An extensive database of airborne trace gas and meteorological observations from the Alpha Jet Atmospheric eXperiment (AJAX)

Emma L. Yates[1,2], Laura T. Iraci[1], Susan S. Kulawik[1,2], Ju-Mee Ryoo[1,3], Josette E. Marrero[1,†], Caroline L. Parworth[1,‡], Jason M. St. Clair[4,5], Thomas F. Hanisco[4], Thao Paul V. Bui[1], Cecilia S. Chang[1,2], Jonathan M. Dean-Day[1,2]

[1]NASA Ames Research Center, Moffett Field, CA 94035, USA

[2]Bay Area Environmental Research Institute, Moffett Field, CA 94035, USA

[3]Science and Technology Corporation, Moffett Field, CA 94035, USA

[4]Atmospheric Chemistry and Dynamics Laboratory, NASA Goddard Space Flight Center, Greenbelt, MD 20771, USA

[5]Joint Center for Earth Systems Technology, University of Maryland Baltimore County, Baltimore, MD 21228, USA

†Now at Sonoma Technology, Inc., Signal Hill, CA 90755, USA

‡Now at Aclima, Inc., San Francisco, CA 94111, USA

Correspondence to: Emma L. Yates (emma.l.yates@nasa.gov) and Laura T. Iraci (laura.t.iraci@nasa.gov)

Abstract. The Alpha Jet Atmospheric eXperiment (AJAX) flew scientific flights between 2011 and 2018 providing measurements of trace gas species and meteorological parameters over California and Nevada, USA. This paper describes the observations made by the AJAX program over 229 flights and approximately 450 hours of flying. AJAX was a multi-year, multi-objective, multi-instrument program with a variety of sampling strategies resulting in an extensive dataset of interest to a wide variety of users. Some of the more common flight objectives include satellite calibration/validation (GOSAT, OCO-2, TROPOMI) at Railroad Valley and other locations, and long-term observations of free-troposphere and boundary layer ozone allowing for studies of stratosphere-to-troposphere transport and long-range transport to the western United States. AJAX also performed topical studies such as sampling wildfire emissions, urban outflow, and atmospheric rivers. Airborne measurements of carbon dioxide, methane, ozone, formaldehyde, water vapor, temperature, pressure, and 3-D winds made by the AJAX program have been published at NASA's Airborne Science Data Center.

1 Introduction

The Alpha Jet Atmospheric eXperiment (AJAX) was a flight measurement program designed to provide year-round observations to support satellite data validation and document the atmospheric composition of a coastal region in close proximity to urban and agricultural areas. The ongoing nature of the program was ideal for studies of changes in both the

atmosphere and the remote sensing instruments in service during the period 2011 to 2018. The AJAX program was the result of a public-private partnership between NASA Ames Research Center (NASA ARC) and H211 LLC, which owns the Alpha Jet, a tactical military aircraft modified for scientific observations (see Figure 1, left). This partnership provided the opportunity to acquire measurements of trace gas species and meteorological parameters over California and Nevada, USA, producing a unique multi-year, multi-objective, multi-instrument airborne dataset, with 229 flights and ~450 flight hours. The distribution of AJAX flights by common flight sampling locations and by year and season are shown in Figure 1 (middle and right).

The AJAX program flew on average 2-3 science flights a month, as determined by aircraft availability and weather conditions. Flight opportunities were typically arranged the week prior to a science flight, in which consideration of synoptic conditions, time of year and science objectives determined desired flight objectives and target locations. This flexibility and rapid turnaround meant that the AJAX program could respond to short-lived regional scientific opportunities, such as sampling emissions from wildfires or atmospheric rivers. This flexibility also allowed for the participation in, and collaboration with, more traditional field campaigns (e.g., CABOTS (Faloona et al., 2020), LVOS (Langford et al., 2015), DISCOVER-AQ-CA (Johnson et al., 2014), SEAC⁴RS (Toon et al., 2016)). Table 1 lists some of the more common AJAX flight objectives, definitions, and number of flights associated with each objective; note that many AJAX flights accomplished multiple objectives in a single flight.

Given the wide variety of flight objectives shown in Table 1, AJAX data have been used to investigate a variety of science questions including:

i. Satellite validation: AJAX regularly sampled vertical profiles of the atmosphere from ~8 km to the lowest safe altitude within the boundary layer (1000 ft (~300 m) in congested (urban) areas and 500 ft (~150 m) over sparsely populated regions (FAR, 2023)). For example, the regularly-used satellite validation site, Railroad Valley (RRV), NV is a common AJAX flight target location. The AJAX program has a long record of vertical profile observations of CO_2 and CH_4 in conjunction with satellite overpasses and has been used as comparison data for GOSAT (Tanaka et al., 2016), GOSAT-2 and OCO-2 (Kulawik et al., 2017). In addition, AJAX flights also performed vertical profiles of ozone and formaldehyde under TROPOMI, and these data are still to be fully analyzed (Parworth et al., 2018).

ii. Free tropospheric ozone: Free tropospheric ozone trends over the western US has been an active area of research for many years. Data provided by flights from the AJAX program have proved to be a useful asset in assessing ozone trends and identifying extreme events, such as stratosphere-to-troposphere transport and long-range transport of pollution (e.g., Faloona et al., 2020; Fine et al., 2015; Langford et al., 2018; Ryoo et al., 2017; Yates et al., 2013). The long-term, sustained observational approach adopted by the AJAX program is of value in assessing changes in free-troposphere ozone trends (Chang et al., 2022; Lin et al., 2015).

iii. Fires: AJAX has sampled a total of 14 wildfires over 17 different flights, with some fires sampled more than once (Iraci et al., 2021b). Iraci et al. (2022) provides an overview of wildfire emissions sampled by AJAX. In addition,

there have been focused analyses of emissions from California's Rim wildfire during 2013 and Soberanes fire during 2016 as reported by Baker et al. (2018), Yates et al. (2016) and Langford et al. (2020).

iv. Urban and point source emissions: The Alpha Jet's fast cruising speeds can be useful in reducing uncertainties due to the evolution of the boundary layer during flight sampling. AJAX can essentially take a "snapshot" of the boundary layer and has been used to assess outflow of CO_2 and CH_4 emissions from urban centers (Ryoo et al., 2019), and emission point sources (Leifer et al., 2018; Tadić et al., 2017), as well as the transport of emissions (Leifer et al., 2020).

v. Atmospheric rivers: The flexibility of the AJAX program was an advantage to provide a rapid response to measure the early phase of atmospheric river events as demonstrated by Ryoo et al. (2020). AJAX examined terrain-trapped airflows during an atmospheric river event which impacted northern California in March 2016, providing spatially resolved 3-D wind observations along a portion of the coastline where wind profiler data did not exist.

vi. Central Valley: AJAX collaborated with NASA's DISCOVER-AQ-CA field campaign in 2013 to sample the boundary layer in California's San Joaquin Valley, a region with some of the most severe ozone and PM2.5 pollution in the U.S. AJAX ozone, CO_2 and CH_4 data have been used for observational and model studies of air quality and greenhouse gas emissions in this region (e.g. Faloona et al., 2020; Cui et al., 2019; Johnson et al. 2014; Johnson et al. 2016; Yates et al., 2015) as well as the impacts on surrounding, rural areas (Yates et al., 2020).

This paper provides a detailed overview of the AJAX program and its data products. We present a summary of the range of flight objectives, sampling strategies, airborne instrumentation, and data processing to produce AJAX trace gas and meteorological data products. An overview of the AJAX dataset is presented, including discussions on long-term trends and greenhouse gas correlations and a case study highlighting the use of AJAX data in support of satellite validation of lowermost tropospheric products from OCO-2 and GOSAT.

2 Description of airborne platform, sensors, and data processing

The Alpha Jet has four external wing-pods; the two outboard pods on each wing contain fuel, and the two-inboard pods (painted white in Figure 1) are redesigned fuel tanks which can house scientific instruments, as shown in Figure 5. The instrument wing-pods can each carry a payload of 245 kg within 0.15 m^3 of available space, and modifications in the rear cockpit allow for electrical (on/off) control of the instrument payload. A NASA Airworthiness Statement was issued following thorough Airworthiness and Flight Safety Reviews of the scientific instrumentation and planned operations. During all science flights the Alpha Jet operated as a public-use aircraft under a flight release issued by NASA Ames Research Center.

The AJAX payload was developed incrementally. AJAX flew its first science flight in January 2011 with an ozone (O_3) monitor. In June 2011 a greenhouse gas monitor was added, measuring carbon dioxide (CO_2), methane (CH_4) and water vapor (H_2O). In June 2013, the Meteorological Measurement System (MMS) was added to measure 3-D winds, temperature, and

pressure, and lastly the addition of the Compact Formaldehyde Fluorescence Experiment (COFFEE) was completed in December 2015.

The Alpha Jet flies at speeds between 75-230 m/s with a ceiling height of 12 km, however science flights were typically flown
95   at speeds of ~140 m/s (~500 km/h) up to an altitude of 8 km (limited by decreasing performance of the greenhouse gas instrument at higher altitudes).

2.1 Ozone monitor

AJAX observes O_3 mixing ratios using a commercial, UV absorption based, O_3 monitor (2B Technologies Inc., model 205, S/N 734), modified for flight worthiness and improved performance. Details of the instrument have been reported by Yates et
100   al. (2013). Modifications of the O_3 monitor include an upgrade of the pressure sensor and pump to allow measurements at high altitudes (low pressures), the inclusion of 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 (perfluroalkoxy-polymer, PFA) with a backward-facing inlet positioned on the underside of the instrument wing-pod. Air is delivered through a 5 µm PTFE (polytetrafluoroethylene) membrane filter to remove fine particles prior to analysis.

105   The O_3 monitor has undergone thorough instrument testing in the laboratory and in a pressure- and temperature- controlled environment to determine the precision, linearity, and overall uncertainty. Eight-point calibration tests (ranging from $0 - 300$ ppbv) were performed before and/or after most AJAX flights using an O_3 calibration source (2B Technologies, model 306, S/N 045, referenced to the WMO scale). The uncertainty of AJAX O_3 measurements is estimated from the sum in quadrature of the laboratory precision (1-σ over 3 min at 10 s sampling resolution), variability in laboratory calibrations (the differences
110   observed in the calculated zero offset when sampling the ozone source set to 0 ppb), repeatability (maximum difference in ozone observed from three calibrations on the same day) and pressure dependance (the change in ozone observed with pressure changes while sampling 50 ppb ozone in environmental chamber tests). The overall uncertainty of the airborne AJAX O_3 data are estimated to be ~3.0 ppbv, based on laboratory and chamber testing; a breakdown of the uncertainty terms is shown in Table 2.

115   AJAX performed regular flights to Trinidad Head (THD) in conjunction with ozonesonde launches for comparison and validation of AJAX O_3 data and to the Table Mountain Facility (TMF) for comparison with the TMF tropospheric ozone lidar (TMTOL) and TMF ozonesonde. Figure 6 shows an example of coincident ozone profiles taken over TMF on 24 May 2013 by AJAX, TMTOL and TMF ozonesonde. Comparisons show very good agreement between the airborne, TMTOL and TMF ozonesonde measurements, well within measurement uncertainties, despite spatial and temporal differences in data sampling.

120   To provide quality controlled O_3 data, we applied standard processing to the raw data as follows:

— Removal of datapoints outside of a predefined operational limit. If the reported flow rate through the sample cell fell below 1 L/min, those datapoints were removed from the dataset.

— Removal of outliers in the data due to instrument instability. If ΔO_3 (O_3 (i)- O_3 (i+1)) is greater than the 1σ standard deviation of the entire O_3 data, those datapoints (O_3(i) and O_3(i+1)) were removed from the dataset.

125   — Calibration based on the linearity and zero-offset factors calculated from the closest calibration to the flight (typically ±1 day).

    — Averaging of O_3 data from 2 s resolution to report at 10 s resolution, improving precision and the overall quality of the finalized dataset.

2.2 Greenhouse gas sensor

130 AJAX provides in situ measurements of CO_2 and CH_4 using a commercial, Cavity Ring Down Spectroscopy (CRDS), sensor (Picarro Inc. G2301-m, S/N 634-CFDDS2120) which has been modified for flight. Details of the sensor have been reported elsewhere (Tadic et al., 2014, Tanaka et al., 2016). Briefly, the AJAX GHG sensor has been re-packaged into two separate enclosures (analyzer and electronics) to fit within the wing-pod. Additional modifications to the original sensor for flight readiness included fans for thermal management, an additional filter on the inlet, and vibration isolators. The GHG sensor is

135 powered on ~1 hr prior to flight take-off in a pre-flight routine that ensures the laser temperature reaches its operational temperature of 45 °C and commences measurements early into each science flight.

The GHG sensor has undergone thorough laboratory and environmental chamber testing to evaluate its precision, linearity, and overall accuracy under a variety of operating conditions. Calibrations are performed before and/or after most AJAX flights using NOAA ERSL whole air standards certified by the WMO Central Calibration Laboratory for CO_2 and CH_4. The overall

140 uncertainty of aircraft measurements was estimated considering the precision (1σ), repeatability of calibrations, in-flight variance due to cavity pressure fluctuations, uncertainty in water vapor corrections, and pressure dependence of measurements based on environmental chamber studies.

AJAX CO_2 and CH_4 measurements were subjected to the following quality control procedures to generate the finalized GHG dataset:

145   — Removal of instrument lag time (5 s), calculated given the length of the inlet tubing and measured flow rate.

    — Removal of outliers in the data due to in-flight instability of the optical cavity pressure. Datapoints where the cavity pressure deviated by more than 0.2% were removed from the dataset.

    — Removal of datapoints outside of operational temperature limits. Datapoints were removed when the cavity temperature was >45.15 °C or <44.95 °C.

150   — Water vapor corrections. AJAX applies the water vapor corrections described by Chen et al. (2010) using simultaneous water observations.

    — Application of calibration based on the closest calibration to the flight (typically ±1 day).

    — Averaging of GHG data from 3 Hz resolution to report at 3 s resolution, improving precision and the overall quality of the reported, finalized dataset.

155 Typical overall uncertainties for AJAX CO_2 and CH_4 are 0.16 ppmv and 2.2 ppbv respectively as shown in Table 3 and reported by Tanaka et al. (2016).

2.3 Formaldehyde instrument: COmpact Formaldehyde FluorescencE Experiment

AJAX collected in-situ observations of formaldehyde (HCHO) using a custom-built instrument called COmpact Formaldehyde FluorescencE Experiment (COFFEE), which was designed specifically to join the AJAX payload. A detailed overview of COFFEE is presented by St. Clair et al. (2017), with updates to the optical design in St. Clair et al. (2019). Briefly, COFFEE utilizes non-resonant laser-induced fluorescence (NR-LIF) to measure formaldehyde, with 300 mW of 40 kHz 355 nm laser output exciting multiple formaldehyde absorption features and the resulting fluorescence measured by two photomultiplier tube detection axes. Fluorescence signal is collected for 500 ns, with 5 ns resolution, at each laser pulse and then averaged to 1 s. The 1 Hz fluorescence signal shape is processed post-flight to calculate formaldehyde mixing ratios.

Data collected prior to April 2018 (prior to the optical changes described in St. Clair et al., 2019) have a water dependence term in the measurement uncertainty that is not present in data collected afterward. Overall measurement uncertainty is ± (20% of HCHO + 200 pptv + 230*[H_2O % by volume]) before April 2018 and ± (20% of HCHO + 200 pptv) in April 2018 and after. All final HCHO data passes multiple quality control steps, including:

— All instrument engineering parameters (temperatures, pressures etc.) are nominal.

— Laser is fully warmed up, as indicated by the power monitor photodiode.

— Final laboratory calibrations are applied.

2.4 Meteorological Measurement System (MMS)

AJAX reports calibrated, science-quality static pressure, static temperature, and 3-dimensional winds using the NASA-Ames developed Meteorological Measurement System (MMS). MMS consists of three major systems: An air motion sensing system, an inertial navigation system and a data acquisition system. MMS has been deployed on multiple NASA aircraft and has been involved in numerous NASA field campaigns. A detailed overview of MMS instrumentation and basic concepts has been described by Scott et al. (1990).

The accuracy of MMS pressure, temperature and wind data was achieved through a combination of laboratory sensor calibrations and analysis of dedicated in-flight maneuvers. Lab calibrations include determination of inertial navigation system time delays by means of executing a series of fixed axis rotations on a dedicated pendulum test platform. The custom-designed apparatus allows recording angular position in addition to attitude, position, velocity and acceleration data from the INU. This information was combined with In-flight maneuvers that included a series of pitches, yaws, and a box pattern featuring a roll/unroll sequence after every 90 degrees of heading angle rotation. Adjustments were made to attack and yaw angle measurements to account for rotation of the radome orifices with respect to the aircraft reference frame, which would shift slightly as the wing pod (where MMS instrumentation was integrated) was removed and reinstalled for maintenance purposes. Local calibration constants were determined by eliminating aircraft motion artifacts from pressure, temperature, and wind measurements. However, since the Alpha-Jet operated within a broad regime of aerodynamic conditions (e.g., Mach number and angle of attack), further statistical analysis was applied to individual calibration results to generate a map of sensor

corrections applicable to the entire flight envelope. Calibrations were updated on a periodic basis as time allowed for dedicated MMS maneuver flights, which were intentionally flown in relatively calm air to avoid contamination by atmospheric motions (i.e., waves and turbulence). During these flights, a series of MMS maneuvers was often repeated at a series of altitudes and aircraft velocities, permitting efficient reanalysis and update of MMS calibration statistics.

The primary products of MMS are pressure (precision of ±0.3 hPa with accuracy of 0.5%), temperature (±0.3 K, 0.2%), horizontal wind (±1m/s, ~3.3%) and vertical wind (±0.3 m/s). The derived parameters are potential temperature, true air speed, turbulence dissipation rate and Reynolds number. MMS reporting parameters are GPS positions, velocities, accelerations, pitch, roll, yaw, heading, angle of attack, angle of sideslip, dynamic total pressure, and total temperature.

3 Sampling strategy

AJAX flights were typically 2 hours in duration, starting and ending at Moffett Field, CA (37.415 °N, 122.050 °W); take-off times were generally between 10:00 and 12:00 local time. Horizontally the measurements covered an area of 32° to 42° N and -125° to -115° W (Figure 1, middle panel). Flight patterns varied considerably depending on the flight objective (see Table 1), which was determined ~1 week prior to the flight date, based on weather, science, and aircraft availability. Some of the more common AJAX flight patterns are discussed below.

3.1 Tropospheric profiling

AJAX has a long-term record of designing and executing science flights to perform tropospheric profiling, often coincident with satellite overpasses, a Total Carbon Column Observing Network (TCCON) site, or ozonesonde launches from Trinidad Head, CA (THD). The sustained, long-term approach by AJAX allows for validation of remotely-sensed data products during the evolution of the instrument/mission lifetime, providing a unique dataset and a powerful asset for continued data evaluation. In total, vertical profiles of trace gases (≥8 km to boundary layer) have been recorded in 127 times in the AJAX data record, including 58 flights over RRV with the primary purpose of collecting CO_2 and CH_4 measurements for validation of GOSAT, GOSAT-2 and OCO-2 satellite data products (Tadić et al., 2014; Tanaka et al., 2016). In addition, AJAX flights targeting offshore vertical profiles coincident with OCO-2 provided data aiding the development of a lowermost tropospheric CO_2 partial column satellite product (Kulawik et al., 2017). AJAX has also collected trace gas vertical measurements coincident with TROPOMI and a TCCON site based at NASA Armstrong Flight Research Center (NASA AFRC), formerly known as Dryden Flight Research Center.

The sampling strategy for a tropospheric profiling flight generally followed this pattern: ascent from Moffett Field reaching a cruise altitude of ~8 km shortly after take-off for transit to a desired location, followed by a descending, spiraling profile with a descent rate of 10 m/s and horizontal diameter of <9 km The bottom of the profile can be at or below 50 m above ground level in accordance with Federal Aviation Regulation in sparsely populated areas (FAR, 2023). After completion of the vertical profile, AJAX typically ascended and returned to Moffett Field with the return transit leg at a lower altitude (e.g., 5 km) than

220   the outbound transit leg. AJAX observations of CO_2 and CH_4 over RRV measured during descending profiles are presented in Figure 2. Annual increases in CO_2 and CH_4 are observed over this long-term record, as discussed further in Section 4.

3.2 Boundary layer observations

AJAX has a record of regular, low-altitude flights observing the San Francisco Bay Area (SFBA), California's Central Valley (CCV) and offshore boundary layers. AJAX take-off and landing data provide a long-term record of the SFBA boundary layer,

225   which has currently been underused. AJAX executed regular flights with the main objective to measure within the SFBA (13 flights), CCV (85 flights) and offshore (14 flights) boundary layers to better understand emission sources and transport. The sampling strategy of boundary layer flights was typically: take-off from Moffett Field and transit towards the first desired location at an altitude left to pilots' discretion. On arrival at the first sampling location AJAX would descend to ≤300 m and from there would sample at altitudes ≤300 m through several designated locations within the study region before transiting

230   and returning to Moffett Field, providing ~1.5 hours of boundary layer observations within a 2-hour flight. An example AJAX flight focusing on boundary observations is shown in Figure 3, which displays O_3, CH_4 and CO_2 collected on one day of a mini-intensive coordinated during a seasonal deployment of multiple EM27/SUN ground-based spectrometers across the San Francisco Bay Area of California (Klappenbach et al., 2021). The flight plan was devised to provide in-situ greenhouse gas measurements near and above multiple column observations on sequential days in Fall 2016.

235   ## 3.3 Source identification

AJAX had several flight objectives to sample and help quantify emissions sources, including wildfires (17 flights), emissions from urban centers (14 flights) and oil and gas infrastructure (17 flights). These flights required a sampling approach that firstly aims to obtain an upwind, clean-air sample before targeting observations from the emission source itself. A typical sampling strategy would be as follows: take-off from Moffett Field, ascend to a transit altitude (pilot discretion), perform a

240   vertical profile descending from ≤ ~8 km to within the boundary layer, upwind as close to the emission source as feasible. AJAX would then perform several circles or passes around the emission source within the boundary layer. Often several altitudes were flown, allowing for 2-3 circles around the source before ascending for the return transit (altitude at pilot's discretion) and landing at Moffett Field. An example source identification AJAX flight is shown in Figure 4 from a flight over the Aliso Canyon, CA natural gas leak in December 2015 (34.315 °N, 118.564 °W). The observed, uncorrelated CO_2 and

245   methane are due to difference in emission sources. High CO_2 was observed downwind of Aliso Canyon and is associated with transport of pollution from the LA basin, whereas extremely high methane values are observed in the vicinity of Aliso Canyon.

4 Dataset overview and demonstrations

Given the wide variety of information contained in the AJAX dataset, here we present an overview of the dataset designed to highlight the data applications. This includes long-term trends observed by AJAX, an example of the changing boundary layer

250 trace gas concentrations relevant to air quality studies and a case study highlighting the use of AJAX data in support of satellite validation of lowermost tropospheric products from OCO-2 and GOSAT.

4.1 Long-term observations and trends

The multi-season, multi-year, multi-objective sampling approach adopted by the AJAX program allows for detailed analysis of a range of questions related to climate and air quality as well as atmospheric trends. For example, Figure 2 shows the

255 increasing mixing ratios of CO_2 and CH_4 between 2011 and 2018 as observed at RRV, NV.

Figure 7 shows the average trace gas mixing ratios for CO_2, CH_4, and O_3 over the entirety of each AJAX flight. The average of measurements within the boundary layer (BL, defined as <2 km) and averages within the free troposphere (FT, defined as >2 km) are also shown for each AJAX flight. Presented in grey are the monthly average mixing ratios from atmospheric baseline stations (Mauna Loa (MLO) for CO_2 and CH_4 (NOAA, 2022a) and Trinidad Head (THD) for O_3 (NOAA, 2022b)).

260 The mean AJAX observations of CO_2 (Figure 7, top panel) are in such good agreement with the surface trends at MLO that they lie directly underneath the other plotted data making them difficult to observe in Figure 7. AJAX FT CO_2 agrees slightly better than AJAX BL CO_2, which is influenced by local source emissions.

AJAX free tropospheric observations of CH_4 (middle panel) track the general trends in the MLO data but are offset slightly (typically less than 50 ppb). The boundary layer observations of CH_4 show the impact of emissions, with most flights observing

265 large CH_4 AJAX BL values, relative to the FT.

O_3, as a secondary pollutant, is impacted by a range of processing conditions (UV, temperature, etc.) and emissions sources. Thus, most AJAX BL O_3 observations show larger and more variable values relative to those observed within the marine boundary layer at the THD surface site, likely indicating that AJAX BL observations are more representative of continental air masses. Detailed comparisons of AJAX to other vertically resolved ozone observations (ozonesondes and lidar) are

270 presented in Langford et al. (2019) and Yates et al. (2017).

Figure 8 (right) shows the relationship between CH_4 and CO_2 measured by AJAX categorized by altitude, showing that observations in the FT (representative of the well-mixed atmosphere) are more closely correlated, while the relationship between CH_4 and CO_2 in the boundary layer is complex, with a wide variety of sources resulting in a large spread of correlations. But when these data are categorized by year (Fig. 8 (left)), patterns can be seen, including the annual increase in

275 CO_2 and CH_4. In 2011, CO_2 was observed between a minimum of 382 ppmv and a maximum of 397 ppmv, and by 2018 this range was 401-453 ppmv. The average increase in the annual mean CO_2 observed by AJAX was ~3.0 ppm/yr between 2011 and 2018. This estimate is in line with global growth rates of CO_2 reported by NOAA from observations at marine surface sites which averaged at 2.43 ppm/yr and varied between 1.90 and 3.03 ppm/yr over the same period (NOAA, 2022). Likewise, the increase in CH_4 between 2011 and 2018 corresponds to an increase in the mean annual CH_4 of ~10 ppbv/yr, which also

280 compares well with the annual global increase of CH_4 reported by NOAA, which averaged at 7.69 ppb/yr and varied between 4.85 and 12.73 ppb/yr over the same period (NOAA, 2022).

4.2 Observations of formaldehyde and ozone in a changing boundary layer

During the California Baseline Ozone Transport Study (CABOTS) campaign in spring and summer 2016 (Faloona et al., 2020) AJAX performed twelve flights (Iraci et al., 2020), several with multiple vertical profiles along a west-to-east transect as shown in Figure 9. This flight track provided an opportunity to assess the changes in the photochemical state of the boundary layer over a relatively short spatial distance (~250 km) between the western-most, offshore profile and the eastern-most profile over Visalia (36.319 °N, 119.393 °W), in California's San Joaquin Valley (SJV). This helped to address one of the main aims of CABOTS, to observe the changes in O_3 along the coast (upwind of California) and within California's Central Valley (Faloona et al., 2020). The SJV is the lowermost part of California's Central Valley and is an area that continues to exceed health-based air quality standards for hazardous air pollutants, including O_3, PM 2.5 and PM 10. The ability of AJAX to observe multiple trace gas species complemented the additional CABOTS measurement suite, including ozonesondes and ozone lidar. For example, HCHO is the third-most abundant oxygenated volatile organic compound in the SJV (Liu et al., 2022), and it undergoes complex photochemical reactions resulting in the production of O_3.

Figure 9 compares the west-to-east profiles for each trace gas species measured by AJAX. Each species shows a good degree of correlation at all locations in the free troposphere (>~2 km). However, in the lower troposphere each species shows significant differences at the varied locations. For O_3, HCHO and CH_4 there is a trend to increasing values from west to east, with the smallest mixing ratios for O_3, HCHO, CO_2 and CH_4 observed offshore at Pt Sur, and the largest mixing ratios over Visalia (KVIS) in line with the SJV's typical spring/summer conditions (high UV, temperature) favoring photochemical processing and O_3 formation in the lowermost troposphere. For H_2O, the profile over Visalia has a similar water vapor content within the boundary layer as the offshore profile due to the presence of rivers, crop irrigation and transpiration (Cooper et al., 2011).

4.3 Satellite validation case study: AJAX data for validation of Lowermost Tropospheric (LMT) products from OCO-2 and GOSAT

AJAX observations of CO_2 can be used to validate OCO-2 lowermost troposphere (LMT; approximately surface to 2.5 km, or surface to 750 hPa) and GOSAT lower troposphere (LT; defined by the retrieved surface pressure (P_{surf}) as 0.6-1 P_{surf} or approximately surface - 510 hPa) estimates of CO_2, and can, to a lesser extent, validate the OCO-2 upper column (U; approximately above 2.5 km, or 750 hPa) and GOSAT upper troposphere (UT; 0.2-0.6 P_{surf}) of CO_2.

To compare the satellite observations to aircraft, we calculate an estimate of how the satellite would observe the AJAX extended profile, given the satellite's sensitivity (averaging kernel), which are different for OCO-2 and GOSAT partial column quantities, as shown in Figure 10(left). The satellite to aircraft comparison is accomplished using Equation 1, where z is the partial column, x is the profile, ak is the averaging kernel, and the a subscript represents the a priori information. $z_{true,ak}$

represents the expected satellite observation of the AJAX extended profile (Rodgers, 2000; Connor et al., 2008; Kulawik et al. 2017).

$$z_{true,ak} = z_a + \sum_i ak_i * (x_{true,i} - x_{a,i}) \tag{1}$$

Since AJAX observations extend from near-surface to ~8-9 km, the AJAX profile needs to be extended both upward and downward to compare to a satellite column data product. Aircraft measurements are extended down to the surface using the lowest measured value. Between the top aircraft observation and 100 hPa, we extended the AJAX profile using the CarbonTracker 2019B model (Jacobson et al., 2020) with a constant offset to match to the topmost AJAX observation. Above 100 hPa, the CarbonTracker 2019B model without offset is used. Detailed overview of the LMT and U partial column products is described by Kulawik, et al. (2017) and LT and UT by Kuze et al. (2022).

Figure 10(right) shows an example of a CO_2 profile measured by AJAX (black); the OCO-2 and GOSAT priors (green), the total column CO_2 (XCO2) measured by OCO-2 (grey), and partial column products. These data were collected on 13 April 2018 during regular nadir overpasses (looking straight down) of both satellites. The satellite observations are plotted as solid lines, with horizontal bars showing the standard deviation of all nearby satellite observations. The GOSAT upper troposphere (UT, dark blue) shows a value slightly larger than XCO2, and the OCO-2 upper partial column (U, light blue) has a lower value than XCO2. OCO-2 and GOSAT both show an enhancement in the lower troposphere (orange and red solid lines, respectively) as compared to XCO2 (grey solid line).

AJAX data with the respective averaging kernels (using Eq. 1) are shown with dotted lines of colors matching the individual satellite partial products. The calculated LMT (orange dotted) and LT (red dotted) partial column averages are very similar to each other and to the direct observations (black). Thus, the AJAX data validate the vertical gradient observed by both GOSAT and OCO-2, with all matches within the satellite errors (estimated by the variability of all satellite matches). Validation of these exciting new GOSAT and OCO-2 vertically resolved products using AJAX in situ data shows the unique usefulness of aircraft profiles and the AJAX dataset of co-located vertical profiles in particular. A future paper will broaden this validation using a complete set of AJAX coincident observations over the period 2015-2018.

5 Data overview and availability

The AJAX data described in this manuscript are freely available at NASA's Atmospheric Science Data Center (https://asdc.larc.nasa.gov/project/AJAX, doi: 10.5067/ASDC/SUBORBITAL/AJAX/DATA001, Iraci et al. 2021a). The data are provided in ICARTT format, which is described at https://www.earthdata.nasa.gov/esdis/esco/standards-and-practices/icartt-file-format lite product is available through NASA Goddard Earth Sciences Data and Information Services Center (GES DISC) at: https://disc.gsfc.nasa.gov/datasets/OCO2_L2_Lite_FP_10r/summary?keywords=oco-2%20r10.

For each AJAX flight, four individual instrument datasets are available for download following the ICARTT file format V2.0. The CO_2 + CH_4 dataset (3 s resolution) and O_3 dataset (10 s resolution) each include time and aircraft position. The HCHO dataset is available at 1 s and includes a time stamp; GPS data can be inferred from either the MMS or greenhouse gas datasets.

345 The MMS dataset includes meteorological parameters and aircraft position archived at 1 s. All AJAX Level 2 datasets (https://doi.org/10.5067/ASDC/SUBORBITAL/AJAX/DATA001, Iraci et al., 2021a) are readily available from NASA's Atmospheric Science Data Center (NASA ASDC, https://asdc.larc.nasa.gov/project/AJAX).

GOSAT data used for this paper are available through Japan Aerospace Exploration Agency (JAXA) GOSAT/GOSAT-2 EROC daily partial column GHG website available at: https://www.eorc.jaxa.jp/GOSAT/CO2_monitor/index.html.

350 **6 Summary**

Measurements collected by the AJAX program provide an extensive dataset of trace gas and meteorological observations that will be of interest to a wide variety of data users. The multi-year, multi-objective, multi-instrument approach provided by the AJAX program was unique from a NASA airborne science perspective and produced a long-term record of vertical profile (up to 8-9 km) and level leg measurements over California and Nevada. The regular sampling approach and ability to vary flight

355 objectives based on season, synoptic and atmospheric conditions allowed for opportunistic flights and flexibility to address several science questions. The AJAX dataset is of interest to those researching areas related to (but not limited to) 1) tropospheric profiling, including satellite validation; 2) boundary layer observations in California, with an emphasis on the San Francisco Bay Area and California's Central Valley; and 3) source identification, including wildfires, urban outflow and emissions from oil and gas and agriculture.

360 This paper provides a detailed overview of the AJAX program and its dataset, including a summary of flight objectives, sampling strategies, airborne instrumentation, data processing and a presentation of the overall datasets and their availability. AJAX observations can be used to validate lowermost troposphere satellite data products as shown in the case study presented in Section 4.3.

Author contribution

365 Funding acquisition, project administration and supervision were carried out by LTI. JEM, J-MR, CLP and ELY worked on data curation, formal analysis, and visualisation. SK worked on validation of OCO-2 products using AJAX data. JMS, TFH, TPB, CC and JD-D provided resources and performed investigations. ELY prepared the manuscript with contributions from all co-authors.

Acknowledgements

370 The NASA AJAX program recognizes support from Ames Research Center Director's funds, the NASA Postdoctoral Program (JEM, J-MR, CLP, JT, TT, ELY), the OCO-2 Science Team, the Rapid Response and Novel research in the Earth Sciences (RRNES) program element, and the California Air Resources Board (Contract No. 17RD004), as well as by NASA's Atmospheric Composition Program through the Internal Scientist Funding Model and the Campaign Data Analysis and Modeling (20-ACCDAM20-0083) program. LTI acknowledges support from the NASA Earth Science Research and Analysis

375 Program during data collection and analysis. TFH and JSC acknowledge support from the Goddard Internal Research and Development (IRAD) program. Technical contributions from C. Camacho, W. Gore, P. Hamill, E. Quigley, M. Roby, J. Tadic, T. Tanaka, T. Trias, and Z. Young made this program possible. The authors gratefully recognize the support and partnership of H211 L.L.C., with particular thanks to K. Ambrose, T. Cardoza, R. Fisher, T. Grundherr, J. Kerr, J. Lee, B. Quiambao, R. L. Sharma, D. Simmons and R. Simone. Data discovery, accessibility and archiving were made possible through the efforts of

380 the Atmospheric Science Data Center staff: K. Phillips, M. Buzanowicz, N. Jester, N. Arora, and S. Haberer. The OCO-2 lowermost tropospheric analysis was funded from ROSES 17-OCO2-17-0013 project, "Reducing the impact of model transport error on flux estimates using CO2 profile information from OCO2 in concert with an online bias correction." JAXA-GOSAT partial column density products were funded by JAXA GOSAT program. Authors gratefully acknowledge A. Kuze and H. Suto of JAXA for consultation regarding the GOSAT partial column products. The TMF data used in this publication

385 were obtained from Dr. Thierry Leblanc as part of the Network for the Detection of Atmospheric Composition Change (NDACC) and are available through the NDACC website www.ndacc.org.

References

Baker, K. R., Woody, M. C., Valin, L., Szykman, J., Yates, E. L., Iraci, L. T., Choi, H. D., Soja, A. J., Koplitz, S. N., Campuzano-Jost, P., Jimenez, J. L., and Hair, J, W.: Photochemical model evaluation of 2013 California wild fire air quality

390 impacts using surface, aircraft and satellite data, Sci. Total Environ., 637, doi: 10.1016/j.scitotenv.2018.05.048, 2018.

Chang, K.-L., Cooper, O. R., Rodriguez, G., Iraci, L. T., Yates, E. L., Johnson, M. S., Gaudel, A., Jaffe, D. A., Bernays, N., Clark, H., Effertz, P., Leblanc, T., Petropavlovskikh, I., Sauvage, B., and Tarasick, D. W.: Diverging ozone trends above western North America boundary layer decreases vs. free tropospheric increases, submitted, in-review, 2022.Chen, H., Winderlich, J., Gerbig, C., Hoefer, A., Rella, C. W., Crosson, E. R., Van Pelt, A. D., Steinbach, J., Kolle, O., Beck, V., Daube,

395 B. C., Gottlieb, E. W., Chow, V. Y., Santoni, G. W., and Wofsy, S. C.: High-accuracy continuous airborne measurements of greenhouse gases (CO_2 and CH_4) using the cavity ring-down spectroscopy (CRDS) technique, Atmos. Meas. Tech., 3(2), 375-386. doi: 10.5194/amt-3-375-2010, 2010.

Cooper, O.R., Oltmans, S.J., Johnson, B.J., Brioude, J., Angevine, W., Trainer, M., Parrish, D.D., Ryerson, T.R., Pollack, I., Cullis, P.D., Ives, M.A., Tarasick, D.W., Al-Saadi, J., Stajner, I.: Measurement of western U.S. baseline ozone from the surface

400 to the tropopause and assessment of downwind impact regions. J. Geophys. Res. 116, doi:10.1029/2011JD016095, 2011.

Connor, B. J., H. Boesch, G. Toon, B. Sen, C. Miller, and D. Crisp, Orbiting Carbon Observatory: Inverse method and prospective error analysis, J. Geophys. Res., 113, D05305, doi:10.1029/2006JD008336, 2008.

Cui, Y. Y., Vijayan, A., Falk, M., Hsu, Y.-K., Yin, D., Chen, X. M., Zhao, Z., Avise, J., Chen, Y., Verhulst, K., Duren, R., Yadav, V., Miller, C., Weiss, R., Keeling, R., Kim, J., Iraci, L. T., Tanaka, T., Johnson, M. S., Kort, E. A., Bianco, L., Fischer, M. L., Stroud, K., Herner, J., and Croes, B.: A Multiplatform Inversion Estimation of Statewide and Regional Methane Emissions in California during 2014–2016, Environ. Sci. Technol., 53 (16), 9636-9645, doi: 10.1021/acs.est.9b01769, 2019.

Faloona, I. C., Chiao, S., Eiserloh, A. J., Alvarez, R. J., Kirgis, G., Langford, A. O., Senff, C. J., Caputi, D., Hu, A., Iraci, L.T., Yates, E. L., Marrero, J. E., Ryoo, J-M., Conley, S., Tanrikulu, S., Xu, J., and Kuwayama, T.: The California Baseline Ozone Transport Study (CABOTS), Bull. Am. Meteorol. Soc., 101, doi: 10.1175/BAMS-D-18-0302.A, 2020.

Federal Aviation Regulation (FAR), 91.119 Minimum safe altitudes: https://www.faa.gov/about/office_org/field_offices/fsdo/lgb/local_more/media/FAA_Guide_to_Low-Flying_Aircraft.pdf last access: 3 January 2023.

Fine, R., Miller, M. B., Yates, E. L., Iraci, L. T., and Sexauer Gustin, M.: Investigating the influence of long-range transport on surface O_3 in Nevada, using observations from multiple measurement platforms, Science of the Total Environment, 530-531, doi: 10.1016/j.scitotenv.2015.03.125, 2015.

GOSAT: https://www.eorc.jaxa.jp/GOSAT/GPCG/index_GOSAT.html last access: 3 January 2023.

Hamill, P., Iraci, L. T., Yates, E. L., Gore, W., Bui, T. P., and Tanaka, T.: A new instrumented airborne platform for atmospheric research, Bull. Am. Meteorol. Soc., 97(3), 397-404. doi: 10.1175/bams-d-14-00241.1, 2016.

Iraci, L. T., Ryoo, J.-M., and Yates, E. L.: Spatial Variation of Vertical Ozone Distribution over California, Final Project Report for Contract No. 17RD004 submitted to California Air Resources Board, 2020.

Iraci, L. T., Yates, E. L., Marrero, J. E., Parworth, C. L., Ryoo, J.-M., Tanaka, T., Bui, T. P., Dean-Day, J., Chang, C., Hanisco, T., and St. Clair, J.: Airborne Trace Gas Measurements Collected by the Alpha Jet Atmospheric eXperiment (AJAX) Project: 2011 – 2018, available at Atmospheric Science Data Center [data set], https://doi.org/10.5067/ASDC/SUBORBITAL/AJAX/DATA001, 2021a.Iraci, L. T., Yates, E. L., Marrero, J. E., Parworth, C. L., Ryoo, J.-M., and Tanaka, T.: Compendium of Airborne Trace Gas Measurements Collected in and around California Fire Plumes by the AJAX Project, available at Atmospheric Science Data Center [data set], https://doi.org/10.5067/ASDC/AJAX/wildfire, 2021b.

Iraci, L.T., Parworth, C.P. Yates, E. L., Marrero, J.E., and Ryoo, J-M.: A collection of airborne measurements and analyses of trace gases emitted from multiple fires in California, Earth Space Sci., 9, EA002116, doi: 10.1029/2021EA002116, 2022.

Jacobson, A. R., Schuldt, K. N., Miller, J. B., Oda, T., Tans, P., Mund, J., Ott, L., Collatz, G. J., Aalto, T., Afshar, S., Aikin, K., Aoki, S., Apadula, F., Baier, B., Bergamaschi, P., Beyersdorf, A., Biraud, S. C., Bollenbacher, A., Bowling, D., Brailsford, G., Abshire, J. B., and Chen, G.: CarbonTracker CT2019B. Model published 2020 via NOAA Global Monitoring Laboratory, 2020.

Johnson, M. S., Yates, E. L., Iraci, L. T., Loewenstein, M., Tadić, J. M., Wecht, K. J., Jeong, S., and Fischer, M. L.: Analyzing

435   source apportioned methane in northern California during Discover-AQ-CA using airborne measurements and model simulations, Atmos. Environ., 99, doi: 10.1016/j.atmosenv.2014.09.068, 2014.

Johnson, M. S., Xi. X., Jeong, S., Yates, E. L., Iraci, L. T., Tanaka. T., Loewenstein, M., Tadíc, J. M., and Fischer, M. L.: Investigating seasonal methane emissions in Northern California using airborne measurements and inverse modelling, J. Geophys. Res., 22, doi: 10.1002/2016JD025157, 2016.

440   Klappenbach, F., Chen, J., Wenzel, A., Forstmaier, A., Dietrich, F., Zhao, X., Jones, T., Franklin, J., Wofsy, S., Frey, M., Hase, F., Hedelius, J., Wennberg, P., Cohen, R., and Fischer, M.: Methane emission estimate using ground based remote sensing in complex terrain, EGU General Assembly 2021, online, 19-30 April2021, EGU21-15406, https://doi.org/10.5194/egusphere-egu21-15406, 2021.

Kulawik, S. S., O'Dell, C., Payne, V. H., Kuai, L., Worden, H. M., Biraud, S. C., Sweeney, C., Stephens, B., Iraci, L. T., Yates,

445   E. L., and Tanaka, T.: Lower-tropospheric CO_2 from near-infrared ACOS-GOSAT observations, Atmos. Chem. Phys., 17, doi: 10.5194/acp-17-5407-2017, 2017.

Kuze,A., Nakamura,Y., Oda,T., Yoshida,Y., Kikuchi,N., Kataoka, F., Suto, H., and Shiomi, K.: Examining partial-column density retrieval of lower-tropospheric $CO2$ from GOSAT target observations over global megacities, Remote Sens. Environ., 273, 112966, doi: 10.16/j.res.2022.112966, 2022.

450   Langford, A. O., Senff, C., Alvarez II, R. J., Brioude, J., Cooper, O. R., Holloway, J. S., Lin, M. Y., Marchbanks, R. D., Pierce, R. B., Sandberg, S. P., Weickmann, A. M., Williams, E. J.: An overview of the 2013 Las Vegas Ozone Study (LVOS): Impact of stratospheric intrusions and long-range transport on surface air quality, Atmos. Environ., 109, doi: 10.1016/j.atmosenv.2014.04.040, 2015.

Langford, A. O., Alvarez II, R. J., Brioude, J., Evan, S., Iraci, L. T., Kirgis, G., Kuang, S., LeBlanc, T., Newchurch, M. J.,

455   Pierce, R. B., Senff, C., and Yates, E.L.: Coordinated profile of stratospheric intrusions and transported pollution by the Tropospheric Ozone Lidar Network (TOLNet) and NASA Alpha Jet eXperiment (AJAX): Observations and comparison to HYSPLIT, RAQMS and FLEXPART, Atmos. Environ., 174, doi: 10.1016/j.atmosenv.2017.11.031, 2018.

Langford, A. O., Alvarez, R. J., Kirgis, G., Senff, C. J., Caputi, D., Conley, S. A., Faloona, I. C., Iraci, L. T., Marrero, J. E., McNamara, M. E., Ryoo, J.-M., and Yates, E. L.: Intercomparison of lidar, aircraft, and surface ozone measurements in the

460   San Joaquin Valley during the California Baseline Ozone Transport Study (CABOTS), Atmos. Meas. Tech., 12, 1889-1904, doi: 10.5194/amt-12-1889-2019, 2019.

Langford, A.O., Alvarez, R. J., Brioude, J., Caputi, D., Conley, S. A., Evan, S., Faloona, I. C., Iraci, L. T., Kirgis, G., Marrero, J. E., Ryoo, J-M., Senff, C., and Yates, E. L.: Ozone production in the Soberanes smoke haze: Implications for air quality in the San Joaquin Valley during California Baseline Ozone Transport Study, J. Geo. Phys. Res., 125, doi:

465   10.1029/2019JD031777, 2020.

Leifer, I., Melton, C., Fischer, M. L., Fladeland, M., Frash, J., Gore, W., Iraci, L. T., Marrero, J. E., Ryoo, J-M., Tanaka, T., and Yates, E.L.: Atmospheric characterization through fused mobile airborne and surface in situ surveys: Methane emissions quantification from a producing oil field, Atmos. Meas. Tech., 11, doi: 10.5194/amt-11-1689-2018, 2018.

470 Leifer, I., Melton, C., Chatfield, R., Cui, X., Fischer, M. L., Fladeland, M., Gore, W., Hlavka, D. L., Iraci, T. L., Marrero, J. E., Ryoo, J-M., Tanaka, T., Yates, E. L., and Yorks, J. E.: Air pollution inputs to the Mojave Desert by fusing surface mobile and airborne in situ and airborne and satellite remote sensing: A case study of interbasin transport with numerical model validation, Atmos. Environ., 224, doi: 10.1016/j.atmosenv.2019.117184, 2020.

Lin, M., Horowitz, L. W., Cooper, O. R., Tarasick, D., Conley, S. C., Iraci, L. T., Johnson, B., LeBlanc, T., Petropavlovskikh, I., and Yates, E. L.: Revisiting the evidence of increasing springtime ozone mixing ratios in the free troposphere over western

475 North America, Geophys. Res. Lett., 42, doi: 10.1002/2015GL065311, 2015.

Liu, S., Barletta, B., Hornbrook, R. S., Fried, A., Peischl, J., Meinardi, S., Coggon, M., Lamplugh, A., Gilman, J. B., Gkatzelis, G. I., Warneke, C., Apel, E. C., Hills, A. J., Bourgeois, I., Walega, J., Weibring, P., Richter, D., Kuwayama, T., FitzGibbon, M., and Blake, D.: Composition and reactivity of volatile organic compounds in the South Coast Air Basin and San Joaquin Valley of California, Atmos. Chem. Phys., 22, 10937-10954, doi: 10.5194/acp-22-10937-2022, 2022.

480 NOAA, 2022a, https://gml.noaa.gov/ccgg/trends/data.html

NOAA, 2022b, https://gml.noaa.gov/dv/site/THD.html

Parworth, C. L., Marrero, J. M., Yates, E.L., Ryoo, J.-M., and Iraci, L. T.: Proposed trace gas measurements over the western United States for TROPOMI validation, 20th EGU General Assembly, EGU2018, Proceedings from the conference held 4-13 April, 2018 in Vienna, Austria, p.5672, 2018.

485 Rodgers, C. D.: Inverse methods for atmospheric sounding: theory and practice, London, WorldScientific, 2000.

Ryoo, J-M., Johnson, M. S., Iraci, L. T., Yates, E. L., and Gore, W.: Investigation sources of ozone over California using AJAX airborne measurements and models: Assessing the contribution from long-range transport, Atmos. Environ., 155, doi: 10.1016/j.atmosenv.2017.02.008, 2017.

Ryoo, J.-M., Iraci, L. T., Tanaka, T. Marrero, J. M., Yates, E. L., Fung, I., Michalak, A. M., Tadíc, J., Gore, W., Bui, T. P.,

490 Dean-Day, J. M., and Chang, C. S.: Quantification of CO_2 and CH_4 emissions over Sacramento, California, based on divergence theorem using aircraft measurements, Atmos. Meas. Tech., 12, doi: 10.5194/amt-12-2949-2019, 2019.

Ryoo, J. -M., Chiao, S., Spackman, J. R., Iraci, L. T., Ralph, F. M., Martin, A., Dole, R. M., Marrero, J. E., Yates, E. L., Bui, T. P., Dean-Day, J. M., and Chang, S. C.: Terrain Trapped Airflows and Precipitation Variability during an Atmospheric River Event, *J. Hydrometeor.*, 21, 355-375, doi: 10.1175/JHM-D-19-0040.1, 2020.

495 Scott, S. G., Bui, T. P., Chan, K. R., and Bowen, S. R.: The meteorological measurement system on the NASA ER-2 aircraft, J. Atmos. Ocean. Tech., 7, 525-540, https://doi.org/10.1175/1520-0426(1990)007%3C0525:TMMSOT%3E2.0.CO;2, 1990.

St. Clair, J. M., Swanson, A. K., Bailey, S. A., Wolfe, G. M., Marrero, J. E., Iraci, L. T., Hagopian, J. G., and Hanisco, T. F.: A new non-resonant laser-induced fluorescence instrument for the airborne in situ measurement of formaldehyde, Atmos. Meas. Tech., 10, 4833-4844. doi:10.5194/amt-10-4833-2017, 2017.

500   St. Clair, J. M., Swanson, A. K., Bailey, S. A., and Hanisco, T. F. (2019). CAFE: a new, improved nonresonant laser-induced fluorescence instrument for airborne in situ measurement of formaldehyde. Atmos. Meas. Tech., *12*(8), 4581-4590. https://doi.org/10.5194/amt-12-4581-2019.

Tadíc, J. M., Loewenstein, M., Frankenberg, C., Butz, A., Roby, M., Iraci, L.T., Yates, E. L., Gore, W., and Kuze, A.: A comparison of in situ aircraft measurements of carbon dioxide and methane to GOSAT data measured over Railroad Valley

505   playa, Nevada, USA. *IEEE* Trans. Geosci. Remote Sens., 52, 7764-7774. doi: 10.1109/TGRS.2014.2318201, 2014.

Tadíc, J. M., Michalak, A. M., Iraci, L. T., Llic, S. C., Biraud, S. C., Feldman, D. R., Bui, T. P., Johnson, M. S., Loewenstein, M., Jeong, S., Fischer, M. L., Yates, E. L., and Ryoo, J-M.: Elliptic cylinder airborne sampling and geostatistical mass balance approach for quantifying local greenhouse gas emissions, Environ. Sci. Tech., 51, doi: 10.1012/acs.est.7b03100, 2017.

Tanaka, T., Yates, E. L., Iraci, L. T., Johnson, M. S., Gore, W., Tadic, J. M., Loewenstein, M., Kuze, A., Frankenberg, C.,

510   Butz, A., and Yoshida, Y.: Two-year comparison of airborne measurements of CO_2 and CH_4 with GOSAT at Railroad Valley, Nevada, *IEEE* Trans. Geosci. Remote Sens., 54, 4367-4375. doi:10.1109/TGRS.2016.2539973, 2016.

Toon, O. B., Maring, H., Dibb, J., Ferrare, R., Jacob, D. J., Jensen, E. J., Luo, Z. J., Mace, G. G., Pan, L. L., Pfister, L., Rosenlof, K. H., Redemann, J., Reid, J. S., Singh, H. B., Thompson, A. M., Yokelson, R., Minnis, P., Chen, G., Jucks, K. W., Pszenny, A.: Planning, implantation, and scientific goals of the Studies and Emissions and Atmospheric Composition, Clouds

515   and Climate Coupling by Regional Surveys (SEAC4RS) field mission, J. Geophys. Res., doi: 10.1002/2015JD024297, 2016.

Wolfe, G. M., Hanisco, T. F., Arkinson, H. L., Blake, D. R., Wisthaler, A., Mikoviny, T., Ryerson, T. B., Pollack, I., Peischl, J., Wennberg, P. O., Crounse, J. D., St. Clair, J. M., Teng, A., Huey, L. G., Liu, X., Fried, A., Weibring, P., Richter, D., Walega, J., Hall, S. R., and Ullmann, K.: Photochemical evolution of the 2013 California Rim Fire: synergistic impacts of reactive hydrocarbons and enhanced oxidants, Atmos. Chem. Phys., 22, 4253-4275, doi: 10.5194/acp-22-4253-2022, 2022.

520   Yates, E. L., Iraci, L. T., Roby, M. C., Pierce, R. B., Johnson, M. S., Reddy, P. J., Tadíc, J. M., Loewenstein, M., and Gore, W.: Airborne observations and modeling of springtime stratosphere-to-troposphere transport over California, Atmos. Chem. Phys., 13(24), 12481-12494. doi: 10.5194/acp-13-12481-2013, 2013.

Yates, E. L., Iraci, L. T., Austerberry, D., Pierce, R. B., Roby, M. C., Tadíc, J. M., Loewenstein, M., and Gore, W.: Characterizing the impacts of vertical transport and photochemical ozone production on an exceedance area, Atmos. Environ.,

525   109, doi: 10.1016/j.atmosenv.2014.09.002, 2015.

Yates, E. L., Iraci, L. T., Singh, H., Tanaka, T., Roby, M. C., Hamill, P., Clements, C. B., Lareu, N., Contezac, J., Blake, D. R., Simpson, I. J., Wisthaler, A., Mikoviny, T., Diskin, G. S., Beyersdorf, A. J., Choi, Y., Reyerson, T. B., Jimenez, J. L., Campuzano-Jost, P., Loewenstein, M., and Gore, W.: Airborne measurements and emission estimates of greenhouse gases and other trace constituents from the 2013 California Yosemite Rim wildfire, Atmos. Environ., 127, 293-302. doi:

530   10.1016/j.atmosenv.2015.12.038, 2016.

Yates, E. L., Johnson, M. S., Iraci, L. T., Ryoo, J. -M., Pierce, R. B., Cullis, P. D., Gore, W., Ives, M. A., Johnson, B. J., Leblanc, T., Marrero, J. E., Sterling, C. W., and Tanaka, T.: An assessment of ground-level and free-tropospheric ozone over California and Nevada, J. Geophys. Res. 122. doi:10.1002/2016JD026266, 2017.

Yates, E. L., Iraci, L. T., Tarnay, L. W., Burley, J. D., Parworth, C., and Ryoo, J-M.: The effect of an upwind non-attainment area on ozone in California's Sierra Nevada Mountains, Atmos. Environ., 230, doi: 10.1016/j.atmosenv.2020.117426, 2020.

535

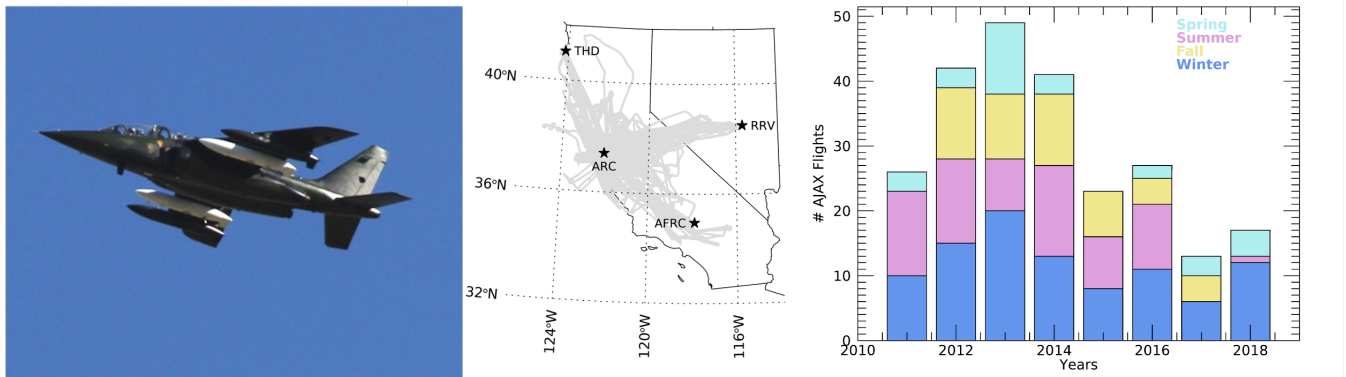



Figure 1: AJAX in flight, before landing at Ames Research Center (ARC), located at Moffett Field, CA (left), map of AJAX flight tracks (middle), and AJAX flights by year and season (right). (AFRC = Armstrong Flight Research Center, RRV = Railroad Valley, THD = Trinidad Head).

540

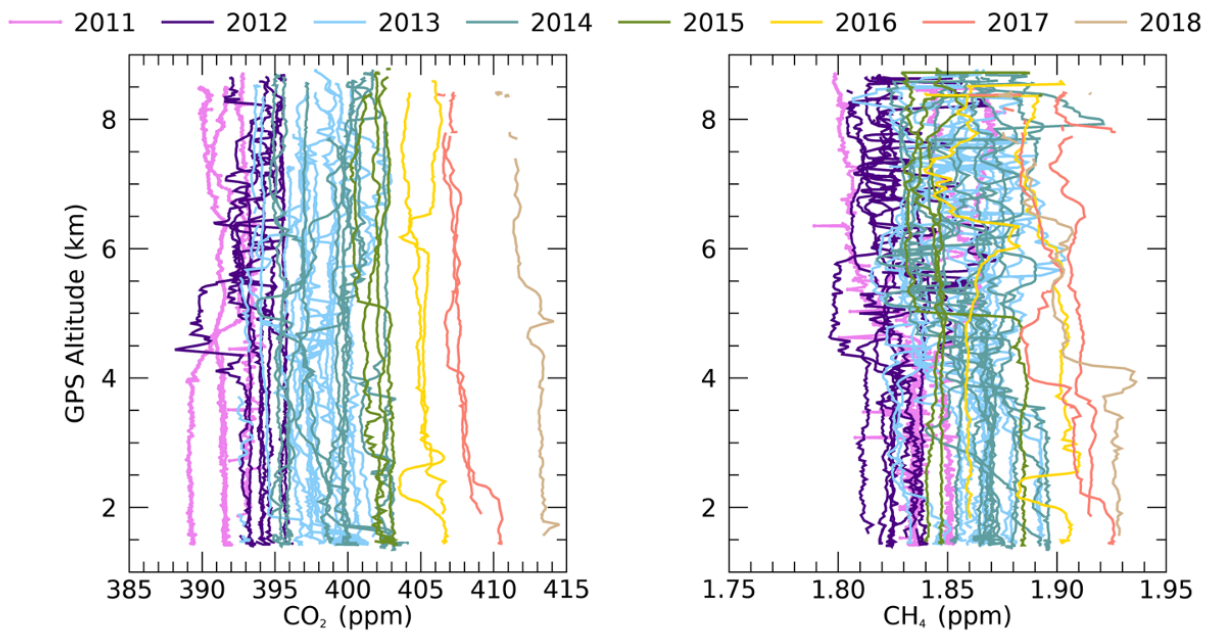



Figure 2. AJAX observations of CO₂ and CH₄ over Railroad Valley, NV (surface elevation = 1437 m asl), colored by year of observation.

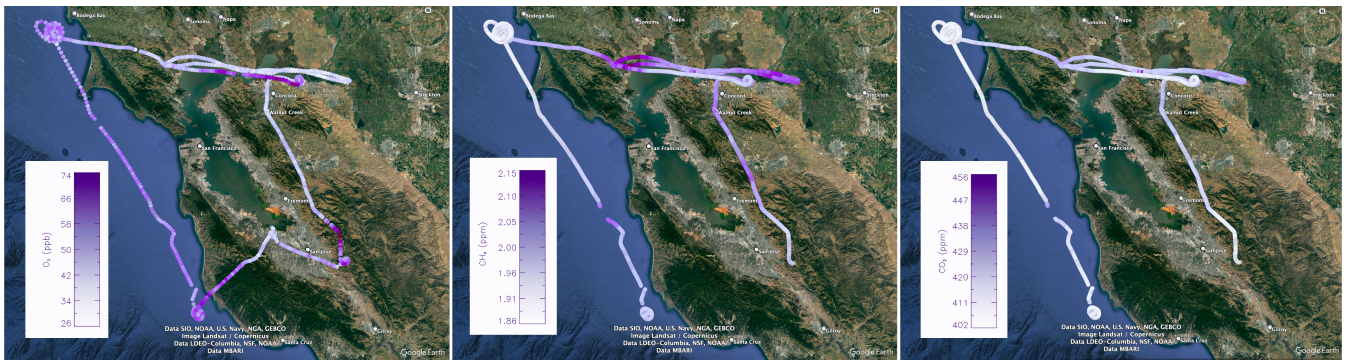



Figure 3. AJAX Flight #202 on 2 November 2016: Observations offshore and over the San Francisco Bay Area of ozone (left), methane (middle) and CO_2 (right) shown in the Google™ Earth map. Note that warm-up and shut down of the GHG instrument causes a loss of data on take-off (for most flights) and landing (for some flights; the post-flight procedures were modified in 2017 to retain landing data in most flights after this date).

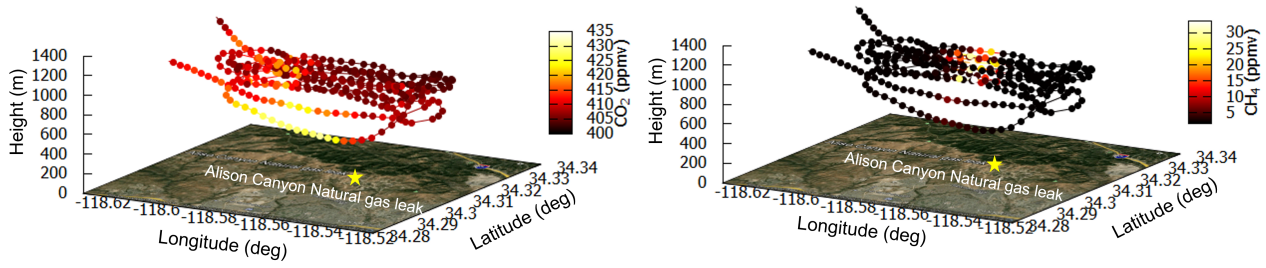



Figure 4. AJAX Flight #175 on 4 December 2015 sampling emissions from the Aliso Canyon gas leak shown in the Google™ Earth map. Maps show carbon dioxide (CO_2, left) and methane (CH_4, right) over the emission source.

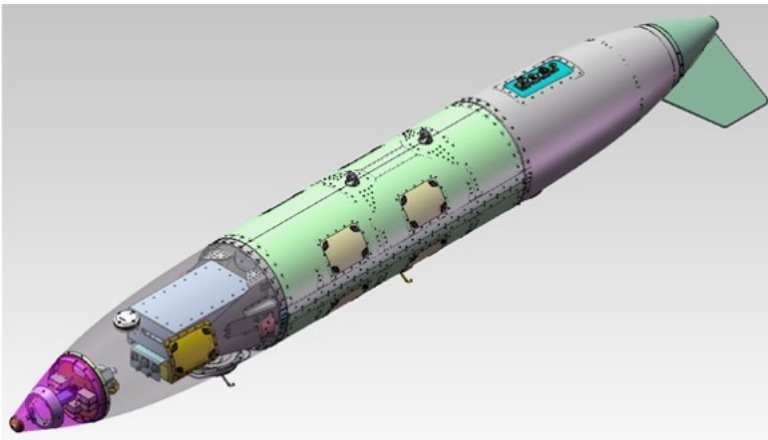



Figure 5. CAD drawing of an AJAX wing-pod showing the location of the ozone sensor and MMS in the nose-cone section of the wing-pod. The greenhouse gas sensor is located in the middle (green) section, and its inlet can be seen below the pod. Signal and electrical connections to the aircraft are made at the top, aft panel (cyan).

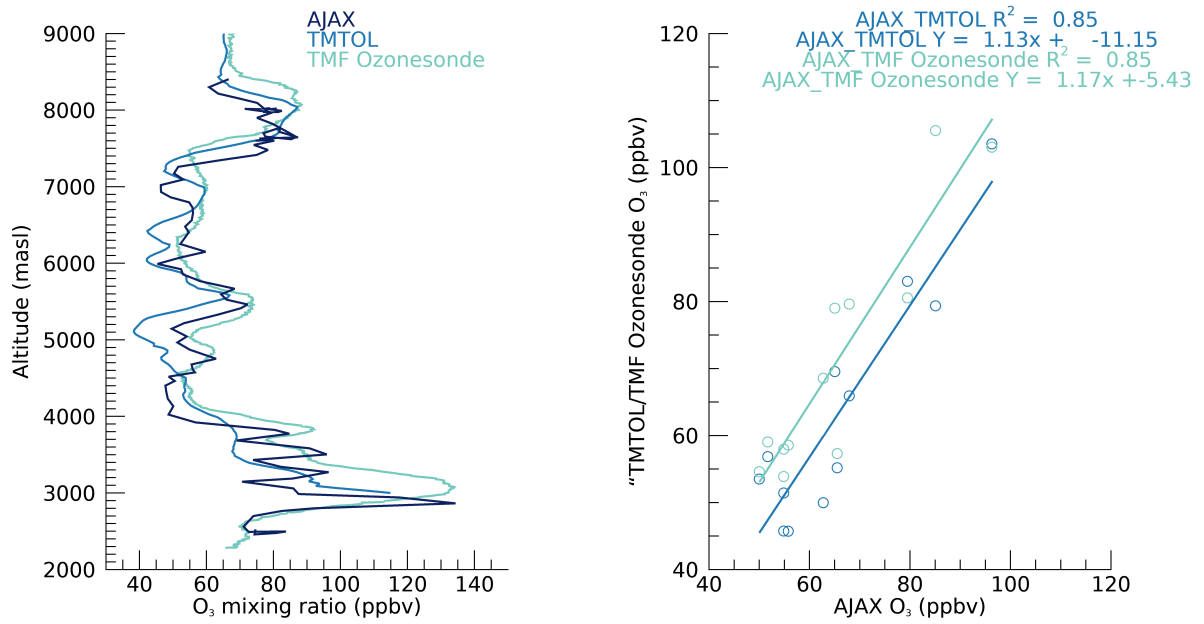



Figure 6. Ozone profiles taken on 24 May 2013 measured by AJAX, Table Mountain Facility tropospheric ozone lidar (TMTOL) and the Table Mountain Facility (TMF) ozonesonde. Correlations of AJAX observations with TMF ozonesonde and TMTOL measurements are shown in the right panel, based on the mean ozone within 500 m vertical resolution window.

560

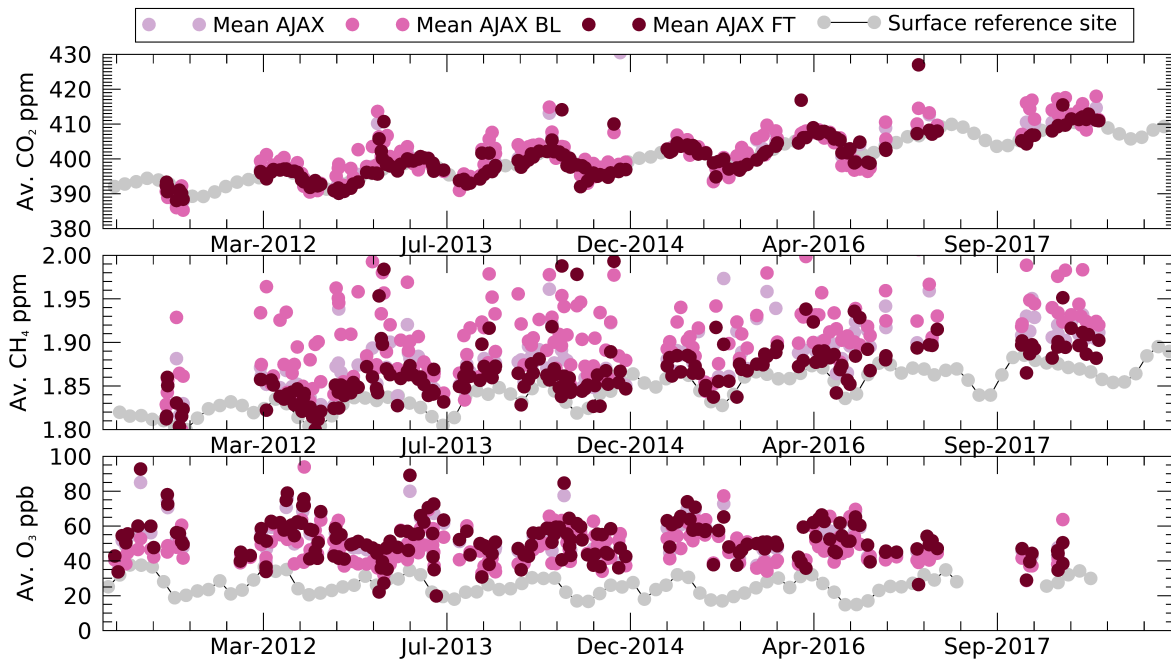
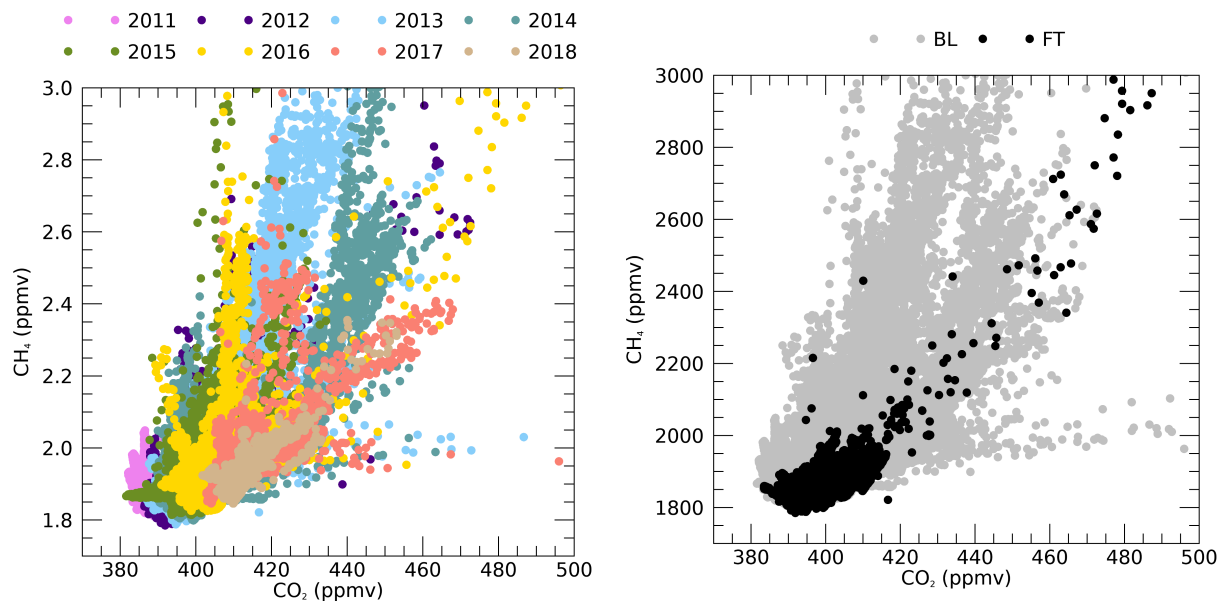



Figure 7. Average CO₂ (top), CH₄ (middle), O₃ (bottom) over the entirety of each individual AJAX flight, the average of data collected below 2 km from each individual flight and above 2 km. Monthly mean values from a surface reference site are shown for comparison: Mauna Loa for CO₂ and CH₄ and Trinidad Head for O₃.

565   **Figure 8. Relationship between CH₄ and CO₂ measured by AJAX separated by year (left) and by free troposphere/boundary layer (FT>3 km<BL) measurements (right).**

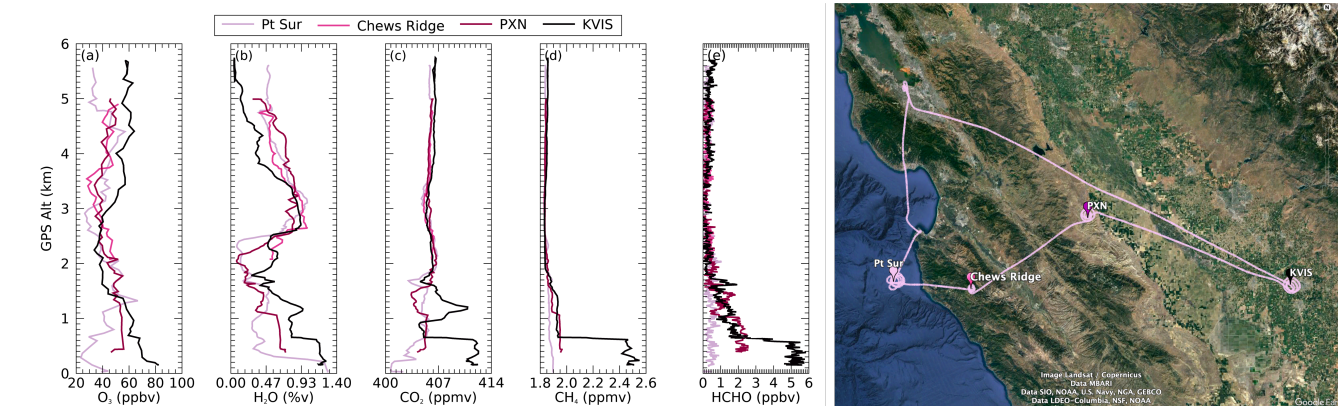



Figure 9. Vertical profiles of O₃ (a), H₂O (b), CO₂ (c), CH₄ (d) and HCHO (e) over Pt Sur (offshore, grey), Chews Ridge (elevation = 1521 m asl, pink), Panoche (PXN, elevation = 634 masl, purple) and Visalia (KVIS, black). AJAX flight track and profile locations
570   **are shown in the Google™ Earth map on the right. Data are taken from AJAX flight #192 on 21 July 2016.**

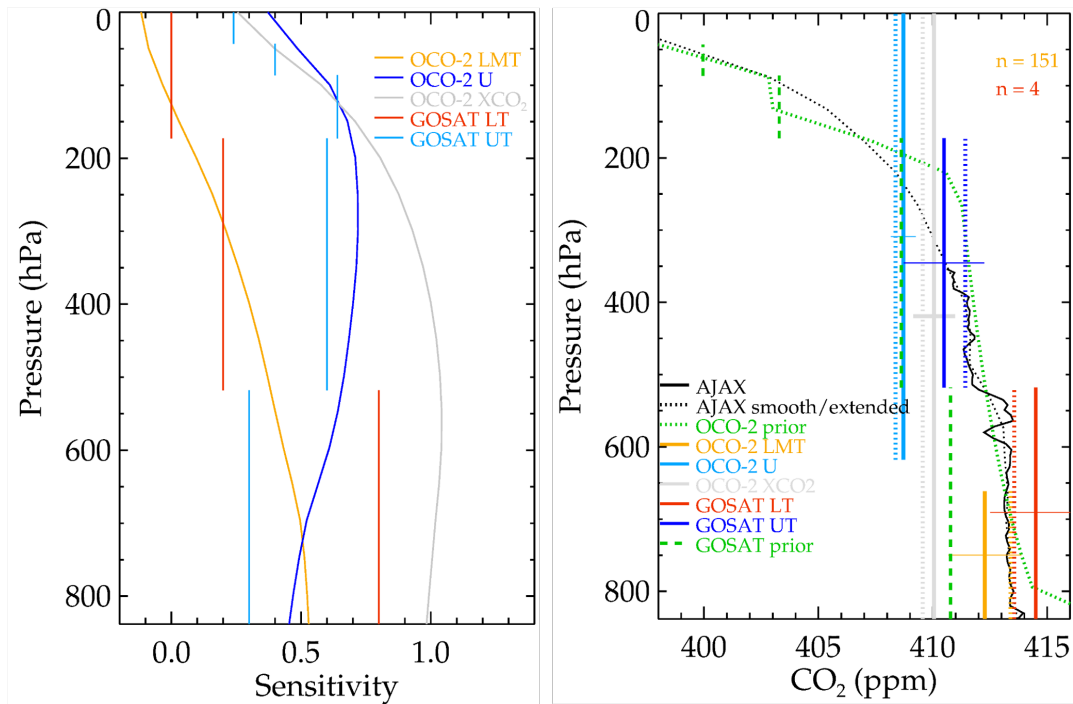



Figure 10. (left) Comparison of averaging kernel (sensitivities) of the different products, where 0 corresponds to no sensitivity and 1 corresponds to perfect sensitivity for OCO-2 LMT (orange), OCO-2 U (dark blue), OCO-2 XCO2 (gray), GOSAT LT (red) and GOSAT UT (light blue). Note that the GOSAT averaging kernel is on 5 layers, whereas the OCO-2 averaging kernel is on 20 levels.
(right) Comparison of each of the quantities from (a) to AJAX (black), where AJAX with the corresponding averaging kernel applied (using Eq. 1) are dotted lines, and priors are green dashed/dotted lines. Note that AJAX data with OCO-2 LMT (orange, dotted lines) and AJAX data with GOSAT LT (red, dotted lines) are nearly the same value, overlaid. The n values are the corresponding number of satellite observations that are with the coincidence criteria, and averaged together, where the error bars shown correspond to the standard deviation of the satellite values

Flight Objective	Description	# of flights
Tropospheric profiling:		
On/Off	At least one vertical profile over land and at least one offshore (>5 km)	40
Profile	Vertical profile from ~ 8 km to surface	127
RRV	Vertical profile over Railroad Valley, NV (elev. ~1.4 km), usually coordinated with GOSAT overpass	58
TCCON	Vertical profile over TCCON instrument at NASA's Armstrong Flight Research Center (formerly Dryden)	8
THD	Vertical profile near Trinidad Head observatory, usually coordinated with NOAA ozonesonde	11
VPocean	Vertical profile over ocean, with top altitude > ~5 km	63
Boundary layer observations:		

CABOTS	In support of CARB's California Baseline Ozone Transport Study (Summer 2016)	12
CentralValley	Boundary layer measurements in California's Central Valley	85
DAQ	Focused on the boundary layer over Central California during DISCOVER-AQ field campaign (Jan-Feb 2013)	6
Offshore	Sampling off the California coast, does not include a vertical profile	14
SanBernardinoWestMojave	Boundary layer measurements in the San Bernardino Mountains and/or west Mojave Desert regions.	15
SFBayArea	Boundary layer measurements In the San Francisco Bay Area	13
Source identification:		
Fire	Sampling influenced by one or more fires, usually wildfires	17
O&G	Sampling over known oil and/or gas infrastructure	17
Urban Outflow	Boundary layer measurements around Sacramento, CA	12

Table 1: Definition and description of common AJAX flight objectives and count of flights that contain each objective. Note that many AJAX flights accomplished multiple objectives in a single flight.

Error source (10s sampling resolution)	Ozone (ppb)
Laboratory precision	1.2
Variability in zero offset	1.4
Repeatability	0.6
Pressure dependance	2.2
Overall uncertainty (RMS of 1-σ uncertainties)	3

Table 2. Uncertainty of the AJAX O_3 measurements.

Error source (3s sampling resolution)	CO_2 (ppm)	CH_4 (ppb)
Laboratory precision	0.03	0.3
Accuracy of the standard	0.07	0.3
Variance in flight	0.08	0.8
Uncertainty due to water vapor corrections	0.10	2.0
Repeatability	0.03	0.4
Pressure dependance	0.04	0.1
Overall uncertainty (RMS of 1-σ uncertainties)	0.16	2.2

Table 3. Uncertainty of the AJAX CO_2 and CH_4 measurements.

585