid bar) and percentage of O3 (hatched bar) from the different source regions: altitudes below 6 km for positions east of 130 W in red (local), long range transport from Asia and the western Pacific in green, and upper troposphere/lower stratosphere influence in blue (altitude above 6 km at any location). (b) Probability density function (PDF) of the O3 mixing ratio of different sources at the end of 10-day back trajectories coming to California on 12 May 2016 (36 – 42°N and 118 –125°W, from the surface to ~ 3 km).

Histograms were constructed for all eight AJAX/CABOTS flight days and are collected in Appendix B. In general terms, the air masses sampled in May-June had the largest contributions from the UT/LS compared to flights later in the summer, with a gradual decline seen with time. 15 June and 6 July showed considerable influence from long-range transport (green). Flight days later in the summer had noticeably more input from local sources (red). Oceanic (or "unassigned") trajectories were a significant fraction of the total on 3 June and 21 July.

A summary of results is collected in Table 3, which also indicates the meteorological and trace-gas features used to make each assessment.

Table 3. Summary of ozone source categorization based on trajectory modeling results and in situ measurements. SI = Stratospheric Intrusion, LT = Long range Transport, Local = local influences.

AJAX	Date	Synoptic & Met Features	Major features observed from AJAX	SI	LT	Local
F188	**May 12**	South or Southwesterly wind was dominant. Dry and warm condition.	High ozone is spread down to the lower level down to 3km with very dry air, indicating the transport of stratospheric air. Mostly local with some long-range transport; intermediate high O_3 was observed offshore.	✓	✓	✓
F189	**May 19**	Dry condition with low RH in low altitude (< 700 hPa). Low wind speed with clear skies with high cirrus clouds. Bay area is located under a upper-level trough, which resides over Northern CA.	High O_3 is observed over SELVY around 6, 4, 2 km.	✓	✓	
F190	**June 3**	Low surface pressure system resides on the west of the coastal regions. Mild wind and dry condition. Upper-level trough penetrated down into the mid troposphere.	Downward motions ~2 - 4.5km were found over Chews Ridge => entrainment from FT to BL can be expected. FLEXPART model shows strat intrusion too. O_3-rich Stratospheric dry air intruding to the lower troposphere (~ 3 km) was detected.	✓	✓	
F191	**June 15**	Low surface pressure system resides on the west of the coastal regions of the western U.S. Mild wind and dry condition is observed in CA. Strong jet is shown over the Pacific over the past few days. Dominant wind is southwesterly. Strong wind both offshore and near Visalia (KVIS) around ~5 km.	High O_3 (> 80 ppbv) and very dry air (H_2O < 0.2 %v) are observed at about 5 km: (Note FLEXPART model shows high CO transport from Asia). O3-rich (high PV) Stratospheric dry air intruding to the lower troposphere (~ 6 km) was detected.	✓	✓	
F192	**June 21**	Northwesterly winds with slower wind speeds over the coastal areas of CA. Cold front was found over CA. Clear skies at the 500 mb and 700 mb levels. CA is located in the ridge surrounded by two upper-level troughs. Dominant wind is westerly and south westerly.	Elevated CH_4/CO_2 is observed offshore (Pt. Sur). High O_3 was also observed with low H2O at bodega bay.	✓	✓	✓
F194	**July 6**	Clear skies at the 300 mb, 500 mb and 700 mb levels. No front was developed. However, the trajectories and horizontal wind at June 29, 2016 (not shown) indicates that the strong jet was developed over the Pacific ~10 days ago.	Airmass offshore (Pt. Sur) appears to be transported to Visalia (KVIS).. (shows similar features around 4 – 5 km) – high O_3 but not so low H2O: implying long-range transport and local pollution.		✓	
F195	**July 21**	Jet stream located over the Northern Bay Area. A low pressure system was moving Southeasterly throughout a few days. Clear skies across CA.	Elevated O_3 (dry (low) H_2o) at 2-3 km near Bodega Bay, and elevated CH_4 are observed at Visalia.			✓
F196	**July 28**	Very weak wind, no frontal system developed, warm and dry conditions. No strong jet over Pacific. Localized and stagnant, but turbulent air condition (see AJAX MMS wind data) with different air mass with different wind.	Elevated CO_2 is observed when H_2O is high, indicating locally generated pollution. Elevated CH_4, O_3, and H_2O are observed at about 3-4 km, and 1.5-2.5km over BLANC.			✓

6. Summary and Conclusions

The objective of this project was to calibrate, finalize, and analyze the atmospheric trace gas and related data collected by the NASA AJAX group during the CABOTS of 2016. Task 1 (Finalize data) has been completed and is described in Section 3 of this report. Task 2 (Compare to other CABOTS data sets) has been completed, is described in Sections 5.1 and 5.2, and provides the bulk of the material documented in Appendix A. Correlations among different profile locations (Task 3) are presented here (Section 5.1), and additional comparisons are included in Appendix A. This task also contributed to analyses and figures published by other CABOTS co-investigators. Please see Figures 13, 14, 15, and 16 of Langford et al. (2019) and Figure 10 of Faloona et al. (2020).

Trajectory analyses (Task 4) are summarized in Table 3 above and have been used to assess the source locations for ozone observed over California. The largest contributions to ozone from the UT/LS were identified in May and June, with significantly less influence on flight days in July. Two days in the middle of the campaign were dominated by long range transport, and the last two flights in July were dominated by "local" influences, having their origins ten days prior over the coastal Pacific Ocean and/or California. Local contributions from the Soberanes wildfire were noted on 28 July.

Preliminary results have been presented (Task 5) to the CABOTS Science Team in several virtual meetings (Aug 2018, Sept 2018, Dec 2018, and Feb 2019), with particular focus on the comparison to AJAX data to other data sets (TOPAZ and ozonesondes). Task 6 (NASA ITAR review) is in progress. The Draft Final Report (Task 7) was delivered on 8 January 2020 , with the *objectives* documented in Section 2, *discussion of the work performed* provided in Sections 3 and 4, and inclusive of all *conclusions* reached (Section 5). Electronic datasets have been provided in satisfaction of Task 9. In addition to the agreed-upon data sets (ozone, greenhouse gases, and meteorological parameters), data from a NASA formaldehyde sensor which was co-manifested on several occasions are also included. We hope these data can be of use to others who may further investigate the CABOTS observations. Feedback from CARB staff and Research Screening Committee members informed the execution of Task 8 (Final Report) and Task 10 (Prepare manuscript). Task 11 (Present seminar) will be arranged in consultation with CARB staff.

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