RESEARCH ARTICLE

Investigating large methane enhancements in the U.S. San Juan Basin

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In 2014, a satellite-based map of regional anomalies of atmospheric methane (CH4) column retrievals singled out the fossil fuel rich San Juan Basin (SJB) as the biggest CH4 regional anomaly (“hot spot”) in the United States. Over a 3-week period in April 2015, we conducted ground and airborne atmospheric measurements to investigate daily wind regimes and CH4 emissions in this region of SW Colorado and NW New Mexico. The SJB, similar to other topographical basins with local sources, experienced elevated surface air pollution under low wind and surface temperature inversion at night and early morning. Survey drives in the basin identified multiple CH4 and ethane (C2H6) sources with distinct C2H6-to-CH4 emission plume ratios for coal bed methane (CBM), natural gas, oil, and coal production operations. Air samples influenced by gas seepage from the Fruitland coal formation outcrop in La Plata County, CO, had enhanced CH4, with no C2-5 light alkane enhancements. In situ fast-response data from seven basin survey flights, all with westerly winds, were used to map and attribute the detected C2H6 and CH4 emission plumes. C2H6-to-CH4 plume enhancement correlation slopes increased from north to south, reflecting the composition of the natural gas and/or CBM extracted in different parts of the basin. Nearly 75% of the total detected CH4 and 85% of the total detected C2H6 hot spot were located in New Mexico. Emissions from CBM and natural gas operations contributed 66% to 75% of the CH4 hot spot. Emissions from oil operations in New Mexico contributed 5% to 6% of the CH4 hot spot and 8% to 14% of the C2H6 hot spot. Seepage from the Fruitland coal outcrop in Colorado contributed at most 8% of the total detected CH4, while gas venting from the San Juan underground coal mine contributed <2%.

Keywords: Emissions, Methane, Ethane, Oil and gas, Hot spot

1. Introduction
Atmospheric methane (CH4) is a potent long-lived greenhouse gas (GHG). It is emitted by both anthropogenic and natural sources. CH4 global sum of sources estimated using atmospheric observations as constraints was found to be 500 Tg CH4/yr to 600 Tg CH4/yr (Dlugokencky et al., 2009, 2011; Saunois et al., 2016; He et al., 2020).

CH4 is the primary constituent of natural gas. It is often present in coal seams and can be coproduced with crude oil or liquid condensate. Globally, coal, oil, and natural gas extraction and the natural gas supply chain contribute about 25% of CH4 annual total emissions, or 145 ± 23 (1σ) Tg/yr (Schwietzke et al., 2016). Anaerobic microbes decompose organic matter in landfills, wastewater, livestock manure, rice paddies, and wetlands and produce biogas, which is a mix of CH4 and CO2. Wetlands are the largest natural source of CH4, contributing about one third of total CH4 emissions globally. Enteric CH4 is generated by methanogens in the digestive tract of ruminants. This source alone is estimated at about 100 Tg/yr.

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Emissions from all microbial sources combined are estimated at 325 Tg/yr to 355 Tg/yr (Dlugokencky et al., 2011; Schwietzke et al., 2016; Tian et al., 2016). Incomplete combustion from biomass burning also results in CH₄ emissions, estimated at 43 ± 9 Tg/yr (Schwietzke et al., 2016).

The main atmospheric sink of CH₄ is oxidation by hydroxyl radicals, OH, leading to carbon dioxide, CO₂. The CH₄ global mean atmospheric lifetime is ~9 years. With an atmospheric lifetime much larger than the troposphere interhemispheric ~1 year mixing time, CH₄ is well mixed zonally in the background troposphere and is considered a long-lived trace gas.

Analyses of old air trapped in ice cores show that over the past 650,000 years, atmospheric CH₄ has oscillated between 350 part per billion (ppb) and 850 ppb (Etheridge et al., 2002; Loulergue et al., 2008). Due to increased emissions from agriculture and fossil fuel extraction, atmospheric CH₄ has more than doubled since preindustrial times (Etheridge et al., 2002; Loulergue et al., 2008).

The National Oceanic and Atmospheric Administration Global Greenhouse Gas Reference Network (NOAA GGGRN) has been conducting calibrated high accuracy and high precision measurements of long-lived GHG dry air mole fractions at globally distributed remote locations for over 40 years.

CH₄ measurements from air sampling at a network of locations at different latitudes in the marine boundary layer (MBL) are representative of large-scale MBL gradients. Studies of the atmospheric CH₄ budget, regionally or globally, require robust detection of small changes in CH₄ at the 1 ppb level (Dlugokencky et al., 1994).

The globally averaged marine surface air CH₄ annual mean derived from the NOAA GGGRN observations was 1,853.3 ± 0.9 ppb in 2018 (Dlugokencky et al., 2019). Since 1984, CH₄’s interhemispheric difference has averaged 88 ppb, reflecting larger emissions in the Northern Hemisphere than in the Southern Hemisphere.

NOAA GGGRN measurements show that “background” CH₄ plateaued globally in the early 2000s and started rising again in 2007, reflecting an imbalance between CH₄ global sources and sinks. The year-to-year increase in global mean CH₄ has ranged between 4.6 ppb and 12.7 ppb from 2007 to 2019 (Dlugokencky, 2019). Assuming a constant lifetime, CH₄ emissions have increased stepwise since prior to 2007, reaching a ~40 Tg CH₄/yr increase in 2018.

Calibrated globally distributed measurements of long-lived GHGs and stratospheric ozone-depleting substances are used to track their contributions to increased radiative forcing since 1750. CH₄ is the second largest contributor to present-day total radiative forcing from long-lived GHG, after CO₂: 20% versus 66%, respectively (Butler and Montzka, 2018).

Modeling studies show that climate impacts, such as ocean warming and sea-level rise, from increased radiative forcing due to increased atmospheric burdens of long-lived GHGs will last from centuries to millennia (Zickfeld et al., 2017; Horton et al., 2018). Limiting future impacts of global climate change necessitates dramatic cuts in long-lived GHG emissions without delay (Montzka et al., 2011; IPCC, 2018; USGCRP, 2018; Nisbet et al., 2020).

Annual accounting of GHG sources and sinks by nations has long been required and encouraged by the United Nations Framework on Climate Change. This sector-based accounting often relies on bottom-up (BU) methods, or inventory calculation, which typically use reported activity counts and measured or modeled unit-level mean emission factors. Each year since the early 1990s, the U.S. Environmental Protection Agency (EPA) compiles and reports a national GHG inventory (GHGI) of annual emissions from 1990 to 2 years prior to the inventory report release year (EPA GHGI, 2018).

Atmospheric measurements of trace gases of interest can also be used in combination with measured or modeled atmospheric dispersion/transport to optimize or independently quantify emissions. These atmosphere-based quantification methods are classified as “top-down” (TD).

A few recent atmospheric composition studies have argued that the U.S. shale gas and tight oil boom may be partly responsible for increases in CH₄ observed at many sites around the globe and increases in ethane (C₂H₆) observed at several sites in the Northern Hemisphere after 2006 (Franco et al., 2016; Hausmann et al., 2016; Helmig et al., 2016; Turner et al., 2016). According to the U.S. EPA GHGI, the magnitude of O&G CH₄ emissions has not changed much over the past 10 years. Two recent analyses of NOAA GGGRN aircraft network data show a modest increase in CH₄ vertical gradient (boundary layer—free troposphere) at three sites heavily influenced by O&G operations but no detectable increase at sites in the air outflow on the U.S. East Coast (Bruhwiler et al., 2017; Lan et al., 2019).

However, over the past decade, TD studies at multiple spatial scales have found larger anthropogenic CH₄ emissions than predicted by the U.S. EPA GHGI (Miller et al., 2013; Brandt et al., 2014). Field measurement studies in U.S. O&G basins have found anomalously large emission contributions from a small fraction of facilities or unaccounted for emissions from certain operations or pieces of equipment (Allen et al., 2015a, 2015b; Lyon et al., 2015, 2016; Marchese et al., 2015; Relia et al., 2015; Subramanian et al., 2015; Brandt et al., 2016; Frankenberg et al., 2016; Bell et al., 2017).

New activity and emission data (some from studies referenced above) have led the U.S. EPA to modify the GHGI calculations for petroleum and natural gas systems CH₄ emissions since the baseline year of 1990. The 2020 GHGI recalculation for U.S. natural gas systems CH₄ emissions between 1990 and 2017, for example, resulted in an 8% decrease on average compared to the GHGI report published in 2019 and a 16% decrease for emissions in 2017 (EPA GHGI, 2020). For the year 2018, the EPA GHGI (2020) estimates of U.S. CH₄ emissions for petroleum systems and natural gas systems were 1.45 Tg/yr and 5.60 Tg/yr, respectively.

A new synthesis of O&G facility or process-level emission results, including contributions from super-emitters, derived a larger estimate for U.S. O&G CH₄ emissions of...
13 ± 2 Tg CH₄/yr for 2015 (Alvarez et al., 2018). This is 60% larger than the EPA GHG (2018) estimate. The authors also found that 85% of U.S. O&G CH₄ emissions are from production, gathering, and processing operations, which are confined in O&G producing regions (Alvarez et al., 2018).

Both BU and TD methods have inherent strengths and weaknesses. For example, super-emitter emissions are typically not considered in official BU emission inventories. Another example is that TD regional aircraft studies typically sample midday emissions. In a basin with episodic daytime emissions, it was demonstrated that aircraft-based midafternoon estimates were not representative of daily mean emissions (Schwietzke et al., 2017; Vaughn et al., 2018), but the temporal representativeness question has not been evaluated for other basins.

Another complication for TD large spatial scale CH₄ studies has to do with the quantitative attribution of estimated area emissions among various sources present in the study domain. Some regional studies have used BU estimates for non-O&G emissions (Pe´tron et al., 2014; Karion et al., 2013). The development of reliable measurement-based methods for the attribution of CH₄ emissions at various spatial scales is critical for the advancement of TD GHG emission budgeting and trend analysis.

C₂-₅ light alkanes, also referred to as natural gas liquids, are naturally present, though in varying amounts, in thermogenic natural gas. Associated natural gas coproduced with liquid condensate and light crude oil will have higher ethane to methane (E/M) ratios than dry thermogenic gas and biogenic gas (Sherwood et al., 2013).

Several emission and air composition studies in O&G basins have shown strong correlations between methane and C₂ light alkanes at the surface downwind of sources or higher up in the boundary layer (Katzenstein et al., 2003; Pétron et al., 2012, 2014; Yacovitch et al., 2014; Helmiag et al., 2014; Roscioli et al., 2015; Peischl et al., 2018; Kille et al., 2019; Mielke-Maday et al., 2019). Smith et al. (2016) and Mielke-Maday et al. (2019) have used enhancement ratios from survey flight in situ CH₄ and C₂H₆ measurements in the planetary boundary layer (PBL) over shale gas basins to estimate the contribution of emissions from natural gas operations to the basin total CH₄ emission estimate derived using the aircraft mass balance approach (Karion et al., 2013, 2015). This study presents a variant of the CH₄ plume attribution method based on high-frequency and high-sensitivity in situ CH₄ and C₂H₆ measurements introduced by Yacovitch et al. (2014) and Smith et al. (2015).

In 2014, a major fossil fuel producing region in the southwestern United States came into the spotlight. Kort et al. (2014) published a map of satellite-derived CH₄ column regional anomalies, which showed the U.S. “Four Corners” region (named after the intersection of four state boundaries: Utah, Colorado, New Mexico, and Arizona) was “the largest US CH₄ anomaly viewed from space.”

The regional anomaly, dubbed the U.S. Four Corners CH₄ hot spot, was more specifically located over the San Juan Basin (SJB), which is home to coal, oil, natural gas, and coal bed methane (CBM) extraction. These operations are known to release CH₄, yet as mentioned earlier, U.S. O&G CH₄ emission estimates derived with different methods often disagree. Besides fossil fuel–related sources, the region is also known for CH₄ and CO₂ seepage from the Fruitland coal formation outcroppings in La Plata County in SW Colorado. The Four Corners CH₄ hot spot announcement received national news coverage, and for the local community and its leaders, it fueled an already active debate about the magnitude and impacts of local CH₄ sources.

In April 2015, several research groups funded by NOAA, NASA, and NSF deployed ground and airborne instrumentation in the SJB to further study the Four Corners CH₄ hot spot. The goals were to use coordinated in situ measurements to (1) quantify the region’s total CH₄ emissions, (2) identify CH₄ sources and evaluate their contributions, and (3) document drivers of the region’s CH₄ atmospheric column anomaly.

Smith et al. (2017) addressed the first goal by estimating TD total CH₄ emissions for the region using campaign data from five mass-balance flights. Their results are used in this follow-up study to address research goals 2 and 3 above. In Section 2, we give an overview of the basin topography, its CH₄ sources, and previously reported emission estimates. In Section 3, we describe the ground and airborne sampling platforms, the in situ trace gas instrumentation, and the wind profilers deployed for the campaign. Section 4 details the data analysis and main results. First, we look at boundary layer height and horizontal wind measurements and investigate the diurnal pattern in surface and boundary layer airflow at a few different locations in the mountain basin. We follow with an analysis of on-road in situ and flask air trace gas measurements, which covered major areas of the SJB and sampled CH₄ emission plumes from different sources, including various fossil fuel operations and gas seepage from the Fruitland coal outcrop in La Plata County. Sampling with an instrumented van throughout the basin at different times of the day allowed us to document the accumulation of CH₄ emissions at night in a shallow surface air layer, especially on low-wind nights in areas with nearby sources and lower elevation. In Section 4, we analyze aircraft in situ CH₄ and C₂H₆ measurements from basin survey flights. The TD attribution based on C₂H₆-to-CH₄ enhancement ratios in detected plumes provides a new breakdown of how different sources in Colorado and New Mexico contribute to the observed CH₄ and C₂H₆ hot spots. In Section 5, results are discussed in the context of existing literature. Finally, Section 6 presents the main conclusions and implications of this paper.

2. Study region and background on CH₄ hot spot
2.1. Geography
The SJB is a sedimentary basin in the Colorado Plateau in the southwestern United States (Huffman and Condon, 1993). It has many rivers and is the watershed of the upper San Juan River, which merges with the Colorado River in Utah. The region is rich in minerals and fossil...
fuels, especially coal and CBM (Fassett and Hinds, 1971; Ayers, 2003; Fassett and Boyce, 2005).

The SJB stretches over 160 km north–south and 140 km east–west. The Fruitland coal formation surface outcroppings outline most of the basin’s periphery (Figure 1). Surface elevations range from ~1,550 meters above sea level (masl) near the Four Corners monument to almost 2,200 masl closer to the San Juan Mountain foothills. The basin is surrounded by several mountain ranges, with peaks reaching elevations above 4,000 masl to the north and over 3,000 masl to the east of the basin.

The SJB straddles the border between SW Colorado and NW New Mexico. The land and mineral rights fall under private/state, tribal, or federal ownership (Parikh et al., 2017a). According to the U.S. Census, the region’s population in 2015 was close to 400,000 (Census, 2017).

2.2. San Juan Basin methane sources and hot spot

2.2.1. CH4 sources and their emission estimates

The SJB has been a major producer of coal, CBM, conventional natural gas, and oil for decades (Figure S1, Supplementary Material Text Section 1; Barnes, 1951; Bie bomberman and Clarich, 1951; Murray, 1996). CBM is a type of unconventional natural gas present in an adsorbed form in underground coal deposits. CBM is typically >95% CH4 in volume, and it is extracted using mostly vertical wells and dewatering operations (Haldar, 2018). In spring 2015, the region had two active coal mines, both tapping the Fruitland coal formation and located in San Juan County, NM. There were over 16,000 gas wells, 7,000 CBM wells, and 1,700 oil wells (Parikh et al., 2017a; Table S1) producing from a few different geological formations. The extensive gas infrastructure also includes gas pipelines and centralized gathering, processing, and compression facilities. The region has thousands of inactive or legacy wells and a large number of abandoned coal mines (Nickelson et al., 1988).

In April 2015, operations in the SJB represented 45% of U.S. CBM total production, close to 2.5% of U.S. non-CBM gas total production and only 0.25% of U.S. oil total production (Tables S1 and S2; COGCC, 2018; NM OCD Statistics, 2018; U.S. EIA, 2018a, 2018b). Close to half of the gas produced in the study area was CBM (44.3 billion cubic feet). In April 2015, there were only three active drilling rigs in the basin, all in New Mexico (Baker Hughes, 2015).

We have compiled available BU and TD estimates of CH4 emissions of the SJB in Table 1. Based on data from the U.S. EPA GHGI for 2012 (Maasakkers et al., 2016), anthropogenic CH4 emissions for the SJB add up to 0.40 Tg CH4/yr (45.8 tonnes CH4/h), with gas systems in New
Mexico and Colorado contributing two thirds and one sixth, respectively, of this total. The WESTAR SJB inventory gives a similar estimate for O&G CH4 emissions in 2014 (0.37 Tg CH4/yr or 42.5 tonnes CH4/h; Table 1; Parikh et al., 2017a, 2017b, see also Section 4.3.4).

Since 2007, CH4 flux chamber measurements have been conducted in spring on a grid map over the Fruitland coal outcrop in La Plata County, CO, by consulting company LT Environmental (LTE; COGCC/LTE, 2018). The flux results show large year-to-year variability at individual measurement locations and in total detected seepage area and aggregated gas seepage magnitude.

Based on the 2016 survey total volumetric flux result (18.165 million cubic feet per day or MMcf/day) extrapolated to the entire year, LTE reports an estimated annual CH4 emission from the outcrop in La Plata County of 125.4 Gg CH4/yr or 14.5 tonnes/h. Using the same derivation and LTE 2015 survey total volumetric flux result (16.903 MMcf/day), the hourly mean CH4 source for spring 2015 is 13.5 tonnes/h (LTE, 2015, 2017). Estimates based on LTE field survey results for 2015 and 2016 are similar. However, between 2007 and 2014, LTE total volumetric flux results show large year-to-year variability with a minimum flux below 2 MMcf/day in 2010 and a maximum flux of *11 MMcf/day in 2014 (see slide 14 in LTE, 2017). Due to the highly variable nature of gas seepage, it is expected that LTE aggregate CH4 seepage rate is fairly uncertain.

Overall, the best available BU estimate for the SJB total CH4 emissions around the time of the campaign is close to
60 tonnes CH$_4$/h (Table 1). The 95% confidence interval for the EPA GHGI natural gas systems CH$_4$ emission, the largest CH$_4$ source in the SJB, ranges from $-19\%$ to $+30\%$ around the inventory national estimate. At subnational scales, the emission uncertainties will be larger than for the GHGNI national estimates (Maasakkers et al., 2016). A comparison of the gridded GHGI with a detailed hybrid (BU and TD) inventory for the Barnett Shale gives a relative error ranging from $30\%$ to $50\%$ for grid resolution going from 0.5° to 0.1° (Maasakkers et al., 2016). There is no uncertainty estimate for the Fruitland coal outcrop emission derivation from LTE.

2.2.2. Composition of emissions
Fossil fuel industry emissions in the SJB can have different chemical signatures, due to multiple forms being tapped for production (Ridgley et al., 2013), as well as varying levels of separation and processing at some facilities. Eighty-four percent of the gas produced in the northern SJB in April 2015 was CBM (Table S1), which is very dry gas with mostly CH$_4$, some CO$_2$, and no or very low non-methane hydrocarbons (NMHC, such as C$_2$H$_6$ and C$_3$H$_8$). The southern SJB has been producing substantial volumes of both CBM and conventional natural gas and increasing, yet still modest, volumes of oil (Table S1).

CBM and conventional natural gas sample composition data for the SJB are available from the Global Inventory of Gas Geochemistry Database (Sherwood et al., 2017), which includes data from Rice (1993), the U.S. Geological Survey Geochemistry database (USGS, 2014), and the Colorado Oil and Gas Conservation Commission, among many others worldwide. C$_2$H$_6$-to-CH$_4$ mole-to-mole ratios (C$_2$/C$_1$) for Fruitland formation CBM samples (with reported ethane mole percent > 0.05) range between 0.05% and 12%, with a median of 0.3% in the northern SJB and a median of 3.3% in the southern SJB. For conventional gas samples, C$_2$/C$_1$ ranges between 0.05% and 18.8% for the basin, with a median of 0.4% in the northern SJB and a median of 6.6% in the southern SJB.

Accordingly, one can expect O&G emission plumes detected in the SJB to cover a large range of C$_2$/C$_1$, reflecting a complex mix of dry and wet gas operations and sources.

2.2.3. Background on Four Corners methane hot spot
Regional or local air pollution hot spots are typically caused by large localized pollution sources combined with trapping topography (basin with nearby mountain ranges) and atmospheric transport patterns, for example, recirculation from land/sea breeze, upslope/downslope winds, or surface temperature inversion at night or in winter (Bassandorj et al., 2017; Fast et al., 2007; Helming et al., 2014; Littman et al., 1993; Oltmans et al., 2014; Reddy et al., 2016; Schnell et al., 2009; Stephens et al., 2008).

Atmospheric in situ measurements or remote sensing scans can be powerful tools to map pollution hot spots (Worden et al., 2013; Kort et al., 2014; Lawrence et al., 2015; Jacob et al., 2016) or identify and quantify larger than expected pollution sources (de Gouw et al., 2009; Frankenberg et al., 2016; Karion et al., 2013). Kort et al.’s (2014) U.S. CH$_4$ anomaly map relied on retrievals of total column average CH$_4$ dry air mixing ratio (XCH$_4$, hereafter) from near infrared radiance measurements aboard the SCIAMACHY satellite (Frankenberg et al., 2006, 2011). Globally, the largest regional scale (>100 km) retrievals were observed over the tropics and in Asia and were associated with emissions from wetlands, agriculture, and coal extraction. For the western United States, Frankenberg et al. (2011) mentioned two regions with larger XCH$_4$ column retrievals than their immediate surroundings: the San Joaquin Valley (SJV) in California and the SJB.

High terrain regions such as the Rocky Mountains, the Himalayas, and the Andes have shorter tropospheric columns that result in smaller retrieved XCH$_4$ because CH$_4$ mixing ratios are lower in the stratosphere than in the troposphere (see figure 17 and text in Frankenberg et al., 2011). For future reference (Section 4), we also note that the SCIAMACHY/ENVISAT satellite overpass time for the SJB was midmorning, centered on 10:40 a.m. Local Standard Time (LST, or 11:40 a.m. local daylight saving time).

Kort et al.’s (2014) gridded map of regional 2009 to 2013 average CH$_4$ anomalies used “topography corrected” SCIAMACHY CH$_4$ column retrievals over the United States. Based on these maps at 1/3 degree horizontal resolution, the authors found 10 to 12 grid tiles over the SJB in Colorado and New Mexico with 25 ppb to 40 ppb enhancements over the local background XCH$_4$ column average. It is important to note here that the local XCH$_4$ background can vary by 20 ppb or more across different regions of the United States. The SJB local XCH$_4$ background is among the lowest in the country as the region is surrounded by mountains and less developed and industrialized areas compared to the U.S. Midwest and Eastern States.

Kort et al. (2014) concluded that the SJB was a “persistent anomalous source region,” and the region became known as the “largest U.S. CH$_4$ anomaly viewed from space.” At the very least, it might be described as “the largest local XCH$_4$ anomaly” derived from midmorning 2003 to 2009 SCIAMACHY retrievals. Kort et al. (2014) also estimated that the SJB emitted 0.59 TgCH$_4$/yr (0.50 to 0.67; 2σ).

In April 2015, a follow-up scientific measurement campaign took place in the U.S. Four Corners region. The main goals were to quantify and attribute CH$_4$ emissions using a suite of ground and airborne in situ trace gas and meteorological measurements. Smith et al. (2017) used in situ CH$_4$ and wind data from five flights to derive aircraft mass balance estimates of basin-wide CH$_4$ emissions. The annualized estimates they derived ranged from 0.31 ± 0.13 TgCH$_4$/yr to 0.84 ± 0.30 TgCH$_4$/yr. The authors averaged results from the five flights and reported an annual mean SJB TD flux of 0.54 ± 0.20 TgCH$_4$/yr. This estimate is close to Kort et al.’s (2014) mean estimate for 2003 to 2009. We note that the April 21, 2015, flight, which produced Smith et al.’s lowest mass balance CH$_4$ flux, only covered a portion of the SJB (see flight map in Figure 1; Smith et al., 2017). The low emission estimate for that flight is very likely an underestimation of the full basin emissions.
The aircraft mass balance quantification technique only provides a snapshot estimate of total midday emission for a region (Schwietzke et al., 2017). The technique does not separate and constrain contributions from individual point sources or from various source categories.

In previous O&G CH₄ TD studies, the attribution of emission estimates relied on BU/inventory information (Pétron et al., 2014; Peischl et al., 2015), on the use of additional tracers as markers for different CH₄ source categories (Smith et al., 2016; Mielke-Maday et al., 2019), or on a mix of facility-level measurements and detailed inventory activity data (Vaughn et al., 2018).

During the April 2015 campaign, some fieldwork focused on the airborne detection of individual facility CH₄ plumes with in situ measurements (coal mine and outcrop, mentioned in Smith et al., 2017) or remote sensing (Frankenberg et al., 2016; Thorpe et al., 2017). Two contracted Twin Otters equipped with NASA CH₄ partial atmospheric column remote sensing instruments AVIRIS-NG and HyTES conducted five flights over portions of the SJB. Frankenberg et al. (2016) estimated point source emissions for over 200 detected CH₄ plumes. Some of these point sources were further characterized closely in time after the detection with ground-level infrared plume imaging (Thorpe et al., 2017).

The NASA 2015 survey data set independently confirmed that there is a multitude of CH₄ emitting sources dispersed all over the SJB, including a few anomalously large sources. It also revealed that a small number of point sources contributed a large fraction of the detected CH₄ column total enhancement (Frankenberg et al., 2016).

Here, we pursue the analysis of the April 2015 ground and airborne in situ measurements to independently constrain the mix of sources and meteorological conditions contributing to the Four Corners CH₄ hot spot over the basin. In Section 3, we describe the trace gas and meteorological measurements used in the analysis.

3. Methods and data sets

3.1. Ground-based and aircraft in situ measurements

On the ground, two instrumented vehicles surveyed portions of the SJB from public access roads or with an escort on Southern Ute Indian Tribe (SUIT) and Navajo Nation roads. The NOAA Mobile Laboratory was equipped with three species (CH₄, CO₂, H₂O) or four species (CH₄, CO₂, CO, H₂O) cavity ring down spectrometer (CRDS, Picarro G2301 or G2401) depending on the day (due to instrumental problems) and with the NOAA Global Monitoring Laboratory (GML) programmable flask sampling apparatus (see below). The CU/INSTAAR van was equipped with a CH₄ and δ¹³CH₄ CRDS (Picarro G2132-i) and a 2D anemometer. The ground survey team members each wore a personal trace gas sensor (borrowed from the Farmington BLM office). In a few instances in the southern SJB, they did smell hydrogen sulfide (H₂S), a poisonous gas, downwind of compressor stations or well pads.

Up to five instrumented aircraft flew different patterns mostly midday (11 a.m. to 5 p.m.) and in the boundary layer over the SJB. The NOAA Twin Otter, NOAA P-3, and Scientific Aviation (SA) Mooney were each equipped with a CH₄, CO₂, H₂O CRDS (Picarro G2301), and in situ C₂H₆ spectrometer (Aerodyne; Yacovitch et al., 2014). These high-frequency, high-precision in situ C₂H₆ analyzers have been used to sample single facility or area distributed emission plumes. C₂H₆ and CH₄ plume enhancement ratios have been used to distinguish between biogenic and thermogenic sources in regions with a complex mix of CH₄ sources (Wennberg et al., 2012; Yacovitch et al., 2014, 2015; Hopkins et al., 2016; Smith et al., 2016).

The NOAA Twin Otter and Mooney also collected discrete air samples analyzed by NOAA GML. Horizontal wind speed and direction along the flight track were derived from differential GPS measurements for the Mooney (Conley et al., 2014, 2017) or derived from differential pressure measurements aboard the NOAA P-3 (Hübler et al., 1998). The CRDS analyzers in the ground vehicles and on the Mooney airplane were calibrated at the NOAA GML laboratory soon before and/or after the campaign using a suite of six gas standards spanning below and above ambient levels (1.776 ppb to 3.049 ppb for CH₄). The total uncertainty of the CRDS CH₄ measurements in the laboratory is 0.5 ppb over the calibration range.

Survey flights in a convective (well-mixed) PBL (not too deep) and under low to moderate dispersion (horizontal wind speed < 10 m/s) conditions are ideal to map and attribute emission plumes (Smith et al., 2017). Afternoon flights in the convective PBL and with steady uniform horizontal winds (> ~ 3 m/s) are necessary to capture horizontal gradients of CH₄ and C₂H₆ upwind and downwind of the study region for mass balance area flux calculation (Karion et al., 2013, 2015; Schwietzke et al., 2017).

The NOAA Twin Otter conducted regional mass-balance flights (Smith et al., 2017). The Mooney flew around target facilities (Frankenberg et al., 2016; Conley et al., 2017) and conducted survey flights over portions or the entirety of the SJB. The NOAA P-3 conducted two basin-wide survey flights as part of the SONGNEX aircraft campaign (Peischl et al., 2018). As mentioned earlier, two contracted Twin Otters equipped with NASA CH₄ partial atmospheric column remote sensing instruments AVIRIS-NG and HyTES conducted five flights over portions of the SJB (Frankenberg et al., 2016).

In this study, we use in situ and flask air measurements from the SA Mooney and NOAA P-3 aircraft and the two ground vehicles described above. For all platforms, scientists could visualize in situ measurements on a screen in close to real time. All sampling platforms used one or multiple GPS units to track location and time at 1 Hz. Data recorded by each instrument were merged in real time or with post-processing using the GPS location information. CH₄ in situ measurements are reported in units of dry air mole fraction, which have a total uncertainty <2 ppb at near ambient levels. All measurements are reported using the WMO X2004A CH₄ calibration scale (Dlugokencky et al., 2005; GML CH₄ Scale, 2015). Comparison plots of the in situ and flask CH₄ results for a subset of the flasks collected from the Mooney aircraft are in the supplementary material (Figure S18).
3.2. Discrete air samples

The SA Mooney aircraft and the vans were equipped with NOAA GML programmable flask packages (PFP) and programmable compressor packages. Discrete 20- to 30-s integrated air samples were collected in the PFP borosilicate flasks mostly in the boundary layer over the SJB.

More than 50 trace gases were measured in the NOAA GML laboratories and reported in units of dry air mole fraction. We use the following abbreviations: ppm = \( \mu \text{mol mol}^{-1} \), ppb = nmol mol\(^{-1} \), and ppt = pmol mol\(^{-1} \). The NOAA GML GHG flask air analysis systems use gas-specific analyzers and custom sample inlets and data acquisition and control systems. Results are traceable to internal calibration scales derived using gravimetric methods (Hall et al., 2019). Here, we focus on the flask air analysis for CH\(_4\) and NMHC.

Flask air CH\(_4\) is measured using gas chromatography with flame ionization detection (Dlugokencky et al., 1994). For CH\(_4\), each aliquot of air from a flask is bracketed by an aliquot of standard. The flask air measurement uncertainty is <2 ppb for CH\(_4\) over the nominal standard scale range of 300 ppb to 5,000 ppb (GML CH\(_4\) Scale, 2015). To prevent contamination of the NOAA GML analytical systems and stay within our calibration range, the NOAA van targeted air sample collection in plumes when ambient CH\(_4\) measured with the CRDS was below 5,000 ppb. In situ CH\(_4\) measurements above this threshold were found in plumes downwind of emitting facilities or in small valleys impacted by gas seepage from the Fruitland coal outcrop.

The NOAA GML PFP analysis includes measurements of C\(_2\)-C\(_6\) alkanes and benzene (C\(_6\)H\(_6\)) by gas chromatography–mass spectrometry (GC-MS; system name: Perseus). These species are co-emitted by some O&G CH\(_4\) sources. For the GC-MS, the standard protocol is to analyze air from a reference gas cylinder every five injections. For polluted samples, each flask air injection was bracketed with reference gas injections. This allowed us to closely track any changes in the instrument response when a polluted air sample was analyzed. Additionally, for flask air with CH\(_4\) >2,200 ppb, 20% to 50% of the usual injection volume (480 mL STP) was injected to the GC-MS to reduce the impact of high mixing ratio samples on the instrument detector.

The Perseus light alkanes and benzene measurements are reported on the following calibration scales: ethane (NOAA-2015-PR1), propane (NOAA-2012-PR1), n-butane (NOAA-2012-PR1), i-butane (NOAA-2012-PR1), n-pentane (NOAA-2012-PR1), i-pentane (NOAA-2012-PR1), and benzene (NOAA-2012-PR1). Relative uncertainties associated with NMHC reported mole fractions are typically less than 5% (1 SD).

3.3. Horizontal wind and boundary layer height measurements

To characterize regional atmospheric transport patterns during the campaign, we deployed three wind profilers at fixed locations in the basin (Figure 1). Each system tracked horizontal wind speed and direction. Sunrise and sunset occurred at 6:47 a.m. and 7:41 p.m. on April 9, 2015, and 6:20 a.m. and 8:00 p.m. on April 30, 2015.

The Farmington site (FMT: 36.79N; 108.16W; 1781 masl) located on a flat portion of NW New Mexico is representative of the wind conditions on the western edge of the densely drilled south portion of the SJB. The Navajo State Park site (NLS: 36.81 N; 107.65 W; 1974 masl) is 28 miles (45.5 km) due East from FMT and in the heart of the SJB O&G operations. The southern Colorado site, near Ignacio, CO, is one of the SUIT air quality monitoring stations, UTE1 (37.14N; 107.63W; 1993 masl). It sits in the mountain foothills and is in the middle of the northern SJB natural gas and CBM operations. The San Juan Mountains peak at over 4,000 masl, ~35 miles north of UTE1. Mountain ranges extend north to south ~60 miles east of UTE1 and have summits above 3,500 masl.

Two NOAA 915 MHz Doppler radar wind profilers (White et al., 2015) were deployed at FMT and NLS. We use processed data from the profilers’ long-range mode. The vertical resolution was 100 m, and the first reported level is at 145 m above ground level (magl). Depending on the day and time, most retrieved profiles extend to at least 2,000 magl, some reaching up to 6,000 magl. The profilers operated continuously, doing a scan for 5 min every hour, between April 8 and May 19, 2015 (no data gaps). Here, we focus on the data contemporaneous with the other measurements in April 2015.

A smaller scanning wind Doppler Lidar, WindCube 100S rented from Leosphere, was deployed at UTE1. The Lidar operated continuously from April 11 to April 30, 2015, with a profile scan every 10 min. Retrieved horizontal wind speed and direction are reported for 99 vertical levels distributed between 89 magl and 4,330 magl, but most retrieved profiles do not extend beyond 2,000 magl, and there were multiple data gaps. Vanderwende et al. (2015) describe the data processing for this type of Lidar.

4. Results

In this section, we first investigate the basin wind diurnal patterns near the surface and throughout the atmosphere’s first 2 km. We then present results from the ground mobile multiple species measurements. Finally, we analyze basin survey flight in situ CH\(_4\) and C\(_2\)H\(_6\) measurements to attribute detected enhancements to different sources.

4.1. Horizontal wind and planetary boundary layer height (PBLH) data

Surface meteorological measurements were collected by several agencies in the SJB, and finalized data are available on the EPA AQS Data Mart web portal. Here, we analyze wind data from seven surface stations (Bloomfield, Mesa Verde, Navajo Lake, San Juan Substation, Shamrock, Ute 1, and Ute 3) and from the three profilers (UTE1, UTE3, and FMT) to investigate airflow patterns in the basin (Figure 1 and Table 2).

Figure 2 shows the medians for hourly horizontal wind speed and wind direction for several altitudes up to 2,000 magl from data collected between April 9 (FMT, NLS) or April 11 (UTE1) and April 30, 2015 (all three sites). April 2015 mean wind data from colocated surface measurement systems are also shown. Note that the surface wind
diurnal patterns observed in April 2015 at the surface stations were similar for other times of the year (Figures S2 and S3).

The wind data at all monitoring sites in the area show that median horizontal winds near the surface in the SJB had a strong diurnal cycle in both direction and speed. In April 2015, surface and near surface wind speeds were minimal at night (<2 m/s to 4 m/s depending on the site) and until 11 a.m. or noon LST. At nighttime, the profiler and surface wind data had a strong vertical gradient in the wind direction from the surface to 1 km (20:00 to 9:00 LST) with NE (UTE 1) or E (FMT and NLS) surface winds sloping down from the San Juan Mountains.

Above all three sites, near surface winds switched direction between 8:00 and 11:00 LST. Profiler data for all levels below 500 magl show consistent mean winds from the W or SW for afternoon and evening hours (until 20:00 or 21:00 LST). The UTE 1 and aircraft wind data indicate some divergence in the afternoon westerly flow in the boundary layer above the SJB eastern mountain foothills (aircraft wind data are not shown here but are included in data files).

Above 500 magl, the wind profiler data show uniform winds throughout the day and night, from the SSW (200 to 210°) above UTE 1 or from the SW to W above FMT and NLS (Figure 2). The horizontal wind speed between 500 magl and 2 km agl has a smaller diurnal cycle, with a minimum between 3 m/s and 4 m/s midmorning, when the daytime boundary layer is developing. On most clear-sky days, by 12:00 to 14:00 LST, the mean PBLH was between 9 a.m. and 5 p.m. LST.

The profiler retrievals show that the PBL growth pattern and peak height varied greatly between days at FMT and NLS (Figure S4). The earliest valid PBLH retrievals at 8:00 LST for the FMT profiler have readings below 450 magl. At 11 a.m. LST, the mean PBLH was 490 ± 250 magl at FMT (22 estimates between April 9 and May 1, 2015) and 470 ± 225 magl at NLS (16 estimates). The PBLH daily maximum ranged between 1,000 and 4,000 magl above both locations. PBLH could not be estimated during very cloudy conditions or when turbulent mixing was not strong enough, which explains the shorter PBLH time series on several days.

For the 6 days with PBLH profiler data extending in the evening at NLS and/or FMT, we see that the boundary layer started collapsing between 5 p.m. and 6 p.m. LST. In the late afternoon, rapidly declining sun radiation leads to the sudden halt of convective atmospheric mixing above the Earth’s surface and the resulting collapse of the atmosphere daytime mixed PBL. At night, the atmosphere surface layer depth is a couple hundred meters deep at most. Vertical mixing in this nocturnal surface layer (typically induced by horizontal wind friction) is very limited in the SJB due to very low surface wind speeds (Figures 2 and Figure S2), and therefore at night, surface CH\textsubscript{4} emission plumes are “trapped” and can lead to large surface enhancements near sources, as shown in the next section.

### 4.2. Ground in-situ and discrete sample measurements

We conducted ground mobile measurement on 14 different days between April 11 and April 29, 2015, mostly from public roads. Prior consent was obtained to conduct air measurements from roads on Native American land. Several drives or portions of drives were also conducted with an official escort on Navajo Nation land and SUIT land.

The van’s fast response CO\textsubscript{2}, CH\textsubscript{4}, and CO CRDS measurements were used to map CH\textsubscript{4} levels at the surface in different parts of the basin and to locate and identify CH\textsubscript{4} point sources and direct discrete air sample collection in emission plumes for additional chemical composition data.

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**Table 2.** Surface air quality and meteorological variable monitoring stations in the San Juan Basin region. DOI: https://doi.org/10.1525/elementa.038.t2

| Site Name           | AQS Code | Site Code on Figure 1 | Latitude | Longitude | Elevation (masl) | Data Provider        |
|---------------------|----------|-----------------------|----------|-----------|------------------|----------------------|
| Mesa Verde          | 08-083-9000 | MSV                  | 37.1984  | −108.4907 | 2,172            | National Park Service |
| Ute 1               | 08-067-7001 | UTE 1                | 37.1368  | −107.6286 | 1,983            | SUIT Environmental   |
| Ute 3               | 08-067-7003 | UTE 3                | 37.1026  | −107.8702 | 1,920            | Programs Division    |
| Shamrock            | 08-067-9000 | SHM                  | 37.3038  | −107.4842 | 2,367            | National Forest Service |
| San Juan Substation | 35-045-1005 | FMT                  | 36.7967  | −108.4725 | 1,678            | New Mexico Environment |
| Bloomfield          | 35-045-0009 | BFD                  | 36.7422  | −107.9769 | 1,713            | Department           |
| Navajo Lake         | 35-045-0018 | NLS                  | 36.8097  | −107.6516 | 1,950            |                      |

Data are available through EPA Data Mart (https://aqs.epa.gov/aqsweb/documents/data_mart_welcome.html).
Speciated trace gas emission inventories rely on industry or EPA data to derive a mean emission composition signature for different sources in a sector or basin. As we were planning the fieldwork, we did not have access to recent spatially relevant emission composition data. Therefore, we decided to collect air samples in CH$_4$ emission plumes intercepted with the van for a subset of sources.

Below, we describe examples of surface hydrocarbon enhancements observed along those drives. Later, the surface air sample results support the interpretation of the aircraft in situ and discrete air measurements, which typically reflect emissions from a mix of point and area sources in a larger spatial footprint than the van's samples.

4.2.1 Methane sources and their emission chemical signatures

Significant enhancements of surface CH$_4$ detected by the NOAA van instrument downwind of a facility are
interacting as emission plumes from the upwind facility. Typically, the emission plumes had peak CH₄ enhancements hundreds to thousands of ppb above the local background (1.8 ppm to 2.0 ppm CH₄). To get an emission source chemical signature, three or more flask samples were collected with the van in local background air and downwind in the facility emission plume(s) to capture different enhancement levels in a 15-min to several-hour-long sampling time window. A constant local background and emission composition is assumed for an entire sampling window downwind of a particular facility. The NOAA van collected air in flasks while stationary with the engine turned off and away or upwind from local road traffic.

Table 3 lists the location, date, and time, as well as the C₂H₆ to CH₄ and C₃H₈ to C₂H₆ mixing ratio correlation results for sampled plumes at 20 locations in the SJB and for background samples in the nearby Paradox oil producing basin in SE Utah.

Gas seepage plumes were sampled at five locations along the Fruitland coal outcrop in La Plata County where CH₄ emissions have been observed and reported (LTE, 2015). In remote outcrop sampling locations (April 12 and 14 outcrop locations #4 and 5 in Table 3), C₂H₆ in samples with elevated CH₄ (2.5 ppm to 5.2 ppm) was less than 1 ppb above the local background and was uncorrelated with CH₄ or other light alkanes (Figures S5 to S8). As a result and for the rest of this paper, we assume that SJB outcrop gas seepage has no C₂H₆.

Other sampling locations were either downwind of known facilities and potentially large point sources of CH₄ (San Juan coal mine air shaft, compressor stations, and processing plants) or happened in emission plumes encountered during a survey drive (e.g., downwind of active well pads).

On eight drives in April 2015, we collected three to seven discrete air samples in CH₄ plumes downwind of eight active well pads, three compressor stations, and two processing plants. These O&G facility plumes have highly correlated C₂H₆ and CH₄ with slopes spanning a wide range from very dry gas (<1% in CBM producing areas to wet gas (~17%) in the oil producing region near Counselor (Table 3).

Air samples were collected in Durango and its vicinity in La Plata County in areas with known outcrop gas seepage on April 11 to 14, 2015 (Figures S5 to S7). The sample analysis results show no clear correlation between CH₄ and C₂H₆ (40 samples, Figure S8A). These samples, however, show strong correlations between the C₂₅ alkane levels with similar slopes as the chemical signatures for air samples collected near O&G operations in the SJB: C₂H₆ to C₂H₄ slope of 0.38% (Figure S8B) and iC₅H₁₂ to nC₅H₁₂ slope of 1.1 ppb/ppb. This suggests that the background air in Durango and the nearby surroundings is likely impacted by hydrocarbon emissions from O&G operations in the SJB.

On April 19, 2015, the NOAA van CRDS detected up to 16 ppm CH₄ ~ ¼ mile east and downwind of the San Juan coal mine air shaft. C₂H₆ and CH₄ were strongly correlated in seven discrete air samples with a slope of 0.9% (R² of 0.98, Figure 3; Figures S9 and S10). On April 21, 2015, the NOAA van sampled surface air from dirt roads near the Navajo surface coalmine operations, including Indian Service Road 5082 (Figures S11 and S12). In situ CH₄ measurements were close to background levels (1,880 ppb to 1,910 ppb) the entire time. Four flasks collected during that time had CH₄ and C₂H₆ ranging between 1,880 ppb to 1,906 ppb and 1.7 ppb to 2.7 ppb, respectively, which are very close to background levels. These findings are supported by data from targeted aircraft flights over both active coalmines.

The WESTAR O&G emission inventory project gathered operational data from O&G companies active in the basin. Based on the SJB survey data for 2014, 94% of active CBM and gas wells in the basin had a wellhead gas compressor engine (90% rich burn/10% lean burn) with some form of emission controls, and 99% of oil wells had a gas-fueled rich burn artificial lift engine (Parikh et al., 2017a, 2017b). In the SJB, natural gas engines are the largest O&G source of NOx (Parikh et al., 2017a, 2017b).

On a few road surveys in April 2015, the NOAA van was close enough downwind of an O&G facility to isolate CH₄ emission plumes with elevated combustion markers CO₂ and CO. These plumes may have been the result of a mix of on-site gas venting, leaks, and incomplete combustion emissions from an on-site natural gas-fired engine. Using 10-s moving averages of the CRDS 0.5 Hz CH₄, CO₂, and CO measurements, we report facility plume emission ratios for five centralized compressor stations, one gas plant complex, and 11 well pads (Figure 3, Table 6). CH₄-to-CO₂ (CO-to-CO₂) correlation slopes for intercepted plumes with R² > 0.5 ranged from 0.09 ppm to 2.4 ppm CH₄/ppm CO₂ (2.3 to 93 ppb CO/ppm CO₂) for emission plumes from large NG facilities and from 0.023 ppm to 11.6 ppm CH₄/ppm CO₂ (non-detectable to 1,467 ppb CO/ppm CO₂) for emission plumes from well pads. Some of these intercepted plumes were clearly attributable to compressor engine emissions (CS#1–5 in Table 6) or to natural gas-driven compressor or pumpjack on a wellpad (WP#1,2,4,7,9 in Table 6), given the facility equipment layout and the predominant winds at the time of the sampling.

The NOAA Mobile Laboratory brief fenceline sampling of emission plumes from sites with natural gas combustion engine covered a small fraction of the SJB O&G engine population and emissions. At each site, the sampling occurred for short periods as back then O&G engine emissions had not been identified as a study target. Also, the mobile surface air sampling, despite being random in term of roads traveled, more easily picked up anomalously large enhancements such as the ones reported here.

Despite these limitations, we compare our observed emission plume ratios with the WESTAR inventory emission factors. Figure 3 shows that, especially for CH₄/CO₂, the observed ratios are several orders of magnitudes above the WESTAR inventory emission factors for point/large engines (2.10⁻⁴ ppm CH₄/ppm CO₂ and 2 ppb CO/ppm CO₂) and nonpoint/small engines at well pads (5.10⁻⁵ ppm CH₄/ppm CO₂ and 27 ppb CO/ppm CO₂).
## Table 3. Results from facility emission plume sampling with National Oceanic and Atmospheric Administration GML flasks. DOI: https://doi.org/10.1525/elementa.038.t3

| Samples Date | Time Window UTC | Source Type or Name | Latitude | Longitude | Number of Flasks | C$_2$H$_6$-to-CH$_4$ Slope | R$^2$ | C$_3$H$_8$-to-C$_2$H$_6$ Slope | R$^2$ | Min and Max CH$_4$ (ppb) | Min and Max C$_2$H$_6$ (ppb) |
|--------------|-----------------|---------------------|----------|-----------|------------------|--------------------------|------|--------------------------|------|--------------------------|--------------------------|
| April 11, 2015 | 21:34 to 2:44 | Fruitland outcrop #1 | 37.291   | -107.798  | 3                | None                    |   -- | 0.33                     | 0.97 | 1,947 to 2,397           | 3.8 to 4.0                |
| April 11, 2015 | 21:50 to 22:06 | Fruitland outcrop #2 | 37.290   | -107.797  | 4                | None                    |   -- | 2.017 to 3.797           | 5.3 to 5.7               |
| April 11, 2015 | 22:26 to 22:40 | Fruitland outcrop #3 | 37.236   | -108.867  | 4                | None                    |   -- | 1,944 to 2,990           | 4.5 to 4.6               |
| April 12, 2015 | 14:35 to 15:56 | Fruitland outcrop #4 | 37.316   | -107.668  | 8                | 0.03%                   | 0.74 | None                     | 2.051 to 5.213           | 1.6 to 2.7               |
| April 14, 2015 | 15:49 to 16:03 | Fruitland outcrop #5 | 37.105   | -108.070  | 4                | None                    |   -- | 2,652 to 3,452           | 4.2 to 4.9               |
| April 13, 2015 | 19:55 to 20:10 | Gas well | 36.992   | -107.514  | 3                | 0.34%                   | 0.22 | 2,055 to 3,220           | 6.5 to 10.5              |
| April 14, 2015 | 23:37 to 0:26 | San Juan Gas Plant | 36.764   | -108.361  | 7                | 2.20%                   | 0.21 | 2.063 to 4.749           | 5.7 to 65                |
| April 16, 2015 | 20:39 to 21:25 | Well pad B | 36.874   | -107.990  | 3                | 2.20%                   | 0.16 | 1,916 to 3,125           | 2.8 to 30                |
| April 16, 2015 | 21:47 to 22:05 | Well pad C | 36.870   | -107.985  | 3                | 8.10%                   | 0.29 | 2,240 to 3,123           | 27.4 to 100              |
| April 16, 2015 | 23:01 to 23:21 | Well pad E | 36.882   | -108.042  | 3                | 1.0%                    | 0.32 | 2,053 to 5,839           | 10.5 to 390              |
| April 16, 2015 | 23:38 to 23:53 | Well pad F | 36.909   | -108.085  | 3                | 2.40%                   | 0.34 | 2,132 to 2,309           | 8.0 to 12.2              |
| April 17, 2015 | 22:00 to 22:40 | Well pad | 36.651   | -107.940  | 6                | 2.65%                   | 0.20 | 1,937 to 2,985           | 2.9 to 30                |
| April 17, 2015 | 1:34 to 1:56 | Well pads near counselor | 36.217   | -107.512  | 5                | 16.8%                   | 0.94 | 2,094 to 6,848           | 42.6 to 870              |
| April 19, 2015 | 18:56 to 20:10 | Underground San Juan coal mine | 36.796   | -108.375  | 7                | 0.90%                   | 0.10 | 2,005 to 2,801           | 3.8 to 10.9              |
| April 21, 2015 | 16:17 to 16:32 | Williams Plant | 37.144   | -107.787  | 4                | 12.40%                  | 0.54 | 1,979 to 2,844           | 2.0 to 110               |
| April 21, 2015 | 19:26 to 21:07 | Navajo surface coal mine | 36.527   | -108.483  | 4                | –                      | –     | 1,880 to 1,905           | 1.7 to 2.7               |
| April 21, 2015 | 22:45 to 1:18 | Compressor GCP1-3026 | 36.724   | -107.850  | 7                | 9.20%                   | 0.99 | 1,900 to 3,317           | 3.1 to 136               |
| April 23, 2015 | 19:42 to 19:54 | Turley compressor | 36.769   | -107.791  | 5                | 8.10%                   | 0.99 | 1,926 to 4,748           | 6.2 to 233               |
| April 28, 2015 | 17:01 to 19:18 | Utah Paradox Basin, background in area | 36.527   | -108.483  | 6                | 6.10%                   | 0.96 | 1,904 to 1,965           | 2.6 to 6.1               |
| April 29, 2015 | 20:35 to 20:57 | Gas well pad | 37.013   | -107.628  | 4                | 1.75%                   | 0.95 | 1,993 to 2,479           | 5.3 to 13.7              |
| April 29, 2015 | 21:47 to 22:07 | Compressor | 36.815   | -107.619  | 3                | 16.1%                   | 0.162| 2,457 to 5,586           | 14.2 to 65               |

R$^2$ is only reported for sites with four or more discrete samples. Slope unit is ppb/ppb.
Our results suggest that upstream and midstream facilities with natural gas burning engines may have temporally varying combustion efficiencies and that some could have significant leaks. More coordinated and representative research for a representative subset of sites would be necessary to better characterize and mitigate emissions and fat tail drivers for \(\text{CH}_4\), \(\text{VOCs}\), \(\text{CO}\), and other air pollutants from enhanced recovery operations with natural gas engines and from centralized gathering and processing stations.

4.2.2. Durango to Farmington transects and nighttime \(\text{CH}_4\) accumulation at the surface in the SJB

The North–South road between Durango, CO, and Aztec (or Farmington), NM, was driven multiple times during the campaign. Figure 4 shows the change in surface \(\text{CH}_4\) levels for morning, afternoon, and evening drives, respectively. Small-scale \(\text{CH}_4\) spikes of several hundred ppb to several ppm above baseline levels attributable to nearby point source emissions were recorded along all drives.

Three drives completed before 10 a.m. LT (April 17, 20, and 29, 2015) show higher overall \(\text{CH}_4\) levels for both the baseline and local (100 m to a few kilometers long) enhancements compared to later in the day drives. On these drives, \(\text{CH}_4\) baseline levels are at background levels at 1.9 ppm to 2 ppm in Durango (37.27N) and then start increasing slowly south of 37.2N to reach 2.3 ppm to 2.8 ppm depending on the day. Baseline \(\text{CH}_4\) levels increase more sharply around 37.1N to reach over 4 ppm where the Animas and Florida rivers meet near 37.06N (near Hwy 550 and county roads 213 and 110). On the New Mexico side, the two drives with measurements obtained before 8 a.m. LT have baseline \(\text{CH}_4\) levels ranging between 2.9 ppm and over 10 ppm (detected near Aztec, NM).

Afternoon and evening measurements also show spatial and day-to-day variability in baseline \(\text{CH}_4\) levels but of much smaller magnitude. Measured baseline \(\text{CH}_4\) levels along the transect were contained between 1,880 ppb and 2,000 ppb between noon and 5 p.m. and between 1,888 ppb and 2,100 ppb between 6 p.m. and 10 p.m. (all LSTs).

To further look into the accumulation of \(\text{CH}_4\) at the surface in the SJB at nighttime, the CU/INSTAAR van conducted six back and forth transects between Durango and Aztec, starting at 10 p.m., 2 a.m., and 6:48 a.m. on April 19 to 20, 2015. The mean surface winds between 10 p.m. and 8 a.m. that night at the five monitoring stations ranged from 0.7 m/s at Shamrock to 2.5 m/s at the San Juan Substation. In situ \(\text{CH}_4\) from the different drives is described in SM Section 2.4 and shown in Figure S13. The later transects between 2:00 a.m. and 7:25 a.m. show increased \(\text{CH}_4\) south of 37.2N compared to the back and forth transect between 10 p.m. and 11 p.m. The CU/INSTAAR van took different roads for the northern portion of transects going southward and northward. County Road 213 was taken for the northbound transects. It follows the Animas River and is at a lower elevation compared to Highway 550. Surface \(\text{CH}_4\) levels were significantly higher along CR213.

On April 20, 2015, the NOAA van also took advantage of the low surface winds to map \(\text{CH}_4\) and NMHC accumulation along a loop between Durango-Farmington-Navajo Lake-Durango (4 a.m. to 8:30 a.m. LST) on major roads and close to major riverbeds. The NOAA van in situ \(\text{CH}_4\) measurements and analysis results for 17 flasks collected along the drive are shown in Figures S14 and S15. Surface \(\text{CH}_4\) was elevated for most of the drive, and \(\text{CH}_4\) and NMHC mixing ratio enhancements in flasks collected in New Mexico are strongly correlated. The van measurements illustrate the increased impact of local \(\text{CH}_4\) sources on surface air composition at night and early morning when limited air circulation leads to the pooling of emissions near sources, especially in low elevation portions of the basin.

4.2.3. Ground flask results summary

To further investigate the air composition in the basin, discrete air samples were collected in NOAA flasks on the ground (212 samples) and by aircraft (101 samples; Figure 5). Unfortunately, the NOAA van did not sample surface air in the SJB uniformly. The lack of paved access roads and limited sampling time did not give us the opportunity to explore with the van a large area south of Bloomfield and north of Counselor. Boundary layer air over this area was sampled, however, by aircraft (Section 4.3). In this section, we will focus on summarizing the main findings for the multi species analysis of surface air samples.

Figure 6 (Panels A to D) shows correlation plots for \(\text{CH}_4\) and several NMHC analyzed by NOAA GML in the surface flasks. The flask data show a large spread in \(\text{C}_2\text{H}_6\)-to-\(\text{CH}_4\) correlation slopes with lower slopes (<5%) for samples collected in the northern SJB, where CBM and dry natural gas are produced and where there is degassing of \(\text{CH}_4\) from the Fruitland coal outcrop. Three samples...
collected downwind of the Williams natural gas processing plant near Ignacio, CO, have a $\text{C}_2\text{H}_6$-to-$\text{CH}_4$ correlation slope of 12.4% (Table 3). The facility treats raw natural gas from Colorado and New Mexico and is likely a complex mix of sources with different emission signatures. Air samples collected in New Mexico had a much wider range of $\text{C}_2\text{H}_6$-to-$\text{CH}_4$ correlation slopes from 0% to >15%. In the next section, this observation is confirmed by the more spatially extensive aircraft in situ $\text{CH}_4$ and $\text{C}_2\text{H}_6$ measurements over the SJB.

### 4.3. Aircraft in situ and discrete sample measurements

Two aircrafts equipped with fast-response highly sensitive $\text{CH}_4$ and $\text{C}_2\text{H}_6$ measurements mapped levels in the PBL throughout the SJB. They mostly sampled the convective boundary layer air between 11 a.m. and 5 p.m. LT on clear-sky days.

On steady wind days, mass balance downwind transects were conducted later in the afternoon to capture the outflow from “fresh” (as opposed to “accumulated”) emissions. $\text{CH}_4$ enhancements measured in the PBL downwind on the north or eastern edge of the SJB during mass-balance flights reached 30 to 90 ppb (Figure 1 in Smith et al., 2017). These downwind levels are a direct function of upward cumulative $\text{CH}_4$ emissions, how fast the airmass traveled over the source region, and the depth of the PBL (see details on aircraft mass balance method and implementation in Karion et al., 2013).

Here, we analyze PBL (<3,000 masl) $\text{CH}_4$ and $\text{C}_2\text{H}_6$ enhancements for five SA Mooney and two NOAA P-3 survey flights during midday and afternoon hours and for...
latitudes between 35.00N and 37.52N. During these flights, PBL horizontal winds were mostly from the West, which allowed us to attribute intercepted emission plumes to sources in Colorado for latitudes above the 37N State borderline and in New Mexico for latitudes below 37N.

Figure 5. Map of flask air sampling locations for the NOAA Mobile Laboratory (ML, red circles) and the NOAA P-3 and SA Mooney aircraft (blue) during the April 2015 campaign. DOI: https://doi.org/10.1525/elementa.038.f5

Figure 6. Correlation plots for C2H6-to-CH4, C3H8-to-C2H6, and iC5H12-to-nC5H12 in the ML (A to C) and aircraft (D to F) SJB discrete air samples in April 2015. DOI: https://doi.org/10.1525/elementa.038.f6
4.3.1 In situ CH4 and C2H6 data analysis

Date and time windows for the SA Mooney and NOAA P-3 flights are listed in Table 4. Horizontal winds along the flight track were mainly from the West (see Mean Wind Tables 5 and 6), which is the typical flow in the region in the afternoon. Figures S16 and S17 show the flight track color-coded by CH4 or C2H6 for each flight, as well as a C2H6 versus CH4 correlation plot color-coded by latitude. Figure S18 shows a comparison of flask and in situ CH4 and C2H6.

Each survey flight consisted of several latitudinal transects, close to perpendicular to the mean horizontal wind direction in the PBL. Multiple CH4 and C2H6 emission plumes were intercepted on each flight. The detected enhancements in an emission plume reflect the emission source composition and strength as well as atmospheric dispersion from the point source and the aircraft.

The mean horizontal wind speed in the PBL was lowest at 3.5 m/s on the April 9, 2015, flight. This flight had the largest range of enhancements for CH4 and C2H6 of all survey flights. This was likely a combination of the early takeoff time (11:50 a.m. LT), the lower wind speed, and limited vertical dilution of the plume due to lower PBL height (below 3,000 masl). The flight mean horizontal wind speed was highest (10 m/s to 11 m/s) on April 6 and April 20, 2015. Measured CH4 and C2H6 mixing ratios covered a wider range on April 20, 2015, than on April 6 and April 20, 2015. The flight mean horizontal wind speed (below 3,000 m) on April 6 and April 20, 2015, was 3.5 m/s on the April 9, 2015, flight. This flight had the lowest mean horizontal wind speed in the PBL. Multiple CH4 and C2H6 emission plumes were intercepted on each flight. The detected enhancements in an emission plume reflect the emission source composition and strength as well as atmospheric dispersion from the point source and the aircraft.

| Date          | Time window       | Aircraft          |
|---------------|-------------------|-------------------|
| April 6       | 2:30 p.m. to 6:00 p.m. | SA Mooney         |
| April 9       | 11:50 a.m. to 4:20 p.m. | First survey     |
| April 19      | 12:00 p.m. to 3:30 p.m. | Second survey    |
| April 30      | 1:40 p.m. to 6:30 p.m. | SA Mooney         |

All SA Mooney survey flights show multiple enhancements of CH4 (up to 300 ppb) and C2H6 (up to 35 ppb) above the local background levels in the PBL. These enhancements are detected in various locations over Colorado, New Mexico, and sometimes on the edges of the SJB. C2H6 to CH4 correlation plots show clusters of data along various transects.
correlation slopes, which depend strongly on latitude. Drier gas operation emission plume \( C_2H_6 \) to \( CH_4 \) slopes (0% to 5%) are observed over CO (>37N), while slopes over New Mexico range mostly between 3% and 10%.

The NOAA P-3 conducted two survey flights with very similar spatial patterns. On March 24, 2015, the flight concluded with 3 N-S transects on the E side of the SJB. On April 19, 2015, the aircraft completed one SJB survey from 10:40 a.m. to 1:19 p.m. (LT) and repeated the same survey over the SJB in the afternoon, leaving the region at 3:30 p.m. Figure S17 shows the NOAA P-3 flight data, separating the April 19, 2015, first and second basin surveys (A and B). The early flight on April 19, 2015, has a lower mean horizontal wind speed, and it exhibits the largest enhancements and a very clear latitudinal dependence (A and B). The early flight on April 19, 2015, has a lower mean horizontal wind speed, and it exhibits the largest enhancements and a very clear latitudinal dependence of the SJB detected \( CH_4 \) hot spot was located over Colorado. In April 2015, wells in Colorado produced 33% of the total detected \( CH_4 \) anomaly and 85% of the total detected \( C_2H_6 \) anomaly located over New Mexico.

Next, we explain the in situ \( CH_4 \) and \( C_2H_6 \) aircraft data analysis (see also SM Text Section 2.7 and Figure S19). We assume that the two aircraft data sets represent independent and representative sampling of the basin \( CH_4 \) emissions on a few days in spring 2015. Similarly to other studies, we attribute \( CH_4 \) emission plumes to different sources depending on whether \( C_2H_6 \) is co-emitted, as is expected for thermogenic gas sources (Smith et al., 2015; Mielke-Maday et al., 2019).

For each flight, we first derive PBL \( CH_4 \) and \( C_2H_6 \) enhancements by subtracting for each flight the observed minimum \( CH_4 \) and \( C_2H_6 \) PBL mixing ratios from the time series. To reduce the impact of high-frequency noise on the \( C_2H_6 \) in situ measurements and reduce the impact of different sampling frequencies and cell volumes for the two trace gas analyzers, we apply a 10-s running average to the \( CH_4 \) and \( C_2H_6 \) enhancement time series, noted \( \Delta CH_4 \) and \( \Delta C_2H_6 \) thereafter.

For data intervals between two successive smoothed \( \Delta CH_4 \) local extrema (from a local minimum to a local maximum or from a local maximum to a local minimum), we calculate the integrated enhancements \( i\Delta CH_4 \) and \( i\Delta C_2H_6 \) along the flight path interval (ppb km). If the maximum \( \Delta C_2H_6 \) in the interval is above 100 ppt, we compute the \( \Delta C_2H_6 \) to \( \Delta CH_4 \) correlation slope, y-axis intercept, and the associated coefficient of determination \( R^2 \).

For each flight, we calculate the total integrated enhancements (TIE) for both \( \Delta CH_4 \) and \( \Delta C_2H_6 \) data intervals have \( R^2 > 0.5 \). The mean y-axis intercept for the correlation plots ranges between 43 ppt and 50 ppt, depending on the flight with a standard deviation between 26 ppt and 63 ppt. This suggests that our single PBL background correction for each flight to derive local enhancements and then interval correlation slopes is sufficient.

To separate different source contributions, we assume that if \( \Delta CH_4 \) and \( \Delta C_2H_6 \) in an interval have an \( R^2 > 0.5 \), the two gases are co-emitted by a nearby fossil fuel source or group of sources in the ratio of their observed correlation slope (similar to ground sampling results in Section 4.2).

Conversely, we assume that flight intervals with no \( \Delta C_2H_6 \) enhancements or with \( R^2 < 0.5 \) are contributed by \( CH_4 \) sources with no \( C_2H_6 \)--such as the degassing sampled along the Fruitland coal outcrop in La Plata County and on SUIT land or by biogenic sources or by a mix of sources with different composition leading to poor correlation between \( CH_4 \) and \( C_2H_6 \).

Aggregated results for both aircraft enhancement data sets are in Tables 5 and 6. Our analysis shows that ~25% of the SJB detected \( CH_4 \) hot spot was located over Colorado. In April 2015, wells in Colorado produced 33% of the SJB gas and CBM production and represented 13.4% of active gas and CBM wells in the basin. Both data sets have ~75% of the total detected \( CH_4 \) anomaly and 85% of the total detected \( C_2H_6 \) anomaly located over New Mexico.

For both aircraft data sets, we identified over 600 plume segments with correlated \( CH_4 \) and \( C_2H_6 \) enhancements.
The distribution of the CH4 enhancements over the SJB is very similar to the downwind CH4 plume measured by the NOAA Twin Otter on two flights (Figure S22). Ninety-five percent and ninety-eight percent of the observed CH4 TIE over Colorado and New Mexico, respectively, were due to intervals with at least 10 ppb CH4 enhancements. We find that 72% to 85% of observed CH4 and C2H6 enhancements were distributed all over the basin (Figures S20 and S21 for Mooney data only). As mentioned earlier, larger enhancements were observed on aerial PBL surveys before noon and when horizontal PBL winds were low (~< 5 m/s). The latitudinal distribution of the CH4 enhancements over the SJB is very similar to the downwind CH4 plume measured by the NOAA Twin Otter on two flights (Figure S22). Ninety-five percent and ninety-eight percent of the observed CH4 TIE for intervals with \( R^2 > 0.5 \) over New Mexico and Colorado, respectively, were due to intervals with at least 10 ppb gradients in CH4.

We find that 72% to 85% of observed CH4 and C2H6 TIE over the SJB were attributable to fossil fuel sources. For the Colorado portion of the basin, the aircraft data analysis results show that 69% to 91% of the observed CH4 and C2H6 were attributable to fossil fuel sources.

If we use \( R^2 > 0.75 \) instead of \( R^2 > 0.50 \), the fractions of well-correlated enhancements are lower, and they explain 50% (63%) of the elevated CH4 for the northern SJB and 58% (63%) for the southern SJB for the SA Mooney (NOAA P-3) flights.

Next, we look at the relative contribution of intervals with different C2H6 to CH4 correlation slopes to the detected CH4 and C2H6 enhancements for the five SA Mooney flights combined (Figure 7). The curve origin at 0% slope is the TIE for intervals with either \( R^2 < 0.5 \), or no C2H6 enhancement, or a \( \Delta C_2H_6 \)-to-\( \Delta CH_4 \) slope less than 0.02%. We observed a much wider range of \( \Delta C_2H_6 \)-to-\( \Delta CH_4 \) emission plume ratios over the southern SJB (0.50% to 66%) compared to the northern SJB (>0.1 to ~9%), as was documented with ground observations (see Section 4.2). Further below, we use these ratios to separate contributions from natural gas, CBM, and oil operations in the southern SJB.

Finally, we investigate how the aircraft data analysis results are affected by flight time. We use the TIE results for six 1-h time windows between noon and 6 p.m. LT, merging all five SA Mooney flights. It is important to note that the basin coverage varies for each hourly interval. The median fraction of CH4 TIE with \( R^2 > 0.5 \) and a \( \Delta C_2H_6 \)-to-\( \Delta CH_4 \) slope >0.02% is 68% over Colorado and 76% over New Mexico, which is in good agreement with the time aggregated Mooney flight results we have presented above. Minimum and maximum hourly fractions are shown in Table S7.

### 4.3.2. NMHC in aircraft flasks and NMHC TD emission estimates

Oil, NG, and CBM operations in the SJB are known to be a significant source of ozone precursor emissions (Parikh et al., 2017a). Our flask measurements in gas plumes showed that when both CH4 and C2H6 were enhanced, measured C3-C5 alkanes were also elevated and well correlated. We conclude that the bulk of the measured C2-C5 alkanes are very likely co-emitted with uniform ratios throughout the basin (Figure 6, Panels D to F). Table 7 gives a summary of NMHC correlation slopes calculated for the SJB aircraft flask data by orthogonal difference regression, assuming a 10% relative uncertainty on all NMHC measurements.

Here, we also derive TD emission estimates for the measured C2-C5 alkanes. We first scale the average, low, and high aircraft mass-balance estimates of total CH4 emissions from Smith et al. (2017) using the ratio of CH4 TIE to CH4 TIE for the five SA Mooney flights. We obtain three emission scenarios: mean, low, and high. The mean basin-scale emission estimate for C2H6 is 5.0 ± 1.9 tonnes/h, with a low-high range of 2.9 to 7.8 tonnes/h (Table 8).

#### Table 7. NMHC correlation slopes for 101 air samples collected by aircraft over the San Juan Basin in April 2015. DOI: https://doi.org/10.1525/elementa.038.t7

| Species         | Slope (ppt/ppt) | \( R^2 \) |
|-----------------|-----------------|----------|
| C3H8 to C2H6    | 0.476 ± 0.014   | 1.0      |
| iC4H10 to C2H6  | 0.086 ± 0.003   | 1.0      |
| nC4H10 to C2H6  | 0.145 ± 0.006   | 0.99     |
| iC5H12 to C2H6  | 0.049 ± 0.002   | 0.98     |
| nC5H12 to C2H6  | 0.041 ± 0.002   | 0.98     |
| nC6H13 to iC4H10| 1.68 ± 0.02     | 1.0      |
| iC4H12 to nC4H12| 1.18 ± 0.02     | 1.0      |
| C2H6 to nC4H14  | 0.21 ± 0.02     | 0.60     |

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To derive emission estimates for the other NMHC, the C₂H₆ flux estimates are scaled by the aircraft samples measurement correlation slopes reported in Table 7. The mean basin-scale emission estimate for the measured C₃–C₅ alkanes is 6.8 ± 2.5 tonnes/h, with a low–high range of 3.9 to 10.6 tonnes/h (Table 8).

Our mean emission estimates for C₃-C₅ adds up to three fourth of the WESTAR inventory total VOC emission estimate of 9.3 tonnes/h (Parikh et al., 2017a, 2017b). Our estimate does not include all VOCs emitted by O&G sources such as higher alkanes and methanol. The WESTAR SJB inventory data publically available do not contain a breakdown of VOC emissions by species, which limits the interpretation of our finding. The WESTAR inventory has 90% of O&G total VOC emissions in the SJB attributed to operations in New Mexico (Parikh et al., 2017b). This is close to the 85% New Mexico versus 15% Colorado partitioning we derived for the SJB C₂H₆ anomaly detected by aircraft in spring 2015.

The emission estimates for C₂H₆ in the SJB are similar in magnitude to emissions from dry gas producing basins reported by Peischl et al. (2018). The emission estimates for C₃-C₅ NMHCs for the SJB are significantly lower than the 25.4 ± 8.2 tonnes/h reported by Pétroń et al. (2014) for the Denver Basin, which produces oil, liquid condensate, and wet gas in NE Colorado.

4.3.3. San Juan Basin methane emission attribution

In this section, we use the aircraft CH₄ to C₂H₆ interval analysis results to quantify relative contributions from different CH₄ sources to the hot spot detected by aircraft.

In the vicinity of the San Juan underground coalmine in NM, the SA Mooney detected 16 plume intervals on five different flights with C₂H₆-to-CH₄ correlation slopes ranging between 0.8% and 1.5% (compared to 0.9% for ground flasks collected in the air shaft emission plume on June 14, 2015). These plumes contributed to 2% of the SA Mooney total CH₄ TIE for NM. The NOAA P-3 detected a plume on each survey in the vicinity of the San Juan coalmine, and these plumes contributed 1.4% of the NOAA P-3 total CH₄ TIE for NM.

We postulate that plume intervals with correlated C₂H₆ and CH₄ (R² > 0.5) and a C₂H₆-to-CH₄ slope between 0.16% and 10%—and not attributed to the San Juan coalmine—are attributable to NG or CBM operations in CO or NM. The iAC₂H₆-weighted mean ΔC₂H₆ to ΔCH₄ correlation slope for these plumes is 2.3% (2.7%) in the northern SJB and 4.6% (4.4%) for the southern SJB in the SA Mooney (NOAA P-3) data set. This reflects the larger fraction of dry gas and CBM production in Colorado. Our analysis shows that 66% (SA Mooney) to 75% (NOAA P-3) of the detected CH₄ enhancements fall in the NG and CBM category. Table 9 and Figure 8 summarize the source attribution results discussed below.

The U.S. Geological Survey reports that most coal beds in the SJB Fruitland formation will produce thermogenic CH₄ that can be wet or dry (Ridgley et al., 2013). Therefore, some CBM wells in the basin produce very dry gas with no detectable C₂H₆. CH₄ emissions or gas vented from these wells will not show co-emitted C₂H₆ and will show up in the “Other” category. This means that 66% is likely a lower bound estimate of the NG and CBM operations combined contribution to the observed SJB CH₄ hot spot.

Wetter gas (with larger fraction of C₂-C₅ relative to C₁-C₅) is often a sign of a less mature hydrocarbon reservoir with natural gas liquids coproduced with CH₄. We postulate that plumes with >10% slopes are due to emissions from condensate or oil and associated gas operations. Only in New Mexico did the aircraft detect plumes with correlated C₂H₆ and CH₄ and slopes >10%. These plumes represented 8% of CH₄ TIE and 13% of C₂H₆ TIE for New Mexico and ~ 6% of the CH₄ TIE for the entire basin.

Oil production in the SJB increased from an average of 7.2x10³ bbl/day over the 2003 to 2009 period to 23.9 x 10³ bbl/day in April 2015. This last figure represented 6% and 0.25% of the State and U.S. total oil production volumes, respectively. Over this period, NG and CBM production in the basin decreased by 34%. In 2019, mean daily production statistics for the basin were lower for oil (~4%), natural gas (~8%), and CBM (~26%) compared to the April 2015 numbers.

The annualized TD mean estimates for the SJB total CH₄ emissions for 2003 to 2009 and April 2015 are similar (Kort et al., 2014; Smith et al., 2017). However, there is no attribution for the early period and for recent years. A better understanding of emission magnitudes and trends for the SJB and other basins would require long-term BU and TD emission quantification and attribution studies.

Based on what has been reported so far, CH₄ seepage from the Fruitland coal outcrop could only occur over Colorado. Based on our surface air sampling in outcrop seepage gas plumes, we postulate that outcrop gas has not co-emitted C₂H₆. Emission plumes with no C₂H₆-to-CH₄ enhancement correlation detected over Colorado were categorized as “CO coal outcrop and Other.” Our analysis shows that CH₄ enhancements detected over Colorado and with no correlated C₂H₆ or a C₂H₆-to-CH₄ correlation slope <0.16% represented a smaller fraction

| Species | Mean Estimated Emissions tonnes/h | Low–High Emission Range tonnes/h |
|---------|----------------------------------|---------------------------------|
| CH₄⁺   | 62 ± 23                          | 35 to 96                        |
| C₂H₆   | 5.0 ± 1.9                        | 2.9 to 7.8                      |
| C₃H₆   | 3.5 ± 1.3                        | 2.0 to 5.4                      |
| iC₄H₁₀ | 0.8 ± 0.3                        | 0.5 to 1.3                      |
| nC₄H₁₀ | 1.4 ± 0.5                        | 0.8 to 2.2                      |
| iC₅H₁₂ | 0.6 ± 0.2                        | 0.3 to 0.9                      |
| nC₅H₁₂ | 0.5 ± 0.2                        | 0.3 to 0.8                      |
| C₃-C₅ total | 6.8 ± 2.5                    | 3.9 to 10.6                     |

*Based on Smith et al. (2017).
of the total CH$_4$ TIE: 8% for SA Mooney data and 2% for the NOAA P-3 data.

We conclude that degassing from the outcrop contributed at most between 2% and 8% of the CH$_4$ hot spot observed by the two aircraft. This finding agrees with Smith et al. (2017). Based on flights targeting emission plumes from known outcrop vents SW of Durango, they argued that these sources "are a small fraction of the basin total," ranging from 0.02 to 0.12 Tg CH$_4$ yr$^{-1}$.

The Colorado outcrop emission contribution to the detected CH$_4$ hot spot is much smaller than the 24% BU emission estimates suggest (Table 9). However, outcrop degassing field results have been shown to be variable from year to year (LTE, 2017). Current outcrop gas seepage emission estimates relying on spatially and temporally extrapolated field data likely carry very large uncertainties.

4.3.4. San Juan Basin natural gas and coal bed methane CH$_4$ emissions and infrastructure loss rates

Here, we use mean, low, and high total CH$_4$ emission scenarios based on results from Smith et al. (2017) and the aircraft data attribution results (Table 9) to derive mean, low, and high CH$_4$ emission estimates for NG + CBM and oil systems (Table 10). Mean estimates for both aircraft data sets are between 11 and 15 tonnes/hr for Colorado and close to 30 tonnes/hr for New Mexico for NG + CBM systems and are 2.8 tonnes/hr to 3.9 tonnes/hr for oil operations in NM. The mean estimates we derive for total CH$_4$ emissions from oil, NG, and CBM operations in the SJB are 44.7 tonnes/hr (Mooney) and 53.2 tonnes/hr (P-3). These estimates are based on the analysis of in situ data from a few flights in spring 2015.

For comparison, the WESTAR 2014 SJB annual emission inventory equates to 42.5 CH$_4$ tonnes/hr, assuming a constant emission rate (Parikh et al., 2017b). Note that Parikh et al. (2017a) warn that the SJB annual CH$_4$ emission estimate may be biased low due to missing or inadequate input data. The authors acknowledge that they did not include GHG for point sources with unknown Source Classification Code. Also, the inventory does not include CH$_4$ emissions for nonpoint O&G sources in CO due to the absence of a breakdown of GHG emissions by gas in a CDPHE emission inventory used as input. Finally, the inventory authors assumed a default control efficiency of 98% and a default capture efficiency of 100% for emission sources controlled by flares, for example, tanks, casing-head gas, and dehydrators, which is likely a best-case scenario.

The SJB total gas output was $8 \times 10^7$ m$^3$/day in April 2015. We use a 90% to 99% range for the produced NG or CBM CH$_4$ content in the SJB. Assuming a molar volume for the produced gas of 23.6 L per mole (pressure = 14.73 psi and temperature = 60°F), we derive mean (low–high) CH$_4$ loss rates for the SJB combined NG and CBM operations of...
1.6% to 2.4% (0.9% to 3.8%) in Colorado and 2.0% to 2.4% (1.1% to 3.8%) in New Mexico during the field study for both aircraft data sets (Table 10).

These leak rates are similar to the 2015 national average estimates based on the EPA GHGI (1.4%) and on Alvarez et al. (2018; 2.3%). They are also comparable to TD results from other U.S. onshore O&G basins studied in 2015 (Figure 5 in Peischl et al., 2018).

### 5. Discussion

The detection of locally or regionally elevated XCH$_4$ with aerial or satellite remote sensing can help identify strong sources, especially if (1) emissions persist over days to weeks (Thompson et al., 2016; Pandey et al., 2019; Varon et al., 2019; Cusworth et al., 2020), several months (de Gouw et al., 2020), or several years (Kort et al., 2014; Schneising et al., 2014; Shim et al., 2018), (2) emissions tend to accumulate at the surface (under strong temperature inversions, e.g., de Gouw et al., 2020) or show distinct plumes (Frankenberg et al., 2016), (3) sources are spatially separated (Cusworth et al., 2018), and (4) the area is well observed from space resulting in sufficient “good” data coverage (high albedo, low cloud, low aerosols, etc., all parts of data quality filters defined for each remote sensor and retrieval data product; de Gouw et al., 2020). It is important to recognize that the improved spatial coverage offered by remote sensing products can hide some of the complexity, biases, and total uncertainty attached to column retrievals, potentially affecting the data analysis and interpretation (Zhang et al., 2017; Varon et al., 2018; see more below).

In our paper, we use the 2015 campaign in situ aircraft data for CH$_4$ and C$_2$H$_6$ to attribute a mean total CH$_4$ emissions estimate derived from flights in April 2015 (Smith et al., 2017) to different sources. This allows us to estimate emissions for O&G sources and to calculate a mean CH$_4$ loss rate for CBM and natural gas operations for the time of the campaign.

We are not able to conduct a comparison of the SJB April 2015 CH$_4$ hot spot attribution with the earlier study by Kort et al. (2014), which looked at total basin-wide emissions. Given the large year-to-year variability in the reported surface chamber CH$_4$ flux measurements of the Fruitland coal outcrop surface degassing in La Plata County, we cannot estimate accurate outcrop mean emissions over 2003 to 2009. Also, we do not have TD constraints to quantify O&G CH$_4$ emissions separately for that earlier time period.

The main difference between the earlier study and our paper lies in our effort to use a suite of long-term and campaign mode measurements to explain the different factors leading to the SJB CH$_4$ pollution as observed at the surface with instrumented vehicles, in the PBL with aircraft, and in the total air column by satellite. The region has strong local sources, emissions trapping due to topography, and wind patterns at night and pristine surrounding mountains. The basin is expected to stand out as an XCH$_4$ local anomaly, but it should not be the only one, as noted further below.

All U.S. O&G basins our team has visited in the last decade have much higher CH$_4$ levels at the surface at

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### Table 10. Summary statistics for CH$_4$ BU emissions, PBL CH$_4$ enhancements, and CH$_4$ loss rates for the SJB. DOI: https://doi.org/10.1525/elementa.038.t10

| Categories | Colorado | New Mexico |
|------------|----------|------------|
|            | Bottom-Up | SA Mooney  | NOAA P-3   | Bottom-Up | SA Mooney  | NOAA P-3   |
| Fraction of basin CH$_4$ TIE (%) | 38.8 | 26.5 | 27.2 | 61.2 | 73.5 | 72.8 |
| Fraction of CO or NM CH$_4$ emissions or TIE attributed to (%) | 32.9 | 69.3 | 91.5 | 83.8 | 65.5 | 73.3 |
| NG + CBM* | 1.0 to 1.1 | 1.6 to 1.7 | 2.2 to 2.4 | 2.2 to 2.4 | 2.0 to 2.1 | 2.2 to 2.4 |
| Oil | - | - | - | 2.6 | 6.1 | 8.5 |
| Underground coal mine | - | - | - | 11.3 | 2 | 1.5 |
| Total for fossil fuel systems | 32.9 | 69.3 | 91.5 | 97.7 | 73.6 | 83.3 |
| Weighted average NG + CBM C$_2$H$_6$-to-CH$_4$ slope* (%) | - | 2.3 | 2.7 | - | 4.6 | 4.4 |
| Mean (low–high) CH$_4$ emissions attributable to NG+CBM systems* (tonnes/h) | (6.5 to 176) | (8.8 to 23.9) | (17 to 46.2) | (18.9 to 51.2) |
| Mean NG + CBM system CH$_4$ loss rate* (emissions/production in %) (low–high) | (0.9 to 2.7) | (1.4 to 3.8) | (1.1 to 3.4) | (1.3 to 3.8) |

*Only plumes with 0.16% < C$_2$H$_6$-to-CH$_4$ slope < 10%.
*Removing the San Juan coal mine contributions.
*Assuming produced NG is 90% or 99% CH$_4$ and using low/high CH$_4$ emission scenario.
night and early morning under low wind conditions. The accumulated pollution from nocturnal emissions will dilute vertically as the boundary layer grows and then be flushed out of the region. This flushing happens sooner over a flat terrain than in a topographically basin like the SJB—which also has downslope surface winds at night that carry emission plumes E/SE (see illustrations in supplementary material Figure S23). Below, we use a simple calculation to translate a typically observed nighttime CH₄ surface air anomaly into an early and midmorning vertical column mean enhancement.

The vertical sensitivity of the SCIAMACHY CH₄ column retrievals from nadir shortwave infrared radiation spectra—for cloud free pixels—mostly follows the air pressure down to the surface for solar zenith angles <70° (Buchwitz et al., 2005; Frankenberg et al., 2006). Therefore, the SCIAMACHY retrievals should be a reasonable estimate of CH₄ air column mixing ratio averages. The total column retrievals should be insensitive to the height of the PBL. Any vertical mixing of CH₄-rich surface air without horizontal dispersion will not change the column average mixing ratio over a location.

With a midmorning overpass time (10:40 a.m. ± 40 min LST), the SCIAMACHY satellite detected columns with enriched CH₄ near the surface, resulting from the accumulation of overnight and early morning emissions. A simple calculation shows that it takes a 4-ppm CH₄ enhancement in a 100 m surface layer (~1% of the atmospheric column over the SJB in pressure) to create a 40 ppb total atmospheric column XCH₄ enhancement above the local background column. This is very close to the largest mean column anomaly depicted in the local anomaly map by Kort et al. (2014). As shown in Section 4.2.2, 2 ppm to 4 ppm surface air CH₄ large-scale enhancements were detected in the SJB by the NOAA and CU INSTAAR vans at night and early morning before the onset of the PBL growth and surface air dilution with cleaner air from above entrainment.

The 2 ppm to 4 ppm surface air enhancements reflect the accumulation of local emissions under nocturnal and early morning low surface air dispersion conditions. Our analysis of wind regimes over the SJB implies that on most days, the SCIAMACHY satellite midmorning overpass time occurs before the nightly CH₄ emissions buildup in the basin has been flushed away. In other words, the satellite likely scanned the region’s atmosphere close to when the PBL CH₄ pollution peaks and when the basin’s airshed stands in stark contrast with “cleaner” air columns over nearby remote (source-free) and high-elevation terrain.

The mean TD CH₄ emission estimate reported by Smith et al. (2017) for the SJB is 62 ± 23 tonnes/h. This is of similar magnitude as aircraft mass balance estimates derived for other U.S. O&G producing regions: the Barnett Shale, TX (66 ± 22 tonnes/h; Peischl et al., 2018), the Uinta Basin, UT, (55 ± 15 tonnes/h; Karion et al., 2013), and the Haynesville Shale, LA, and TX (51 ± 16 tonnes/h; Peischl et al., 2018). Only the Barnett was a mature shale gas formation in the United States by 2009. The Uinta Basin in NE Utah has been producing natural gas for decades. These two regions did not show up as hot spots on the Kort et al. (2014) 2003 to 2009 local anomaly map.

The SJB TD CH₄ emission estimate is also less than half of an inverse model emission estimate for the SJV, CA (Jeong et al., 2016), where dairy livestock is the largest CH₄ source. The southern SJV appears as a smaller CH₄ hot spot than the SJB on the Kort et al. anomaly map. This may be the result of the nocturnal surface CH₄ emission buildup dispersing more effectively by midmorning in the SJV than in the SJB.

A detailed analysis of the various potential factors impacting the detectability of known large CH₄ source regions in different satellite data products would be quite valuable, but (1) it is beyond the scope of this paper and (2) a study team with satellite data experts would better accomplish it.

In the SJB, most afternoons, stronger wind speeds and a deeper PBL acted in concert to reduce the CH₄ pollution levels detected with the instrumented vans near the surface or with the aircraft in the PBL (Figures 2, 4, and S4). A satellite instrument with the same sensitivity as SCIAMACHY, but with a mid to late afternoon overpass time, may not have detected such a large and persistent CH₄ hot spot over the SJB.

The SJB does not stand out as a regional anomaly in a more recent map of TROPOMI (TROPOspheric Monitoring Instrument) retrievals of XCH₄ over the United States for December 2018 to March 2019 (Figure 1 in de Gouw et al., 2020). This may partly be due to the very limited number of valid retrievals for large portions of the Rocky Mountains during the entire period (Figure S1 in de Gouw et al., 2020). The authors explain that cloud cover, low surface albedo, higher topography, and other factors can affect the retrieved CH₄ data quality. Despite a 1:30 p.m. LT TROPOMI overpass time, de Gouw et al. (2020) note the largest enhancement detected in another U.S. oil and natural gas basin, the Uinta Basin in NE Utah, occurred “over the deepest parts of the Basin, consistent with the accumulation of emissions underneath inversions,” a phenomenon reported by earlier studies focused on explaining the basin’s wintertime surface ozone pollution (Schnell et al., 2009; Oltmans et al., 2014).

A reliable GHG emission monitoring system cannot rely exclusively on remote sensing products (Cusworth et al., 2018; Nisbet et al., 2020). Remote sensing instruments can have radiometers calibrated for radiance, but the quantity of interest is the in situ profile of the dry air mole fraction of CH₄. The latter is estimated by a radiative transfer model, which has to incorporate several assumptions about the atmosphere and the surface. It remains a fundamental weakness of remote sensing that GHG atmospheric column retrievals cannot be calibrated. GHG retrievals can only be compared to relatively sparse and never entirely coincident calibrated in situ measurements or evaluated against ground-based remote sensing data products (Wunch et al., 2010, 2011; Geibel et al., 2012; Buchwitz et al., 2015; Loew et al., 2017).

Atmospheric column gradients are affected by emission strength and distributions, air dispersion, and changes in surface elevation. Satellite retrievals based on
shortwave infrared radiation can show XCH\textsubscript{4} and XCO\textsubscript{2} gradients related to localized emissions, but retrievals can also be affected by other spatially variable factors such as water vapor, clouds, aerosols, and surface reflectivity (Buchwitz et al., 2005; Frankenberg et al., 2006, 2008).

The “translation” of atmospheric GHG in situ measurements or remote sensing retrievals into emission estimates or emission trends for a given region and time period requires careful analysis of the data quality and data representativeness as well as advanced and validated surface flux estimation and attribution methods. Coordinated research and operational data collection are necessary to further refine and compare emission detection, estimation, and attribution methods at various scales.

Vaughn et al. (2018) showed the importance of perfect overlap in time and space of emission estimation efforts to successfully compare temporally varying CH\textsubscript{4} emission for the Fayetteville dry shale gas basin in Arkansas in fall 2015. The authors compared midday spatially resolved emission estimates based on aircraft measurements and concurrent facility-level measurement and operational data. The coordinated research in that particular basin revealed the large contribution of daytime gas venting practices for liquid unloading at well pads to midday basin total CH\textsubscript{4} emissions. A follow-up coordinated pilot study evaluated the potential of CH\textsubscript{4} emission detection and cost-effectiveness of two airborne techniques against the traditional ground-based leak detection and repair method (Schwietzke et al., 2018).

Other coordinated studies in one particular basin (San Juan Basin; Frankenberg et al., 2016; Smith et al., 2017; Barnett Shale, TX; Zavala-Araiza et al., 2015), or spread over different U.S. O&G basins (Alvarez et al., 2018), have brought many useful insights on source processes and emission distributions. Field measurements on-site, downwind of facilities, and from the air have demonstrated how anomalously large emitters contribute a substantial fraction of total O&G emissions in a producing region as well as for each natural gas supply chain segment.

Some independent studies using different atmospheric CH\textsubscript{4} observations or retrievals and emission quantification techniques have shown substantial disagreement between estimates of O&G emission trends over the United States (Bruhwiler et al., 2018; Lan et al., 2016; Turner et al., 2016) or over a specific Basin (Schneising et al., 2014; Peischl et al., 2016).

Here, we have shown that in situ multiple species measurements can be used to characterize CH\textsubscript{4} emission plumes and attribute basin CH\textsubscript{4} emissions to different sources. In situ meteorological and trace-gas measurements throughout the region and at different times of the day offered a more complete picture of the SJB CH\textsubscript{4} sources and hot spot dynamics than previously reported. In terms of CH\textsubscript{4} emissions and loss rate for NG and CBM operations, TD estimates for the region did not stand out from other U.S. O&G basins and were in line with Alvarez et al.’s (2018) mean results for the United States in 2015.

Before concluding this paper, we want to note that field operations and emissions captured during the campaign may not be representative of typical or annual conditions in the SJB. Some O&G activity levels can vary in response to multiple independent factors. The study authors and other researchers, who were in the field during the April 2015 campaign, noticed a more visible presence of O&G technicians in charge of checking site operations and servicing equipment. Reliable and effective O&G sources management, leak detection and repair (LDAR), and malfunction or leak prevention are critical components of emissions mitigation. It is possible that emissions mitigation from increased site visits was enhanced during the campaign. Continuous emissions monitoring systems for operators and more frequent remote sensing and in situ observations for independent emissions quantification will support stronger mitigation and policy-making.

6. Conclusions

For several years, nongovernmental organizations, industry, universities, local, and federal government entities in the United States have partnered to advance anthropogenic CH\textsubscript{4} emission quantification methods and mitigation assessments, especially from the natural gas sector (ICF, 2016; Alvarez et al., 2018; Schwietzke et al., 2018).

Our study has focused on improving the scientific understanding of CH\textsubscript{4} emissions in a fossil fuel rich basin in the southwestern United States, the San Juan Basin, close to the U.S. Four Corners. This region had become known as the U.S. largest local CH\textsubscript{4} anomaly or hot spot based on the analysis of SCIAMACHY satellite XCH\textsubscript{4} retrievals between 2003 and 2009 by Kort et al. (2014).

We provide a breakdown of source contributions to the SJB CH\textsubscript{4} hot spot detected by two aircraft equipped with fast in situ CH\textsubscript{4} and C\textsubscript{2}H\textsubscript{6} measurements in April 2015. Extensive O&G and CBM operations throughout the SJB contributed at least 66% to 75%, while gas seepage from the Fruitland coal outcrop in La Plata County, CO, contributed 2% to 8% at most. Seventy-five percent (85%) of CH\textsubscript{4}(C\textsubscript{2}H\textsubscript{6}) total enhancements over the local background air was detected over New Mexico. Oil operations in New Mexico only contributed 5% to 6% of the detected CH\textsubscript{4} anomaly.

The SJB, similar to other topographical basins with local sources, experiences aggravated surface air CH\textsubscript{4} and NMHC in the southern SJB) pollution as surface emissions accumulate under low wind and during surface temperature inversion at night and until midmorning. The SCIAMACHY satellite captured this atmospheric CH\textsubscript{4} overnight buildup during its midmorning overpass. It takes 4 to 6 h for the typical afternoon westerly winds to flush the CH\textsubscript{4} (and any co-emitted gases) pollution buildup outside of the basin. This nocturnal and early morning enhanced surface air hydrocarbon pollution due to emissions in O&G basins deserves more scrutiny, especially the investigation of potentially repeated exposure to elevated air toxic levels among local residents and industry workers (Mielke-Maday et al., 2019).

The mean leak rate estimates for CBM and natural gas operations in the SJB in April 2015 are 1.6% to 2.4% in Colorado and 2.0% to 2.4% in New Mexico (Table 10, Section 4.3.4). These numbers are in line with TD and BU estimates from other U.S. O&G basins. This implies
that the SJB is not the largest U.S. CH₄ anomaly in terms of O&G emissions or leak rate. However, the impact of these sources on the basin airshed can be amplified by the local topography and meteorology.

A more detailed assessment of fossil fuel production sources and emissions on the SJB air quality would require process-level emission studies, similar to other studies that have unfortunately almost exclusively focused on CH₄ so far (Allen et al., 2013; Alvarez et al., 2018; Vaughn et al., 2018; Zaimes et al., 2019). The SJB, having been a major producer of gas and CBM for decades, is home to a large number of older producing wells. It may be insightful to characterize emissions for facilities of different sizes, types, and ages to better estimate current and potential future hydrocarbon emissions from this large and complex source sector.

This work and previous studies have shown the intrinsic value of coordinated research and operational data collection to compare and refine CH₄ emission detection, estimation, and attribution methods at various scales. At this critical time in the planet and human history, reliable, comprehensive, and up-to-date GHG emission estimates by source type and at scales relevant for policy-making are still lacking. However, as much as accurate GHG emission estimates are valuable, even more time critical is the need for “rapid and deep” mitigation, in other words much bolder emission cuts necessitating substantial societal and industry transformations, to meet the Paris Climate Agreement goals (Nisbet et al., 2019, 2020; Anderson et al., 2020).

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Author contributions

• Contributed to conception and design: GP, BM, BV, JK, ET, IM-M, O-S, SS, SC, JP, CS, AW, PT, RS.
• Contributed to acquisition of data: GP, BM, ET, BV, JK, IM-M, OS, BH, SS, SC, JP, PL, EM, MC, AC, CS, BS, TN, SW, CK, TC, MR, BV, RS.
• Contributed to analysis and interpretation of data: GP, BM, BV, IM-M, OW, ED, BH, SS, LB, BV, PT.
• Drafted and/or revised the article: GP, BV, ED, SS, JP, PT, RS, BM, BH, MC.
• Approved the submitted version for publication: GP, BM, BV, ET, JK, IM-M, OS, ED, BH, SS, SC, JP, PL, EM, MC, AC, CS, TN, SW, DK, LB, CK, TC, AW, MR, PT, RS.

Data Accessibility Statement

Surface meteorological data for monitoring stations in the San Juan Basin are archived in the EPA Air Quality System Data Mart and can be accessed via links provided here: https://www.epa.gov/outdoor-air-quality-data.

NOAA P-3 data from the SONGNEX campaign are accessible via Data menu at: https://www.esrl.noaa.gov/csd/projects/songnex/.

NOAA GML in situ and flask multiple species data can be requested from the corresponding author.

Supplemental files

The supplemental files for this article can be found as follows:

Text S1. Contains Tables S1 to S7, Figures S1 to S23, Supplementary Text. Docx.

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