# Downward Trend in Methane Detected in a Northern Colorado Oil and Gas Production Region Using AIRS Satellite Data

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#### Abstract

The oil and gas (O&G) sector is estimated to be the largest contributor to anthropogenic methane (CH4) emissions in Colorado. Since 2004, the State of Colorado has implemented multiple regulations to significantly reduce emissions from the O&G sector. The Denver-Julesburg Basin (DJ Basin) is a significant O&G producing region in northern Colorado, and O&G production here has steadily increased over the last decade. To assess CH4 trends in Northern Colorado, we selected CH4 retrievals from the NASA Atmospheric Infrared Sounder (AIRS) instrument for 2003-2020. The study grid cell includes Denver, Boulder, and much of the dense O&G production in the DJ Basin. We computed mean June-August ascending node AIRS 700 hPa CH4 for each year and subtracted mean June-August CH4 sampled at NOAA's Niwot Ridge (NWR) station, a high-altitude background site. Differences represent estimated enhancement over background. Linear regression shows an annual change of -2.84 ppb +/- 0.8 ppb from 2012-2020 (R-squared 0.90) and an estimated reduction of 56% for 2012-2020, despite substantial increases in O&G production. Local CH4 enhancement is strongly correlated with surface measurements of ethane at Platteville which is in the center of the O&G fields (correlation coefficient 0.96), and this is evidence that reductions in O&G emissions are driving reductions in CH4. We conclude that AIRS CH4 can be used to measure the efficacy of emissions control programs in this region and that regulatory requirements are having an effect.

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8	Key Points:
9 10 11 12 13 14	<ul> <li>Despite substantial increases in local oil and gas production, methane enhancement in the northern Colorado study area declined significantly from 2012-2020.</li> <li>Decreases in both methane enhancement and ethane support the conclusion that emissions reduction efforts have been effective.</li> <li>AIRS satellite methane retrievals can detect changes in boundary-layer concentrations when mixing heights and source emissions are large.</li> </ul>
15	

#### 16 Abstract

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- 18 (CH<sub>4</sub>) emissions in Colorado. Since 2004, the State of Colorado has implemented multiple
- 19 regulations to significantly reduce emissions from the O&G sector. The Denver-Julesburg Basin
- 20 (DJ Basin) is a significant O&G producing region in northern Colorado, and O&G production
- 21 here has steadily increased over the last decade. To assess  $CH_4$  trends in Northern Colorado, we
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- production in the DJ Basin. We computed mean June-August ascending node AIRS 700 hPa CH<sub>4</sub>
- for each year and subtracted mean June-August CH<sub>4</sub> sampled at NOAA's Niwot Ridge (NWR)
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- background. Linear regression shows an annual change of -2.84 ppb +/- 0.8 ppb from 2012-2020
- (R-squared 0.90) and an estimated reduction of 56% for 2012-2020, despite substantial increases
- in O&G production. Local CH<sub>4</sub> enhancement is strongly correlated with surface measurements
- of ethane at Platteville which is in the center of the O&G fields (correlation coefficient 0.96), and
- this is evidence that reductions in O&G emissions are driving reductions in CH<sub>4</sub>. We conclude
- that AIRS  $CH_4$  can be used to measure the efficacy of emissions control programs in this region
- 33 and that regulatory requirements are having an effect.

# 34 **1 Introduction**

35 According to the Colorado Department of Public Health and Environment (CDPHE)

- 36 (2021), the oil and gas (O&G) sector is estimated to be the largest industrial contributor to
- anthropogenic methane (CH<sub>4</sub>) emissions in Colorado. The Denver-Julesburg Basin (DJ Basin) is
- a significant O&G producing region in the northern portion of the State. O&G production in the
- 39 DJ Basin has steadily increased over the last decade. Figure 1 shows O&G production trends for
- 40 2003-2020 for the Wattenberg Field, the primary resource area in the DJ Basin (data from the
- 41 Colorado Oil and Gas Commission, October 2021, <u>https://cogcc.state.co.us/data.html</u>.) Oil
- 42 production increased by 343% and gas production by 297% between 2012 and 2020.

43 For a variety of reasons, including the fact that a portion of the state that includes the DJ Basin has been designated an ozone nonattainment area by the Environmental Protection Agency 44 45 (EPA), Colorado has implemented regulations to significantly reduce emissions of CH<sub>4</sub> and volatile organic compounds (VOCs) from O&G production facilities. Although initial 46 regulations were focused on ozone reduction goals, many control requirements had associated 47 48 CH<sub>4</sub> emission reduction co-benefits. One of the most significant of these regulatory changes 49 since 2004 was in February 2014 when the Colorado Air Quality Control Commission (AQCC) fully adopted EPA's New Source Performance Standards (NSPS), 40 CFR Part 60, Subpart 50 OOOO, into AQCC Regulation No. 6 and adopted more stringent control requirements for VOCs 51 and hydrocarbons for a variety of O&G sources in AQCC Regulation No. 7. The changes to 52 53 Regulation No. 7 in early 2014 included expanded control requirements for hydrocarbon liquid storage tanks, pneumatic controllers, glycol dehydrators, and components; implementation of a 54 leak detection and repair program; and limitations to venting associated with maintenance and 55 liquids unloading from storage tanks. Since 2014, the AQCC has continued to adopt additional 56 measures to reduce CH4 and VOCs from O&G sources, and this is anticipated to continue 57 through 2021 and beyond. The Regional Air Quality Council (RAQC) and CDPHE (2020) 58

- 59 projected that O&G VOC emissions would decline from 279.7 to 119 tons per day from 2011 to
- 60 2020 within the Denver Metro and North Front Range Ozone Nonattainment Area (a 57%
- 61 reduction).





Figure 1. O&G production in the Wattenberg Field within the DJ Basin. The greatest growth
 occurs between 2012 and 2020.

If these controls have been effective, then we would expect to see reductions in CH<sub>4</sub> in 65 the region after the implementation of new rules in 2014. In order to understand the efficacy of 66 past O&G regulations and drivers for future emission control regulations, this analysis uses 67 satellite and surface measurements to examine CH4 trends and reductions over the past two 68 decades in an area of Northern Colorado that includes the Denver metropolitan area and a 69 portion of the DJ Basin north of Denver that contains a high density of O&G wells. While a 70 variety of government agencies, researchers, and other groups have assessed CH4 and VOC 71 72 concentrations in Colorado from surface and aerial measurements, satellite CH<sub>4</sub> data has not yet been widely used to assess CH<sub>4</sub> trends in Colorado. 73

Three recent papers consider trends in surface measurements of CH<sub>4</sub> or non-methane 74 hydrocarbons (NMHCs) in our study area for shorter time periods within our temporal domain 75 (2003-2020). Oltmans et al. (2021) recently reported trends in CH<sub>4</sub> using air samples collected at 76 the National Oceanic and Atmospheric Administration (NOAA) Boulder Atmospheric 77 Observatory (BAO) tower near Erie in southwestern Weld County. For 2008-2016 they found no 78 79 statistically significant trends in CH<sub>4</sub> relative to background values when flows were from the DJ Basin. Ortega et al. (2021) found a positive trend of 0.9 ppb  $\pm$  0.3 % per year for ethane at a site 80 north of Boulder from 2010-2019 using a solar absorption Fourier Transform InfraRed 81 82 instrument. In a source-apportionment study that considered trends from 2013 to 2016, Lyu et al. (2021) show a reduction of approximately 50% in nonmethane hydrocarbons at the CDPHE 83 Platteville site which is located within the DJ Basin in Weld County. They report that "new 84 85 regulations implemented by the state as well as changes in operating practices made by the industry for other reasons might explain the observation that NMHC mixing ratios at the 86

Platteville site were lower in 2016 than in 2013." We will show that our results are generally

consistent with those from each of these studies and also point to a statistically significant
 decline in CH<sub>4</sub> extending from 2012 through 2020.

Satellites and aircraft studies are now common tools for calculating emissions fluxes and 90 91 the contributions of O&G sources to ambient concentrations of CH4 and VOCs, and because of greater spatial coverage these can yield data more representative of regional emissions than a 92 limited number of surface monitoring sites. In addition, satellite measurements are made on a 93 94 regular basis and typically provide greater temporal coverage than aircraft studies. Recently, de Gouw et al. (2020) published analyses of CH<sub>4</sub> data from the TROPOspheric Monitoring 95 Instrument (TROPOMI) on board the Copernicus Sentinel-5 Precursor satellite, which was 96 launched in October of 2017. In their paper, the authors have identified and quantified CH<sub>4</sub> 97 enhancements over many O&G basins in the United States. TROPOMI has high spatial 98 resolution and greater signal sensitivity than older satellite instruments, but there is not yet a long 99 100 enough data record to identify long-term trends.

101 To assess long-term CH<sub>4</sub> trends in Northern Colorado, we selected the Atmospheric Infrared Sounder (AIRS) instrument launched in 2002 on the Aqua Satellite, because of its 102 almost 20 years of data, its ongoing use, and its provision of an accurate estimate of the rates of 103 change in CH<sub>4</sub> from year-to-year. Zhang et al. (2020) concluded that AIRS CH<sub>4</sub> data are suitable 104 105 for analyses of spatial and temporal patterns across the globe. They demonstrated that AIRS CH4 data are closer to surface concentrations than CH<sub>4</sub> data from the SCanning Imaging Absorption 106 107 spectroMeter for Atmospheric CartograpHY (SCIAMACHY) satellite instrument or the Greenhouse Gases Observing Satellite (GOSAT). AIRS has been used by researchers to quantify 108 regional and national CH<sub>4</sub> changes and trends over both short and reasonably long-time scales 109 (Rendana et al., 2021; Yang and Wang, 2020; and Ribeiro et al., 2016). Wu et al. (2019) recently 110 completed a comprehensive, long-term, analysis of CH4 trends across China using AIRS data 111 from 2002-2016. They conclude that AIRS CH<sub>4</sub> concentrations "showed good consistency with 112 the ground measurements of surface CH4 concentration from the World Data Centre for 113 Greenhouse Gases (WDCGG) ( $R^2 = 0.83$ , p < 0.01), indicating that the remotely-sensed CH<sub>4</sub> 114 reflected the spatial and temporal variations of surface CH<sub>4</sub> concentration". Rendana et al. (2021) 115 also found good agreement between AIRS and surface CH<sub>4</sub> measurements with an R<sup>2</sup> of 0.86. 116

Our goal is to generate an initial estimate of changes in CH<sub>4</sub> and an AIRS-based metric for tracking rates of change in CH<sub>4</sub> that can eventually be linked with potentially more robust

trend studies using TROPOMI and future higher resolution satellite instruments.

### 120 **2 Data Sources and Methods**

We used June through August 2020 AIRS Version 7 retrievals for the 700 hPa level for 121 2003-2020 (https://giovanni.gsfc.nasa.gov/giovanni/). Details of this product are provided here: 122 https://disc.gsfc.nasa.gov/datasets/AIRS3STM 7.0/summary. Summer season data were chosen 123 in part to optimize the relevance of our results to assessments of trends in local O&G VOC 124 125 emissions that contribute to ozone exceedances and also because of the higher sensitivities of the AIRS product when surface mixed layers are deep. The AIRS product is particularly well suited 126 for several reasons. Specifically, we selected the AIRS data for this study due to its long period 127 128 of record, the frequency and timing of overflights, and relatively low uncertainty for CH<sub>4</sub>.

We used NOAA Global Monitoring Laboratory (GML) monthly mean CH<sub>4</sub> data collected at Niwot Ridge (<u>https://gml.noaa.gov/</u>) (Dlugokencky et al., 2021) to estimate local, continental background concentrations. Niwot Ridge (NWR) is a high-altitude site at 40.053°N, 105.586°W, and 3,523 meters above sea level (MSL) west of the study area. According to Lan et al. (2022), GML calculates monthly means after first extracting weekly values from a smoothing curve

applied to all valid data using the method described by Thoning et al. (1989).

We also acquired vertical profile data for CH<sub>4</sub> from aircraft flights from the NASA
 summer 2014 field campaign "Deriving Information on Surface Conditions from Column and

137 Vertically Resolved Observations Relevant to Air Quality" (DISCOVER-AQ) (<u>https://www-</u>

138 <u>air.larc.nasa.gov/missions/discover-aq/P3B-Profiles.co2014.html</u>). With these data we assessed

139 the representativeness of AIRS retrieval concentrations.

140 AIRS CH<sub>4</sub> data were also compared with Colorado Department of Public Health and Environment (CDPHE) median annual ethane data measured at Platteville and the Denver 141 CAMP station (from: https://www.colorado.gov/airquality/tech\_doc\_repository.aspx). The 142 Platteville site is at 40.209°N, 104.824°W, and 1,469 meters MSL. The CAMP station is at 143 39.75°N, 104.99°W, and 1,593 meters. Monitoring at these sites was initiated in late 2011. 144 Three-hour samples were collected with summa canisters from 6-9 AM every six days, and the 145 chemical constituents were analyzed following EPA Compendium Method TO-15 (USEPA, 146 147 1999). Ethane and NMHC concentrations at Platteville are largely from O&G sources (Lyu et al., 2021). Ethane trends at this site are representative of O&G emission trends. The 148 149 representativeness of Platteville monitoring for the O&G fields was tested with the NOAA Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) back trajectories (Stein et 150 al., 2015; Rolph et al., 2017) for 2018. 151

The study area for the trend analysis is presented in Figure 2, and it includes most of the Denver-Boulder metro area and most of the O&G wells in the DJ Basin (the grid cell area will be referred to as the DDJB). The AIRS satellite flies over the DDJB region once every 12 hours. The afternoon overflights occur at approximately 13:30 MST and are referred to as ascending node overflights. We used ascending node data for 2003-2020 for a 1° by 1° grid cell centered at

157 40°N and 105°W. This is a standard grid cell from the NASA AIRS CH<sub>4</sub> product.

158We obtained mean June-August 500 hPa heights from the National Center for159Environmental Prediction (NCEP) Reanalysis (<u>https://psl.noaa.gov/cgi-</u>

- 160 <u>bin/data/timeseries/timeseries1.pl</u>) for a 2.5° by 2.5° grid cell centered on 40°N 105°W, and this
- 161 contains our study area. The NCEP Reanalysis meteorological data product is described by
- Kalnay et al. (1996). We correlated year-to-year changes in ethane and CH<sub>4</sub> enhancements with
- 163 year-to-year changes in 500 hPa heights to see if meteorology plays a role in the trends we
- 164 identified. To provide evidence that the AIRS 700 hPa ascending node CH<sub>4</sub> retrievals for the 165 study area are sensitive to planetary boundary layer (PBL) heights, we calculated the correlation
- study area are sensitive to planetary boundary layer (PBL) heights, we calculated the correlation between monthly mean AIRS data and monthly mean PBL heights from the NASA Modern-Era
- 167 Retrospective analysis for Research and Applications MERRA version 2 (MERRA-2) for 2012-
- 168 2020 (https://giovanni.gsfc.nasa.gov/giovanni/).



Figure 2. Grid cell for the NASA AIRS CH<sub>4</sub> product centered on 40°N and 105°W in northern Colorado. The grid cell includes most of the Denver-Boulder metro area and most of the O&G wells (gray dots) in the southwestern portion of the DJ Basin. The grid cell covers mostly lower elevation terrain (~1,500 meters MSL). The foothills account for a small fraction of the grid cell area.

NASA has averaged AIRS data for nine single-footprint measurements in this grid cell. 175 The grid cell covers much of the Wattenberg field, an O&G field located in the DJ Basin, as well 176 as the Denver-Boulder metro area. The high density of O&G wells in the northeast corner of the 177 AIRS grid cell is associated with the Wattenberg Field. Thermally driven upslope flows prevail 178 179 along Colorado's Front Range at 13:30 MST (the satellite overflight time) during the summer (Toth and Johnson, 1985; Reddy and Pfister, 2016; Pfister et al., 2017; Flocke et al., 2019). 180 These flows will often transport CH<sub>4</sub> westward and southwestward from the O&G fields into and 181 across the grid cell by 13:30 MST. For example, using data available from an Iowa State 182 University website 183 (https://mesonet.agron.iastate.edu/sites/dyn\_windrose.phtml?station=EIK&network=CO\_ASOS) 184

- we calculated that winds at Erie at the southern end of the Wattenberg were calm or from the
   north through east-southeast 71% of the time from 9 to 13 MST from June-August, 2015-2020.
- The AIRS CH<sub>4</sub> uncertainty depends on a variety of factors. Uncertainty for an individual footprint and a given day will be much greater than for monthly or seasonal averages or gridded data sets such as the ones we used that include data from multiple footprints. Kulawik et al.
- 190 (2021) report decreases in AIRS CH<sub>4</sub> errors with increasing averaging times, and for a single

191 footprint uncertainty decreases significantly for seasonal averages. Xiong et al. (2015) report a

bias of 0.27 % and a root mean square error (RMS) of 0.87 % for AIRS CH<sub>4</sub> data for 555 hPa to

193 777 hPa based on intercomparisons with aircraft data and using version 6 of the standard NASA

- AIRS product. Interpreting the findings of Xiong et al. (2015), Kulawik et al. (2021) report a
- standard deviation of 16 ppb for 555 hPa to 777 hPa when compared with aircraft data and a bias

196 of 5 ppb near the mid-troposphere.

We used AIRS monthly average data from 2003 to 2020. We computed June-August averages from these monthly means. We also used the GML monthly averages of weekly flask sample data for NWR to estimate local continental background CH<sub>4</sub> and computed June-August averages from these monthly means. Oltmans et al. (2021) also used NWR data for western continental background estimates in their assessment of CH<sub>4</sub> trends at the BAO tower near Erie, Colorado. They point out that upslope conditions at NWR during July "led to a somewhat elevated median mole fractions that likely overestimate the background concentration".

Butterworth (2011) reports that summer upslope flows at NWR, when they are present, typically begin at 12:00 MST, and upslope persisting for at least two consecutive hours occurs on 34.8% of summer days. All but 7 of the NWR sample days for 2012-2020, the key period of our study, were taken prior to 12:00 MST. We subtracted mean June-August Niwot Ridge (NWR) CH<sub>4</sub> concentrations from the mean June-August AIRS DDJB CH<sub>4</sub> values. The difference represents an estimate of the CH<sub>4</sub> enhancement from local and regional sources.

Unlike aircraft studies in this area which use upwind and downwind flights to quantify 210 CH<sub>4</sub> enhancements (see Petron et al., 2014), our approach may not account for the influences of 211 upwind surface sources on background and the calculated DDJB CH4 enhancements. Both the 212 presence of some DDJB CH<sub>4</sub> transport to NWR in July and the transport of CH<sub>4</sub> into the DDJB 213 from the east could introduce biases of unknown magnitude in our estimates of local 214 215 enhancement. If July NWR overestimates background to some degree, then this could introduce a high bias in the calculated percentages of decline in DDJB CH<sub>4</sub>. If the calculated local 216 enhancement includes a relatively unchanging contribution from sources upwind of the DDJB, 217 then this could introduce a low bias in the calculated percentages of decline in DDJB CH<sub>4</sub>. In 218 addition, there are likely other unknown biases inherent in the approach and methods we have 219 220 used.

We initially chose June-August AIRS data because it coincides with the summer ozone 221 222 season when VOC precursors from the DDJB have the greatest impact on area ozone concentrations but also because the signal for local CH<sub>4</sub> enhancement was positive and stronger 223 for this time of year. Box plots of cycles in monthly NWR CH<sub>4</sub> and AIRS 700 hPa ascending 224 CH<sub>4</sub> are plotted in Figure 3. NWR has a minimum in the summer and a peak in the winter 225 through spring. Tropospheric concentrations in the northern hemisphere can increase in the 226 winter because of a seasonal decline in hydroxyl radical concentrations. The hydroxyl radical 227 228 gradually destroys CH<sub>4</sub> in the summer when photochemistry is more active. This is presumably the cause of winter maxima at this site. 229



Figure 3. Box plots of (a) monthly NWR CH<sub>4</sub> for 2003-2020 and (b) monthly AIRS 700 hPa ascending node CH<sub>4</sub> for 2003-2020, with interquartile ranges and lines connecting medians. The larger signal for AIRS data in the warmer months is attributed to greater PBL heights.

In contrast to NWR, the AIRS 700 hPa ascending CH<sub>4</sub> peaks in July through October. 234 The differences in seasonal cycles between NWR and DDJB AIRS 700 hPa CH<sub>4</sub> are likely the 235 result of seasonal differences in how the PBL concentrates or disperses local CH4 as well as the 236 effects of retrieval constraints on the representativeness of the 700 hPa CH<sub>4</sub> product in the PBL. 237 Kavitha et al. (2018), for example, described a peak AIRS underestimation of measured surface 238 concentrations in the winter when shallow boundary layers concentrated CH<sub>4</sub> near the surface. 239 Rakitin et al. (2015) found lower correlations between AIRS CH<sub>4</sub> and surface measurements at 240 Beijing when mixing heights were below 700 meters AGL, since the satellite spectrometer is less 241 sensitive to CH<sub>4</sub> when it is concentrated in a shallow surface layer. They also noted that 242 correlations between surface and satellite measurements for most of their study sites were higher 243 in the summer when surface mixed layers were deeper. 244

According to maps of ERA5 reanalysis estimates presented by Zhang et al. (2020b), the 245 mean daytime peak PBL height over the plains in the DDJB in the winter is 800-1,000 meters 246 above ground level (AGL), and, in the summer it is greater than 2,400 meters AGL. Lower 247 elevations of the DDJB are at ~ 1,500 meters MSL, and NWR is at 3,523 meters MSL. This 248 249 means that NWR CH<sub>4</sub> is likely representative of the free troposphere in the winter and a wellmixed troposphere with surface influences in the summer. McGrath-Spangler and Denning 250 (2012) estimated mean June-August afternoon PBL heights for North America using the North 251 252 American Regional Reanalysis (NARR) and the NASA MERRA reanalysis. Maps of their results show that summer afternoon PBL heights were ~3,000 to ~3,500 meters AGL over the 253 DDJB. They also calculated mean summer afternoon PBL heights from aerosol backscatter 254 255 measurements made by the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite. These PBL heights were ~2,500 meters AGL over the DDJB. AIRS is 256

primarily sensitive to CH<sub>4</sub> above 2,000 meters MSL (Kulawik et al., 2021). Consequently, it
should be sensitive to CH<sub>4</sub> from surface sources mixed within the deep summer PBL over the
DDJB. The altitude of our study area and resulting deep, summer-afternoon, boundary layers
(with mean mixing heights between ~4,000 to ~5,000 meters MSL or ~650 to ~550 hPa) should

result in an increased sensitivity of AIRS CH<sub>4</sub> to PBL concentrations during the summer months.

In contrast to our data in Figure 3b, Oltmans et al. (2021) show a winter peak and 262 summer minimum in CH<sub>4</sub> at the BAO tower in the Wattenberg Field (Figure S3 in their paper). 263 Their analysis for 2008-2015 shows that the median and 95th percentile values are above 1,900 264 ppb and 2,100 ppb, respectively, during the peak month of February, a month with shallower 265 boundary layers. The February mean at NWR for these years is 1872 ppb, and it is only 1831 ppb 266 for the AIRS 700 hPa product in the DDJB. These high concentrations at BAO are clearly 267 associated with surface sources and emissions concentrated in the boundary layer, and the 268 boundary layer in February is likely too shallow for reasonable representation of surface 269 270 concentrations of CH<sub>4</sub> in the Platte Valley using the AIRS product.

271 To provide additional evidence that the DDJB AIRS 700 hPa ascending CH<sub>4</sub> retrieval is sensitive to PBL heights, we calculated the correlation between monthly AIRS data and monthly 272 PBL heights from the MERRA-2 reanalysis (Global Modeling and Assimilation Office, 2015) 273 for all months from 2012-2020. Describing the MERRA-2 reanalysis product, Gelaro et al. 274 (2017) state that reanalysis "is the process whereby an unchanging data assimilation system is 275 used to provide a consistent reprocessing of meteorological observations... The process relies on 276 277 an underlying forecast model to combine disparate observations in a physically consistent manner, enabling production of gridded datasets for a broad range of variables, including ones 278 that are sparsely or not directly observed." The correlation coefficient between DDJB AIRS CH4 279 and the MERRA-2 PBL heights in the DDJB was 0.46, suggesting that the retrieval signal is 280 stronger when the boundary layer is not shallow. We would expect this correlation to be higher 281 in a comparison using afternoon-only mixed-layer heights. The monthly mean PBL heights (for 282 all hours) in January and July were 540 meters and 1,507 meters AGL, respectively. 283

We did not use descending node AIRS data because local flow regimes are not typically moving DJ Basin emissions into and across the grid cell at night (see Toth and Johnson, 1985), and shallow boundary layers can concentrate CH<sub>4</sub> near the surface. Similarly, we did not use colder season data since the AIRS retrievals might underestimate daytime CH<sub>4</sub> when persistent shallow inversions concentrate CH<sub>4</sub> near the surface.

AIRS CH<sub>4</sub> retrieval concentrations for specific levels of the atmosphere are not independent of concentrations at other levels in the atmosphere and are not a true measure of an ambient concentration at a specific level. In the mid-latitudes of the northern hemisphere, the AIRS CH<sub>4</sub> retrievals at 400 hPa have the highest sensitivity to CH<sub>4</sub> concentrations. The AIRS 400 hPa CH<sub>4</sub> retrieval concentrations also have the greatest independence from CH<sub>4</sub> at other levels in the atmosphere (Xiong et al., 2015). AIRS 400 hPa CH<sub>4</sub>, however, is not as sensitive to surface mixed layer concentrations as retrievals at lower levels of the atmosphere.

The amount that each vertical region of the atmosphere contributes to an AIRS retrieval for a standard level is characterized by a curve called an averaging kernel. In their Figure 2, Kulawik et al. (2021) show the averaging kernels for nominal AIRS altitude levels for a tropical location. The shapes of these curves vary by region, but, in general the 700 hPa product level

300 (which is actually designated as 681 hPa) has greater sensitivity to concentrations from 800 hPA

to 500 hPa than products for higher up in the atmosphere. Averaging kernel sensitivities to near-

302 surface CH4 approach zero with increasing altitudes.

For all these reasons we selected AIRS CH4 700 hPa AIRS product. The AIRS 700 hPa retrievals are representative of a deep layer of the atmosphere and are characterized by the broad averaging kernel. These retrievals are sensitive to near-surface concentrations as well as concentrations in layers extending up to the stratosphere. The summer 700 hPa level data are anticipated to be strongly correlated with concentrations at the surface in the DDJB.

#### 308 **3 Results and Discussion**

The sensitivity of AIRS 700 hPa CH<sub>4</sub> to local near-surface conditions was evaluated 309 using aircraft data collected by the DISCOVER-AQ campaign in July and August of 2014. 310 NASA completed 16 flights with repeated vertical spirals over the DDJB. All 6 DISCOVER-AQ 311 vertical spiral or airport missed-approach sites were in the AIRS DDJB grid cell, except for the 312 Ft Collins spiral site. Roughly 50% of the flight samples from the surface to 3,000 meters were 313 collected between 10:30 and 15:15 MST. Figure 4 presents statistics for the DISCOVER-AQ 314 flight measurements of CH<sub>4</sub> and ethane for 500-meter altitude bins. These represent all flight 315 hours and not just the hour of peak PBL height which is generally 13 MST (Zhang et al., 2020b). 316 The median CH<sub>4</sub> concentration for the bin centered at the 1,750-meter MSL level was ~1,900 317 ppb which is the closest of all the altitude bin medians to the July-August 2014 average AIRS 318 700 hPa CH<sub>4</sub> of 1,886 ppb. This suggests that the AIRS 700 hPa retrieval concentrations were 319 320 sensitive to CH<sub>4</sub> in the mixed layer and can be used to track changes associated with emissions into the boundary layer. 321

The representativeness of NWR CH<sub>4</sub> concentrations for background conditions is further 322 supported by comparison of NWR CH<sub>4</sub> to aircraft CH<sub>4</sub> measurements made during DISCOVER-323 AQ. In Figure 4, CH<sub>4</sub> above 3,000 meters drops to background levels and ethane approaches zero 324 indicating that the 3,000 to 5000-meter level was generally free from strong influences from 325 326 boundary layer emissions from O&G sources. The median CH<sub>4</sub> concentrations measured by DISCOVER-AQ from 3,000 to 5,000 meters MSL are ~1830-1840 ppb, and these are 327 comparable to the NWR July-August 2014 average CH<sub>4</sub> concentration of 1,852 ppb at 3,523 328 meters MSL. This is additional evidence that NWR is a reasonable choice for background CH<sub>4</sub> in 329

our study.

Three-hour HYSPLIT back trajectories were obtained for the highest 20 percentile, 40-60 percentile, and lowest 20 percentile Platteville ethane concentration samples for all of 2018 using

40 km Eta Data Assimilation System (EDAS) meteorological fields

334 (<u>https://www.ready.noaa.gov/edas40.php</u>). These coincided with the 6-9 AM MST sample

335 window for each daily measurement. These show that the Platteville site is representative of

emissions and ambient concentrations in the southern Wattenberg field. Figure 5 presents a map

of the 3-hour back trajectory points for 6-9 AM MST which indicates that Platteville ethane and

338 VOC samples are representative of an area that extends far beyond the local footprint of the

339 monitoring site. This is clear evidence that even under the reduced surface wind speed and

vertical mixing regimes expected in the early morning when samples are collected, the Platteville

341 site is representative of emissions across a wide area of the DJ Basin between Denver and

# 342 Greeley.



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Figure 4. Percentile plots of CH<sub>4</sub> (left) and ethane (right) for all flights and spirals of the
DISCOVER-AQ P3-B in July and August 2014. Blue triangles are medians, orange bars
represent the 25th through 75th percentile range, and horizontal blue lines represent the 5th
through 95th percentile range. The 1.25 km level represents missed runway approaches just
above ground level near Greeley.

We acquired plots of ethane concentrations and winds from the experimental forecasting 349 system that is run by the National Center for Atmospheric Research (NCAR) and based on the 350 351 Weather Research and Forecasting (WRF) model with chemistry (WRF-Chem) (Kumar et al., 2020) (https://www.acom.ucar.edu/firex-aq/forecast.shtml). Plots are for 5:00 MST on 30 352 353 January 2022 and 23 July 2021. These are shown in Figure 6. Elevated ethane concentrations are largely from O&G sources. Toth and Johnson (1985) and Neff (1997) describe the features of 354 early morning slope and drainage flows as they move eastward from the foothills west of Denver 355 and northward from the Palmer Divide which is south of Denver and converge as southerly 356 drainage flows along the Platte River Valley from Denver to Platteville. Drainage winds 357 eventually curve eastward following the river valley east of Greeley. These conditions are typical 358 359 when stronger synoptic-scale winds are not present.

Modeled ethane concentrations and winds capture these flows and their effects on ethane and show that ethane pools in the area surrounding Platteville and the O&G field. In addition, Pfister et al. (2017) show average modeled O&G tracer concentrations for 6:00-12:00 MST during the 2014 Front Range Air Pollution and Photochemistry Éxperiment in Figure 8 of their paper. The O&G tracer plot places Platteville in the center of a large area with fairly uniform high concentrations, and the spatial distribution is comparable to the fairly uniform high-

- 366 concentration footprints in Figure 6. Platteville is suitably located to sample ethane and other
- 367 O&G trace gases within the O&G field during the morning hours.



- 369 Figure 5. HYSPLIT three-hour back trajectory points (shown as red dots) for 6-9 AM MST for
- the CDPHE Platteville monitor in 2018. Oil and gas wells are plotted as grey dots.



Figure 6. NCAR WRF-Chem surface wind and ethane concentration forecasts for (a) 5:00 MST
30 January 2022 and (b) 5:00 MST 23 July 2021. These show the effects of typical morning
slope and drainage flows that pool ethane near Platteville and transport ethane eastward near

- Greeley following the curve in the Platte River valley. The Platteville location and label have
- been added.

Annual ethane means, medians, and statistics for 2012-2020 are presented for Platteville 377 and Denver CAMP in Tables 1 and 2, respectively. We used annual data for increased sample 378 sizes and because summer data for 2016 were limited. Table 3 lists the June-August mean DDJB 379 AIRS 700 hPa CH<sub>4</sub>, the number of AIRS retrievals, the June-August NWR CH<sub>4</sub>, the calculated 380 DDJB enhancement in CH<sub>4</sub>, and Platteville annual median ethane. We used the median ethane 381 data in this case to reduce the influence of high values associated with shallow surface mixed 382 layers. Table 3 shows that the estimated DDJB CH<sub>4</sub> enhancement decreased by 55% between 383 2013 and 2020. Platteville median ethane decreased by 73% during the same period. The choice 384

of 2013 as the starting year was based on the peak enhancement in 2013. Since there is the

- possibility of influences from unexplained cofactors each year, the true enhancement peak might
- have occurred in either 2012 or 2013. Later we show a slightly more conservative estimate for CH<sub>4</sub> based on a linear regression for 2012-2020, and this reduction is 56%. Similarly, the
- CH<sub>4</sub> based on a linear regression for 2012-2020, and this reduction is 56%. Similarly, the reduction in median Platteville ethane based on the linear regression for 2012-2020 was 77%.

Year	Mean ethane (ppb)	Median ethane (ppb)	Standard deviation (ppb)	95% confidence level (ppb)	Number of samples
2012	213.9	196	155.4	32.4	91
2013	277.4	212	250.5	70.5	51
2014	187.5	132	222.0	59.4	56
2015	155.0	143	108.9	31.6	48
2016	155.3	119	117.5	30.9	58
2017	177.3	79.6	358.4	91.8	61
2018	100.7	87.2	77.8	19.9	61
2019	97.5	65.3	90.8	23.5	60
2020	81.2	57.3	62.4	16.9	55

**Table 1.** Platteville Ethane Annual Means, Medians, and Statistics for 2012-2020.

# **Table 2.** Denver CAMP Ethane Annual Means, Medians, and Statistics for 2012-2020.

Year	Mean ethane (ppb)	Median ethane (ppb)	Standard deviation (ppb)	95% Confidence level (ppb)	Number of samples
2012	23.5	18.6	15.3	3.2	88
2013	29.5	20.9	23.4	6.2	57
2014	24.3	18.5	19.0	5.0	58
2015	27.8	21.3	18.3	5.2	50
2016	34.9	23.8	27.0	7.0	60
2017	28.0	23.5	19.1	4.9	61
2018	32.2	24.0	24.5	6.3	61
2019	32.5	23.0	27.8	7.1	61
2020	33.6	23.6	24.4	7.8	40

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Table 3. AIRS DDJB CH<sub>4</sub>, NWR CH<sub>4</sub>, DDJB Enhancement, and Platteville Ethane for 2003 2020.

Voar	Mean June-August AIRS CH₄	Number of AIRS retrievals June-	Mean June-August Niwot Ridge CH₄	DDJB CH₄ minus Niwot Ridge CH₄ (ppb)	Platteville annual median
2003	1829.7	76	1814.1	15.6	ethane (ppb)
2004	1840.3	73	1805.9	34.4	
2005	1841.6	72	1819.8	21.7	
2006	1841.6	76	1805.6	36.0	
2007	1848.8	71	1813.3	35.4	
2008	1854.9	73	1826.4	28.5	
2009	1864.2	78	1826.6	37.6	
2010	1860.1	68	1834.9	25.2	
2011	1871.5	68	1839.5	32.0	
2012	1877.8	71	1839.0	38.8	196
2013	1881.4	74	1841.6	39.8	212
2014	1884.0	76	1853.6	30.4	132
2015	1896.0	75	1860.4	35.6	143
2016	1904.3	71	1872.4	31.9	119
2017	1907.8	72	1882.7	25.1	79.6
2018	1920.0	67	1896.8	23.2	87.2
2019	1916.9	75	1897.6	19.3	65.3
2020	1927.8	60	1910.1	17.8	57.3
Percent Reduction 2013 to 2020				55%	73%

Peischl et al. (2018) and Petron et al. (2014) conclude that O&G sources account for 75% of local CH<sub>4</sub> sources. Consequently this 55-56% reduction may underestimate the percent reduction in CH<sub>4</sub> from O&G sources. It is reasonable that the percent reduction in ethane was greater than the percent reduction in CH<sub>4</sub>, since O&G sources account for almost all the ethane at Platteville.

In addition, CDPHE estimates that O&G systems accounted for 59% of CH<sub>4</sub> statewide in 401 2019 (CDPHE, 2021, Exhibit 1-5). While emissions estimates are not specific to the DDBJ grid 402 403 cell, only the statewide O&G sector is estimated to have meaningful emissions reductions from 2010 to 2019 (CDPHE, 2021). All other sectors have CH<sub>4</sub> emissions changes that are either 404 negligible (e.g., residential, commercial, industrial, transportation, and electric power), slightly 405 increasing (e.g., agriculture and waste management), or are not emitting CH<sub>4</sub> in the DDJB grid 406 cell (e.g., coal mining). Consequently, it is reasonable to assume that the trends in the DDJB CH<sub>4</sub> 407 enhancement are predominantly affected by changes within the O&G sector. 408

Figure 7 shows the estimated long-term trends in DDJB CH<sub>4</sub> enhancement based on AIRS measurements in the DDJB grid cell. NWR flask samples are collected weekly for a total of ~13 samples for June-August, while an average of 72 AIRS retrievals are available for each

June-August period. In many cases the sample and retrieval days do not match, and the means of

- 413 each dataset can represent different conditions of continental background and transport for
- 414 Colorado. This could introduce noise in the DDJB CH<sub>4</sub> enhancement time series. Consequently,
- a four-point median smoother and a LOESS curve (Cleveland, 1979) have been fitted to the data
- 416 to show the general trends throughout the entire period.



Figure 7. DDJB June-August CH<sub>4</sub> enhancement from 2003 through 2020 fitted with a 4-point median smoother (red) and a LOESS curve with a 0.4 smoothing parameter (green).

420 A gradual increase which may be associated with growth in O&G extraction activities is followed by a steep downward trend after 2013, and we attribute the decline to DJ Basin O&G 421 emissions reductions. A linear regression through this data for 2003-2012 yields a statistically 422 insignificant trend of 1.25 +/- 1.8 ppb. Oltmans et al. (2021) found no significant trend in BAO 423 CH<sub>4</sub> near Erie in the southwest portion of the Wattenberg from 2008 through mid-2016. We also 424 found no significant trend for the DDJB AIRS enhancement from 2008 through 2016. A plot of 425 426 our results for this period is presented in Figure 8(a). When we consider 2012-2020 (see Figure 8(b)), however, we find that there is a nearly linear and statistically significant downward trend 427 with an  $R^2$  of 0.90. CH<sub>4</sub> declined in the DDJB grid cell at a rate of -2.84 +/- 0.8 ppb (based on 428 the 95% confidence interval) per year. In addition, a linear regression of the ratio of DDJB CH<sub>4</sub> 429 to NWR CH<sub>4</sub> against year (which is independent of local enhancement estimates) shows a 430 decline from 1.022 to 1.008 during this period ( $R^2$  of 0.91). 431

The trends in 2012-2020 annual median Platteville and Denver CAMP ethane (see Figure 9) show a statistically significant linear decrease in Platteville ethane of -19.1 ppb +/- 5.9 ppb per year (based on the 95% confidence interval, R<sup>2</sup> of 0.90) and a statistically significant linear increase in Denver CAMP ethane of 0.7 ppb +/- 0.4 ppb per year (based on the 95% confidence interval, R<sup>2</sup> of 0.68). Lyu et al. (2021) report a 33%-56% contribution from O&G sources to 6-9 AM MST Denver CAMP NMHC. Ortega et al. (2021) found a positive trend of 0.9 ppb  $\pm$  0.3 % per year for ethane north of Boulder from 2010-2019. Denver and Boulder ethane have been

- 439 increasing at a similar rate, although the rate for the former is based on morning-only
- 440 measurements.

Rossabi et al. (2021) analyzed west-east gradients in  $CH_4$  at several sites for 9 to 11 441 three-day periods during July and August of 2014. They found a high Pearson correlation 442 coefficient of 0.77 between CH<sub>4</sub> and ethane in eastern Boulder County and concluded that it is 443 highly likely that natural gas extraction activities are the dominant source of ethane in the region. 444 Their sites extended from the higher terrain west of Boulder, across Boulder, and into the 445 southwestern edge of the Wattenberg Field. Figure 3 of their study shows that CH4 at the 446 Boulder County Public Health site in Boulder was ~ 6 ppb higher than estimated background. 447 Their Figure 3 also shows that CH<sub>4</sub> at two sites in or near the southwestern edge of the 448 Wattenberg Field was ~12 ppb higher than background. Their results, other work we have cited, 449 and our results for ethane at Platteville and Denver show steep spatial gradients in O&G impacts 450 across the region. Nominal increases in ethane at Boulder and Denver are not necessarily 451 inconsistent with steep declines at Platteville, which is in the center of the Wattenberg Field; and 452 we have concluded that the Platteville site is representative of the Wattenberg, based on 453



454 HYSPLIT back trajectories and cited studies.

455

**Figure 8.** DDJB June-August CH<sub>4</sub> enhancement versus year with linear regression and 95%

457 confidence limits for (a) 2008-2016 and (b) 2012-2020. No statistically significant trend was
458 found for CH<sub>4</sub> from 2008-2016 at BAO (Oltmans et al., 2021) or in our analyses for the DDJB,

459 but a robust downward trend is evident for the DDJB from 2012-2020.



Figure 9. Linear regression and 95% confidence limits for l (a) annual median Platteville ethane
 and (b) annual median Denver CAMP ethane, versus year for 2012-2020. Trends for both sites
 are statistically significant.

A regression between the June-August DDJB  $CH_4$  enhancement and annual median Platteville ethane are plotted in Figure 10. The regression is statistically significant and has an  $R^2$ of 0.93, and the Pearson correlation coefficient is 0.96. If Platteville ethane which is largely from O&G sources were reduced to zero, the regression in Figure 10 would yield a remaining  $CH_4$ enhancement of 12 ppb or 30% of the 2012 regression value in Figure 8(b). This is comparable to the 25% local contribution from non-O&G sources in 2012 estimated by Petron et al. (2014).

Figure 11 shows the time series of DDJB CH<sub>4</sub> enhancement and Platteville ethane and the 470 year-to-year changes in each. Results in Figures 10 and 11 provide compelling evidence that the 471 calculated DDJB CH<sub>4</sub> enhancement is sensitive to and tracks with surface boundary layer 472 473 concentrations. These results also show that DDJB CH<sub>4</sub> has been responsive to reductions in O&G emissions within the grid cell. Recall that Rossabi et al. (2021) concluded that it is highly 474 likely that natural gas extraction activities are the dominant source of ethane in the region. In 475 addition, using positive matrix factorization and chemical mass balance methods, Lyu et al. 476 (2021) found that O&G activities accounted for 92%-96% of NMHC at Platteville for 2013-477 2016. The strong correlation and the fact that there have not been substantial changes in ethane 478 in Denver or Boulder suggest that reductions in O&G emissions in the Wattenberg are driving 479 the observed declines in AIRS CH<sub>4</sub> within the DDJB. The correlation between DDJB CH<sub>4</sub> 480 enhancement and median Denver ethane is -0.66. 481



- **Figure 10.** Linear regression between June-August mean 2012-2020 DDJB CH<sub>4</sub> enhancement
- and annual median Platteville ethane (with plotted 95% confidence limits). The fit is robust with an  $R^2$  of 0.93.



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Figure 11. Trends in June-August DDJB CH<sub>4</sub> enhancement and annual median Platteville ethane

in ppb. Panel (a) illustrates June-August mean 2012-2020 DDJB CH<sub>4</sub> enhancement in red with

489 open circles and annual median Platteville ethane shown with blue bars (with plotted regression

lines) and panel (b) shows year-to-year changes in these, with CH<sub>4</sub> in red with open circles and

491 ethane in blue with open squares.

Feng et al. (2022) find that interannual variability in meteorology (especially wind 492 speeds) plays a significant role in trends in CH<sub>4</sub> in the US. Reddy and Pfister (2016) 493 demonstrated that maximum surface ozone concentrations in and near the DDJB are highly 494 correlated with 500 hPa heights. They state that "upper level ridges decrease winds and allow 495 cyclic terrain-driven circulations to reduce transport away from sources". We obtained mean 496 June-August 500 hPa heights from the NCEP Reanalysis for a grid cell centered on 40°N 497 105°W, and this contains the DDJB. The correlation coefficients for interannual differences in 498 Platteville ethane, DDJB CH<sub>4</sub> enhancement, and NWR CH<sub>4</sub> versus the interannual differences in 499 500 hPa heights were calculated for 2013-2020; and these are 0.69, 0.74, and 0.26 for Platteville, 500 the DDJB, and NWR, respectively. Even though we used annual data for ethane, it appears that 501 the impacts of summer meteorology are strong enough to have a significant effect on variations 502 in yearly values. The correlation is very high for DDJB CH4 enhancement and low for NWR 503 CH<sub>4</sub>. While reductions of emissions in the DDJB are likely responsible for the overall trends, the 504 more subtle variations in Figure 11 are probably caused, at least in part, by year-to-year changes 505 in meteorology. The strong correlations with weather for both ethane and CH<sub>4</sub> enhancement 506 support the hypothesis that the AIRS product is detecting real changes in year-to-year CH<sub>4</sub> 507 within the PBL. 508

Oltmans et al. (2021) estimated background propane, a tracer for O&G emissions, at the 509 BAO tower near Erie in the DJ Basin and subtracted this from ambient data. Using only data 510 from 2008-2015 and scaling the annual changes in local contribution to 2011 data, they projected 511 512 a -1.5% change per year in propane through 2021 and compared this with the RAQC's projected emissions inventory reductions of -6.5% per year. We have not estimated background ethane, 513 also an O&G tracer, at the Platteville monitor, but we used the regression statistics for median 514 Platteville ethane versus year and scaled the -19.1ppb slope by the 2012 regression value of 515 197.8 ppb to obtain an average annual decline of -9.7% from 2012 through 2020 (with a range of 516 -12.6% to -6.7%, based on the lower and upper 95% confidence intervals for the slope). 517

518 This rate of decline is not based on data from an earlier period but on an analysis of data 519 through 2020, and the rate of decline of the emissions inventory is just above the upper limit of 520 the calculated range for 2012-2020 at Platteville. Trends in both Platteville ethane and the DDJB 521 CH<sub>4</sub> enhancement provide evidence that the regulatory requirements reflected in the emissions 522 inventories are having a significant effect.

The focus of this paper is CH<sub>4</sub> trends within the DDJB, but to further verify our method 523 and the ability of the AIRS 700 hPa CH<sub>4</sub> product to detect warm-season trends in the high-524 altitude western US, we completed a preliminary analysis of trends in CH<sub>4</sub> enhancements for the 525 Uintah Basin O&G fields in Utah, a major O&G production region in the western US. The floor 526 of the basin is at ~1,500 meters MSL, and summer mean peak PBL heights are comparable to 527 those in the DDJB (McGrath-Spangler and Denning, 2012; Zhang et al., 2020b). We used the 528 standard AIRS version 7 product for the two AIRS grid cells that contain the basin (see 529 supplementary Figure S1). We calculated June-August mean AIRS CH<sub>4</sub> and subtracted the mean 530 June-August NWR CH<sub>4</sub> data calculated for our study. 531

The resulting trend for the Uintah Basin CH<sub>4</sub> enhancement for 2014-2020 is plotted in Figure 12. Lin et al. (2021) show that basin natural gas production increased rapidly, peaking in 2012 and 2013, and then declined rapidly. The number of producing gas wells peaked in 2015

- and then declined rapidly. They also show that oil production and the number of oil wells peaked
- in 2014-2015, and then declined unevenly after this. They found that decreases in CH<sub>4</sub> after 2015
   were related to declining O&G production and not to a reduction in CH<sub>4</sub> leaks. We show a
- statistically significant linear decrease of -2.77 ppb +/- 2.47 ppb ( $R^2$  of 0.62) from 2014-2020.
- 520 Linet al. (2021) a local de la collega de la collega de la lifference de lifference de la terres
- 539 Lin et al. (2021) calculated basin  $CH_4$  enhancement by taking the difference between
- 540 surface concentrations at a site in the center of the O&G field and one on the far western edge of 541 the basin. For a trend analysis with linear regression, they used afternoon-only concentrations for
- 4 April-September to avoid cold pooling events that concentrate CH<sub>4</sub> in shallow surface layers.
- Using monthly means, they found a decreasing trend of -4.63 ppb with an R<sup>2</sup> of 0.241 for 2015-
- 544 2020. Their enhancements (from the regression line) range between  $\sim 20$  ppb to  $\sim 45$  ppb, whereas
- the AIRS-based summer season enhancements are from 7 ppb to 29 ppb.
- 546 Using a combination of measurements and modeling, Lin et al. (2021) report that
- emissions decreased by approximately 50% during this period, and we calculate a 60% reduction
- in CH<sub>4</sub>. This percentage was derived from the regression values for 2015 and 2020. There is
- reasonable agreement between the AIRS-based results and theirs. This is further evidence that
- the AIRS 700 hPa CH<sub>4</sub> product can be used to detect changes consistent with surface
- concentrations in and near a major O&G production area for periods and locations with deep
- 552 vertical mixing.



- 554 Figure 12. AIRS 700 hPa CH<sub>4</sub> enhancement for the Uintah Basin in Utah with a linear
- regression and 95% confidence limits for 2014-2020. The fit is statistically significant with an  $R^2$  of 0.62.

#### 558 4 Conclusions

We selected a one-degree NASA AIRS grid cell centered at 40°N and 105°W in northern 559 Colorado to assess changes in CH<sub>4</sub> from 2003 through 2020. This grid cell includes Denver, 560 Boulder, and Greeley, Colorado, and much of the Wattenberg O&G field in the Denver 561 Julesburg O&G basin. O&G emissions from this area are a significant source of CH<sub>4</sub>, ethane, and 562 VOC precursors to ozone. Colorado implemented regulations to significantly reduce emissions 563 from O&G sources in early 2014 and has implemented additional emissions control requirements 564 since (Colorado Regulation No. 3, No. 7, and No. 22, https://cdphe.colorado.gov/aqcc-565 regulations). Our study was designed to use AIRS CH<sub>4</sub> to assess the effects of these controls (and 566 operational changes implemented by the industry voluntarily) on ambient CH<sub>4</sub> and to determine 567 whether this approach could be used to track the effectiveness of emissions controls. Surface 568 measurements alone have limited spatial coverage compared with satellite instruments. Aircraft 569 studies are typically limited in temporal coverage. Newer and more advanced satellite 570 instruments such as TROPOMI do not provide data prior to about 2016-2017. Our study shows 571 that AIRS data can serve as a bridge to more robust analyses based on TROPOMI and other 572 satellite instruments that will soon become operational. 573

We computed mean June-August ascending node AIRS 700 hPa CH<sub>4</sub> for each year and subtracted June-August mean NWR CH<sub>4</sub>. NWR is a high-altitude surface monitoring site immediately west of our grid cell. The differences represent the enhancement over background within the grid cell. Vertical profiles of CH<sub>4</sub> from aircraft flights within the grid cell during the July-August 2014 DISCOVER-AQ field campaign were used to verify the sensitivity of the 700 hPa AIRS product to near-surface concentrations and to confirm that the Niwot Ridge site is a reasonable choice for background concentrations.

We find 55% and 73% reductions in DDJB  $CH_4$  enhancement and Platteville ethane, 581 582 respectively, between 2013 and 2020 and a 56% reduction in DDJB CH<sub>4</sub> enhancement using a linear regression for 2012-2020. These significant reductions occurred even though oil 583 production increased by 343% and gas production by 297% from 2012-2020. The reduction in 584 CH<sub>4</sub> is comparable to the projected 57% reduction in O&G VOC emissions for 2011-2020 from 585 the 2020 draft ozone State Implementation Plan for the ozone nonattainment area that includes 586 the DDJB (RAQC and CDPHE, 2020). We are unable to calculate biases or uncertainties for the 587 55-56% CH<sub>4</sub> reduction, but our estimates of CH<sub>4</sub> enhancements for the Uintah basin suggest that 588 biases inherent in this approach may be minimal. 589

Smoothing curves applied to the grid cell enhancement show a gradual increase in CH<sub>4</sub> 590 591 through 2013 followed by a noticeable decline. A linear regression of grid cell enhancement versus year shows a 2.84 ppb +/- 0.8 ppb decrease per year from 2012 through 2020 with an  $\mathbb{R}^2$ 592 value of 0.90. Grid cell enhancement is strongly correlated with Platteville ethane. Platteville is 593 in the center of the O&G fields. Using HYSPLIT back trajectories for 2018, we demonstrated 594 595 that this site is representative of a large area of the Wattenberg field, a major source region for CH<sub>4</sub>. The correlation coefficient between grid cell CH<sub>4</sub> enhancement and Platteville ethane was 596 0.96, and the  $R^2$  for a linear regression between these was 0.93. Platteville ethane dropped from 597 196.0 ppb in 2012 to 57.3 ppb in 2020. Based on a linear regression, we calculate an annual rate 598 of change of -9.7% relative to 2012 (with a range of -12.6% to -6.7%). Denver median ethane 599

concentrations ranged from 18.5 to 24.0 ppb during the period with a slight increasing trend of
 0.7 ppb +/- 0.4 ppb per year.

A preliminary analysis of trends in CH<sub>4</sub> enhancement for the Uintah Basin O&G production area based on AIRS 700 hPa CH<sub>4</sub> and NWR data compares well with the decline of surface CH<sub>4</sub> described by Lin et al. (2021). This is additional evidence that the AIRS product can detect changes directly related to surface concentrations for periods and locations with deep vertical mixing.

The grid cell CH<sub>4</sub> enhancement should be considered an estimate that is representative of 607 608 and proportional to true enhancement above background within the boundary layer. The strong correlation between grid cell CH<sub>4</sub> enhancement and Platteville ethane and the relative stability of 609 Denver and Boulder ethane concentrations over time suggest that reductions in O&G emissions 610 are driving the reductions in CH<sub>4</sub> that are evident in the AIRS CH<sub>4</sub> product. We conclude that 611 AIRS CH<sub>4</sub> can be used to measure the efficacy of O&G emissions control programs in this 612 region and that these controls, as well as other voluntary design changes such as tankless systems 613 614 and process centralization, are having an effect. Further intercomparisons between AIRS and TROPOMI CH<sub>4</sub> trends for the years of overlapping records would help scientists quantify 615 percent reductions in CH<sub>4</sub> in recent years and understand the biases and uncertainties in these 616 estimates. A comprehensive picture of the effects of emissions controls will require continued 617 analyses of surface, aircraft, and satellite measurements for this region. Evidence and results 618 presented here also suggest that the AIRS CH<sub>4</sub> data could be useful in tracking progress in 619 620 achieving DDJB greenhouse gas emissions reductions.

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634

### 635 **Open Research**

- 636
- The primary datasets used in this study include AIRS and MERRA-2 reanalysis data from the
- 638 NASA Giovanni website, NCEP Reanalysis data from the NOAA Physical Sciences Laboratory,
- ethane monitoring data from the CDPHE Air Pollution Control Division, and monthly CH<sub>4</sub> data
- for Niwot Ridge from the NOAA Global Monitoring Laboratory. Key datasets are available in a
- data repository (<u>https://doi.org/10.5281/zenodo.7038756</u>). Statistical calculations were

completed with NCSS 8.0 and Microsoft Excel. Graphs, plots, and maps were prepared with 642 NCSS 8.0, Surfer 12, Excel, and Igor Pro.

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850	Journal of Geophysical Research: Atmospheres
851	Supporting Information for
852	Downward Trend in Methane Detected in a Northern Colorado Oil and Gas
853	Production Region Using AIRS Satellite Data
854	P. J. Reddy <sup>1</sup> , C. Taylor <sup>2</sup>
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856	<sup>2</sup> Ramboll US Consulting, Fort Collins, Colorado
857	
858	Contents of this file
859	
860	Text S1
861	Figure S1
862	Introduction
863	This document contains supplementary figure S1 which shows the location of the

This document contains supplementary figure S1 which shows the location of the standard AIRS version 7 product grid cells that contain the Uintah Basin oil and gas production region in Utah.

### 866 S1. The AIRS Version 7 Grid Cells Over the Uintah Basin

AIRS Version 7 grid cells were obtained from the NASA Giovanni website

868 (https://giovanni.gsfc.nasa.gov/giovanni/). Details of this product can be found here:

869 https://disc.gsfc.nasa.gov/datasets/AIRS3STM\_7.0/summary. The two cells are 1° by 1° in size

and have been joined in Figure S1. One grid cell is centered on 40°N, 110°W, and the other is

871 centered on 40°N, 109°W.



Figure S1. Merged AIRS grid cells covering the Uintah Basin in Utah. The map source is GoogleEarth.