



# Quantification of Methane Emissions from Discrete Sources

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April 23, 2024

# Quantification of Methane Emissions from Discrete Sources

A DISSERTATION PRESENTED  
BY  
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TO  
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FOR THE DEGREE OF  
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IN THE SUBJECT OF  
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## Quantification of Methane Emissions from Discrete Sources

### ABSTRACT

Various point source methane quantification approaches enable comprehensive estimation of methane emissions, which is essential for effective methane monitoring and mitigating strategies. This study uses remote sensing data from projects like MethaneAIR and MethaneSAT to develop robust quantification methods for methane emissions. We explore techniques such as modified integrated mass enhancement (mIME), divergence integral (DI), ratio method, and Geostatistical Inverse Model (GIM) approach, considering factors like the nature of the sources, their size, the background noise, and topography.

In the first chapter, we aim to develop rigorous point source quantification approaches for MethaneAIR and MethaneSAT satellites. We explore different techniques to quantify methane emissions from discrete sources and devise a method based on large eddy simulations. Although initially tested on high-resolution Chinese satellites due to the early stage of the MethaneAIR and MethaneSAT projects, these algorithms pave the way for faster development of methods tailored to our projects.

In the second chapter, we apply our quantification methods to controlled release experiments and evaluate their performance on MethaneAIR data. Our findings show good agreement between estimated and released emissions as well as mIME and DI quantification methods. We develop a decision tree to determine the most suitable method for specific scenarios, enhancing our understanding of method limitations and informing the development of MethaneSAT algorithms.

The final chapter focuses on a detection limit analysis of MethaneAIR to evaluate MethaneSAT's potential performance. Using large eddy simulations, we estimate MethaneSAT's detection limit as a function of noise across different resolutions. Additionally, we demonstrated successful point source estimation based on the GIM, bridging the gap between point source and area source quantification techniques. The findings better prepare us for MethaneSAT data analysis.

In conclusion, this study lays the foundation for methane point source quantification for the MethaneAIR and MethaneSAT projects. The lessons learned extend beyond our projects; they can allow the appropriate selection of methane emission quantification approaches for various scenarios.

# Contents

|                                                              |      |
|--------------------------------------------------------------|------|
| TITLE PAGE                                                   | i    |
| COPYRIGHT                                                    | ii   |
| ABSTRACT                                                     | iii  |
| TABLE OF CONTENTS                                            | iv   |
| LIST OF FIGURES                                              | vii  |
| LIST OF TABLES                                               | xiv  |
| DEDICATION                                                   | xv   |
| ACKNOWLEDGMENTS                                              | xvii |
| o INTRODUCTION                                               | i    |
| 1 METHANE QUANTIFICATION METHODS                             | 5    |
| 1.1 Introduction . . . . .                                   | 6    |
| 1.2 Methods . . . . .                                        | 8    |
| 1.3 Results for GF5 and ZY1 Satellites . . . . .             | 13   |
| 1.4 Conclusion . . . . .                                     | 16   |
| 2 CONTROLLED RELEASE EXPERIMENTS                             | 17   |
| 2.1 Introduction . . . . .                                   | 18   |
| 2.2 Data . . . . .                                           | 19   |
| 2.2.1 MethaneAIR Research Flights . . . . .                  | 19   |
| 2.2.2 MethaneAIR Data Retrievals . . . . .                   | 22   |
| 2.3 Methods . . . . .                                        | 23   |
| 2.3.1 Methane Point Source Emission Quantification . . . . . | 23   |
| 2.3.2 Wind Validation . . . . .                              | 27   |

|       |                                                                           |           |
|-------|---------------------------------------------------------------------------|-----------|
| 2.3.3 | Error Analysis . . . . .                                                  | 28        |
| 2.3.4 | Targeted Emissions Sources . . . . .                                      | 29        |
| 2.4   | Results and Discussion . . . . .                                          | 32        |
| 2.4.1 | Blinded Volume-Controlled Release Experiment (2021) . . . . .             | 32        |
| 2.4.2 | Blinded Volume-Controlled Release Experiment (2022) . . . . .             | 36        |
| 2.4.3 | Confidence Levels of the Emission Estimates . . . . .                     | 38        |
| 2.5   | Conclusions . . . . .                                                     | 40        |
| 3     | <b>OBSERVING SYSTEM SIMULATION EXPERIMENTS</b>                            | <b>42</b> |
| 3.1   | Introduction . . . . .                                                    | 43        |
| 3.2   | Methods . . . . .                                                         | 44        |
| 3.2.1 | Scenes of interest for the OSSE . . . . .                                 | 45        |
| 3.2.2 | The Quantification Limits . . . . .                                       | 47        |
| 3.2.3 | Prototypes of the GIM framework . . . . .                                 | 48        |
| 3.2.4 | Emission Estimates from the GIM . . . . .                                 | 51        |
| 3.3   | Results . . . . .                                                         | 53        |
| 3.3.1 | MethaneAIR Observations . . . . .                                         | 53        |
| 3.3.2 | Reprojection of MethaneAIR onto the MethaneSAT Grid . . . . .             | 57        |
| 3.3.3 | The Quantification Limits Based on Critical Noise Levels (OSSE) . . . . . | 61        |
| 3.3.4 | The GIM Outputs . . . . .                                                 | 64        |
| 3.4   | Conclusions . . . . .                                                     | 69        |
| 4     | <b>CONCLUSIONS</b>                                                        | <b>71</b> |
|       | <b>APPENDIX A ADDITIONAL INFORMATION</b>                                  | <b>73</b> |
| A.1   | Microisotropy of the Anadarko Basin . . . . .                             | 73        |
| A.1.1 | The Story . . . . .                                                       | 73        |
| A.1.2 | The Draft . . . . .                                                       | 74        |
| A.2   | WRF Benchmarking . . . . .                                                | 76        |
| A.3   | Calculations . . . . .                                                    | 76        |
| A.3.1 | mIME Effective Wind Speeds . . . . .                                      | 76        |
| A.3.2 | Divergence Integral . . . . .                                             | 77        |
| A.3.3 | Gridding by Nearest Neighbor and Physics-Based Methods . . . . .          | 80        |
| A.3.4 | York Regression . . . . .                                                 | 80        |
| A.3.5 | Mean Percentage Error . . . . .                                           | 80        |
| A.4   | Additional Results . . . . .                                              | 81        |
| A.4.1 | Miscellaneous Methane Sources . . . . .                                   | 81        |
| A.4.2 | MethaneAIR and MethaneSAT comparison . . . . .                            | 82        |
| A.4.3 | Blinded Volume-Controlled Release Experiments . . . . .                   | 86        |
| A.4.4 | Unlit Flare . . . . .                                                     | 97        |
| A.4.5 | MVD & Pipeline . . . . .                                                  | 99        |

|       |                                                                        |     |
|-------|------------------------------------------------------------------------|-----|
| A.5   | Analysis Methods . . . . .                                             | 101 |
| A.5.1 | Wind Analysis . . . . .                                                | 101 |
| A.5.2 | Wind Product Analysis . . . . .                                        | 102 |
| A.5.3 | Threshold Analysis . . . . .                                           | 103 |
| A.5.4 | Decision Tree . . . . .                                                | 103 |
| A.5.5 | Additional Attempts to Determine Contamination Near the Methane Plumes | 106 |
| A.6   | Cropped Blinded-Volume controlled release Plumes . . . . .             | 107 |
| A.7   | MethaneAIR flight tracks . . . . .                                     | 113 |

|            |     |
|------------|-----|
| REFERENCES | 121 |
|------------|-----|

# Listing of figures

|     |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |    |
|-----|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 1.1 | Observed (left) and modeled (right) plumes of (a) plume k on August 2, 2020, and (b) plume x on November 1, 2019. The modeled plume was generated by using WRF-Chem-LES with topography and one-way coupling to full HRRR meteorology. We used the "contourLines" function in R to identify the enclosing area with a detection threshold of $\Delta XCH_4$ (threshold = 0.045, ppm, i.e. 2.5% of total $XCH_4$ ), defining the dashed black line in the main image (red box). The selected plume is in the solid white box at the bottom right corner. The red sun-cross symbols indicate the source locations. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | 14 |
| 2.1 | Examples of the MethaneAIR data on 6 August 2021 (a) MiVida Gas Processing Plant (pink triangle) from RFo6 segment 10 (approximately 2km x 2km image) and (b) mosaic image (75 km x 120 km) of the entire RFo6 scene from all the segments including the segment shown in Figure 2.1. The prominent gap areas in this picture were not imaged during the overlapping turns of the pattern. The white dashed line indicates the approximate wind direction at the processing plant site. Full flight track information can be found in A.7. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | 21 |
| 2.2 | <b>a:</b> MethaneAIR image of a blinded volume-controlled release (orange square) and an unlit flare (pink triangle) observed on 3 August 2021. The white dashed line indicates the approximate wind direction at the unlit flare site. <b>b:</b> A sample of the isolated unlit flare plume using the mIME method. The isolated plume is superimposed on a ©Google Map 2022 satellite layer. <b>c:</b> Three example rectangles are used to calculate the flux divergence. <b>d:</b> Calculated flux divergence as a function of distance from the source to the downwind edge of the rectangle. Circles indicate the position of the example rectangles are shown in Figure 2.2c. The fluctuations of the apparent flux with distance from the source reflect the influence of eddy scale motion as well as contributions of excess methane from nearby sources. The influence of eddy-scale motions is evident in the oscillation at approximately 250 m intervals, the apparent length scale for eddies at this overpass. The surface flux is estimated by averaging the flux divergence over several eddy scales to average out $\partial m / \partial t$ . In this example, we averaged the DI from 80 – 700m to avoid influence from other sources nearby that increase the DI beyond 700m from the source. . . . . | 26 |

|     |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |    |
|-----|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 2.3 | Volume-controlled release experiment results after unblinding. The results presented in this figure are based entirely on the mIME method. The black circles represent the post-unblinding estimates plotting against the reported metered emissions. The red square represents no detection. The red circles represent data flagged as below the detection limit (A.16). The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from the resamples. The orange is the Ordinary Least Squares (OLS) fit. The p-value of 0.075 from the paired t-test between the estimated emissions and Stanford emissions suggests that we cannot reject the null hypothesis that the population mean of the differences is 0 with a 95% confidence interval. . . . .                                                                                                                                                                                                                                                                                            | 35 |
| 2.4 | Volume-controlled release experiment results after unblinding. The results presented in this figure are based on the average between the mIME and the DI method. The black circles represent the post-unblinding estimates plotting against the reported metered emissions. The red circles represent data flagged as low-quality data points by the Stanford team. MethaneAIR flags none of these data points. The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from the resamples. The orange is the Ordinary Least Squares (OLS) fit. The p-value of 0.082 from the paired t-test between the estimated emissions and Stanford emissions suggests that we cannot reject the null hypothesis that the population mean of the differences is 0 with a 95% confidence interval. . . . .                                                                                                                                                                                                                                                      | 37 |
| 2.5 | mIME vs DI estimated emissions for all valid cases from all the research flights of interest in 2021. Using the algorithms presented here and meteorological fields from HRRR, MethaneAIR observations of $\text{XCH}_4$ enabled accurate estimation of methane emission from point sources within 30% errors, close to zero bias, and a detection limit of 200 kg/hr. The black circles represent the blind volume controlled release mIME estimates plotting against the DI emission estimates. The green circles represent the unlit flare mIME estimates plotting against the DI emission estimates. The circles representing emission estimates under 500 kg/hr are open. The circles representing emission estimates over 500 kg/hr are solid. The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from resampling. The paired t-test between the Divergence Integral and the mIME emissions ( $p = 0.12$ , rate > 500 kg/hr) indicates no statistically significant difference between the population means for the two methods. . . . . | 39 |
| 3.1 | The summary of the OSSE framework. MethaneAIR flight data provide the setting for WRF-LES OSSE runs and are image-processed to simulate gridded MethaneSAT data. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | 45 |
| 3.2 | OSSE targets: A. Anadarko (MethaneAIR-X 59, 7 October 2023), B. Eagle Ford (Methane-X 03, 2 June 2023), C. Haynesville (MethaneAIR-X 17, 27 June 2023), and D. Permian (MethaneAIR RFo6, 6 August 2021). . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           | 46 |

|      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |    |
|------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 3.3  | Illustration of the Jacobian domain compared to the GIM produced $\text{XCH}_4$ and WRF-LES $\text{XCH}_4$ outputs. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                    | 51 |
| 3.4  | Example of scene data with large (3000 kg/hr) and small (300 kg/hr) emission sources Permian Basin – MethaneAIR Flight RFo7, Aug 9, 2021. This observed plume suggests three stages depending on the mixing level and distance from the source. . .                                                                                                                                                                                                                                                                                                            | 54 |
| 3.5  | Example of clustered methane sources in the Permian from MethaneAIR RFo6 on August 6, 2021 (left) and projected MethaneAIR data onto the MethaneSAT grid (right) with 20 ppb rms noise. . . . .                                                                                                                                                                                                                                                                                                                                                                | 55 |
| 3.6  | Example of cloud effects on the plume. This example comes from MethaneAIR-X flight number 3 on June 1st, 2023. The estimated emission from DI at the MethaneAIR resolution is 1009 kg/hr (left). Once the plume is projected on the MethaneSAT grid, it is impossible to identify or quantify the plume regardless of the horizontal (middle) or vertical (right) cross-track orientation. The dark plume is $\text{CO}_2$ from a power plant, and the methane plume is from a natural gas liquifaction plant (Ethan Manennin, private communication). . . . . | 56 |
| 3.7  | Example of pixel orientation effects on the shape of the plumes. This example comes from MethaneAIR-X flight number 24 on July 19th, 2023. The estimated emission from DI at the MethaneAIR resolution is 1164 kg/hr (left). Once the plumes were reprojected on the MethaneSAT grid, the emissions were $594 \pm 481$ kg/hr when the cross-track direction was horizontal across the scene (middle) and $1381 \pm 796$ kg/hr when the cross-track direction was vertical across the scene (right). . . . .                                                    | 57 |
| 3.8  | DI results based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis). The York regression suggests a $0.94 \pm 0.13$ slope and $-207 \pm 98$ intercept. The SMA regression suggests a $0.97 \pm 0.14$ slope and $421 \pm 88$ intercept. . . . .                                                                                                                                                                                                                                                                         | 59 |
| 3.9  | DI results under 5,000 kg/hr based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis). The York regression suggests a $0.90 \pm 0.11$ slope and $-181 \pm 90$ intercept. The SMA regression suggests a $0.53 \pm 0.08$ slope and $799 \pm 80$ intercept. . . . .                                                                                                                                                                                                                                                       | 60 |
| 3.10 | The plot shows the critical noise values (ppb) as a function of nominal emission rates (kg/hr) based on mIME. The green line represents the critical values for the 1x1 MethaneSAT grid. The red line represents the critical values for the 2x1 MethaneSAT grid. The blue line represents the critical values for the 4x1 MethaneSAT grid. . . . .                                                                                                                                                                                                            | 62 |
| 3.11 | The plot shows the critical noise values (ppb) as a function of nominal emission rates (kg/hr) based on DI. The green line represents the critical values for the 1x1 MethaneSAT grid. The red line represents the critical values for the 2x1 MethaneSAT grid. The blue line represents the critical values for the 4x1 MethaneSAT grid. . . . .                                                                                                                                                                                                              | 63 |
| 3.12 | The left figure is the $\text{XCH}_4$ from the WRF-LES-HRRR. The right figure is the $\text{XCH}_4$ from the GIM Point Source Quantification, reconstructing the plume from the Jacobian of the WRF field. The source strength has been optimized by minimizing the observed (left) and reconstructed (right). . . . .                                                                                                                                                                                                                                         | 65 |

|      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |    |
|------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| 3.13 | This figure shows a time series of mean quantified emission from point sources based on the GIM framework as a function of time in the simulation. We found consistent emission estimates of 1247 kg/hr, similar to the nominal emission rate. . . . .                                                                                                                                                                                                                                                                | 66 |
| 3.14 | The left figure is the XCH <sub>4</sub> from the WRF-LES based on a random Gaussian noise background of $1.0 \pm 0.1$ kg/hr per pixel. The right figure is the XCH <sub>4</sub> from the GIM with the same emission field. . . . .                                                                                                                                                                                                                                                                                    | 67 |
| 3.15 | The mean difference between the GIM and WRF-LES from various OSSE runs. Since WRF-LES has more details from the eddies, the magnitudes are higher than the patterns suggested by the GIM. Overall, the difference is around -9.7 (-14.0, -5.3) mol · km <sup>-2</sup> · s <sup>-1</sup> . . . . .                                                                                                                                                                                                                     | 68 |
| A.1  | WRF's runtime as a function of cores (left) and the speedups as a function of cores . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                         | 76 |
| A.2  | DI results based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis) when the MethaneSAT noise is 40 ppb. . . . .                                                                                                                                                                                                                                                                                                                                                              | 83 |
| A.3  | DI results under 5,000 kg/hr based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis) when the MethaneSAT noise is 40 ppb. . . . .                                                                                                                                                                                                                                                                                                                                            | 84 |
| A.4  | The nominal emission for this figure is 928 kg/hr. A. The panel shows the estimated emission using the light IME method as a function of noise at 1x1 MethaneSAT resolution. B. The panel shows the estimated emission using the light IME method as a function of noise at 2x1 MethaneSAT resolution. C. The panel shows the estimated emission using the light IME method as a function of noise at 4x1 MethaneSAT resolution. . . . .                                                                              | 85 |
| A.5  | The nominal emission for this figure is 928 kg/hr. A. The panel shows the absolute difference between the nominal emissions and the emissions from the light IME method as a function of noise at 1x1 MethaneSAT resolution (red). B. The panel shows the absolute difference as a function of noise at 2x1 MethaneSAT resolution (green). C. The panel shows the absolute difference as a function of noise at 4x1 MethaneSAT resolution (blue). D. The coefficient of variation as a function of the noise. . . . . | 86 |
| A.6  | Post-unblinding best estimates of methane emissions from the controlled release experiments during RFo4 and RFo5 using the mIME method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16. . . . .                                                                                                                                                                                                                                                                               | 88 |
| A.7  | Post-unblinding methane emission estimates from the single-blind volume-controlled release experiments during RFo4 and RFo5 using the DI method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16. . . . .                                                                                                                                                                                                                                                                      | 89 |



|      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      |     |
|------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| A.8  | An analogous plot of Figure 3. The results presented in this figure are based on the “best estimates” reported on 1 February 2022. The black circles represent the post-unblinding comparison between the reported and estimated emissions. The red square represents the original comparison, which was later identified as no detection and removed. The red circles represent the flagged data points determined by the decision tree (Section A.5.4). The blue solid line is the post-unblinding York fit. The orange is the Ordinary Least Squares (OLS) fit. The comparisons shown by the York and OLS fits are notional and do not actually pertain to the information exchanged in the unblinding. The errors in the MethaneAIR data had not yet been assessed in detail and, in fact, were over-estimated. The MethaneAIR team did not know the metered rates and the associated uncertainties were not documented. . . . . | 95  |
| A.9  | An analogous plot of Figure 3. The results presented in this figure are based on the post-unblinding estimates using the DI results. The black circles represent the post-unblinding comparison between the reported and estimated emissions. The blue solid line is the post-unblinding York fit. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           | 96  |
| A.10 | Methane emission estimates from an unlit flare during RFo4 and RFo5 using the mIME method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           | 97  |
| A.11 | Methane emission estimates from an unlit flare during RFo4 and RFo5 using the DI method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                             | 98  |
| A.12 | Barplot of the miscellaneous sources quantified in the Delaware Basin. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | 99  |
| A.13 | ASOS surface wind comparison from the single-blind volume-controlled release experiments during (a) RFo4 on 30 July 2021 and (b) RFo5 on 3 August 2021. The purple crosses represent ASOS winds from the Midland International Air & Space Port (MAF), which is 15 km from the single-blind volume-controlled release site. The orange x’s represent ASOS winds from the Odessa-schlemeyer Field Airport (ODO), which is 17 km from the single-blind volume-controlled release site. The magenta circles represent surface winds from the LES at the flare. The blue triangles represent surface winds from the LES at the single-blind volume-controlled release site. The dark green vertical lines represent times when MAIR flew over the targets. All LES winds during the controlled release experiments and unlit flare observation agree with the ASOS winds (two-sided t-test p-values are greater than 0.05). . . . .      | 101 |

|      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |     |
|------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| A.14 | Estimated emission rates with the mIME using different wind products vs. the known emission rates from the Stanford team. From left to right: estimated emission rates based on effective winds from WRF-LES-HRRR, HRRR, and measured winds. The paired t-test between the estimated emissions and Stanford reported emissions suggests that we cannot reject the null hypothesis that a true difference in means is equal to 0 within 95% confidence interval for the estimated emissions based on WRF-LES-HRRR and Stanford observed winds with p-values of 0.166 and 0.309 respectively. However, the test suggests the alternative hypothesis of a true difference in means between the emissions derived from HRRR winds and the reported emissions with a p-value of 0.0408.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              | 102 |
| A.15 | The thresholds do not affect the IME values of the plume of interest.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                           | 103 |
| A.16 | Updated decision tree classifying quality levels of emission estimates. Most of the steps were developed purely based on our analysis of the data and signal processing. The steps in the red dashed box were added after the unblinding process. First, we filter out all the estimates that (i) have the absolute differences between mIME emission estimates using thresholds of 1 and 2 standard deviations divided by emission estimate using a threshold of 1 standard deviation to be greater than 0.5 or (ii) no emission estimates at the threshold of 2 standard deviations. After that, for each estimate with SNR below 1 or significant interference was observed, we classify the case as either no detection (Endpoint 0) or poor quality (Endpoint 1). The estimates are moderate quality when the SNR is between 1 and 2.5 (Endpoint 2). Only the mIME method can be used. When the SNR is above 2.5, we checked if the plume size is greater than one eddy. If the plumes are smaller than one eddy, only the mIME method can be used (Endpoint 3). All three different methods developed were applied if the plumes are larger than one eddy (Endpoint 4). For the last step, the WRF-HRRR-LES were compared to the nearby ASOS winds and later assigned quality flags based on agreement to the observed winds. If the winds disagree, the wind quality is low. When winds agree, the wind quality is high. | 105 |
| A.17 | Filtered single-blind volume-controlled release RFo4 Segment 1.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 108 |
| A.18 | Filtered single-blind volume-controlled release RFo4 Segment 2.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 108 |
| A.19 | Filtered single-blind volume-controlled release RFo4 Segment 3.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 108 |
| A.20 | Filtered single-blind volume-controlled release RFo4 Segment 4.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 108 |
| A.21 | Filtered single-blind volume-controlled release RFo4 Segment 5.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 109 |
| A.22 | Filtered single-blind volume-controlled release RFo4 Segment 6.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 109 |
| A.23 | Filtered single-blind volume-controlled release RFo4 Segment 7.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 109 |
| A.24 | Filtered single-blind volume-controlled release RFo4 Segment 8.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 109 |
| A.25 | Filtered single-blind volume-controlled release RFo4 Segment 9.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 110 |
| A.26 | Filtered single-blind volume-controlled release RFo5 Segment 1.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 110 |
| A.27 | Filtered single-blind volume-controlled release RFo5 Segment 3.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 110 |
| A.28 | Filtered single-blind volume-controlled release RFo5 Segment 5.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 111 |
| A.29 | Filtered single-blind volume-controlled release RFo5 Segment 7.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 111 |

|      |                                                                          |     |
|------|--------------------------------------------------------------------------|-----|
| A.30 | Filtered single-blind volume-controlled release RFo5 Segment 9. . . . .  | 111 |
| A.31 | Filtered single-blind volume-controlled release RFo5 Segment 11. . . . . | 111 |
| A.32 | Filtered single-blind volume-controlled release RFo5 Segment 13. . . . . | 112 |
| A.33 | Filtered single-blind volume-controlled release RFo5 Segment 15. . . . . | 112 |
| A.34 | Filtered single-blind volume-controlled release RFo5 Segment 17. . . . . | 112 |
| A.35 | Filtered single-blind volume-controlled release RFo5 Segment 21. . . . . | 112 |
| A.36 | Filtered single-blind volume-controlled release RFo5 Segment 23. . . . . | 113 |
| A.37 | MethaneAIR RFo4 flight track . . . . .                                   | 113 |
| A.38 | MethaneAIR RFo5 flight track . . . . .                                   | 113 |
| A.39 | MethaneAIR RFo1E flight tracks . . . . .                                 | 114 |
| A.40 | MethaneAIR RFo3E flight tracks . . . . .                                 | 114 |

# List of Tables

|     |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |    |
|-----|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----|
| I   | Specification comparison for MethaneAIR and MethaneSAT . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | 3  |
| I.1 | Summary of GaoFen-5 Satellite’s specifications . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               | 7  |
| I.2 | Summary of ZY-1 Satellite’s specifications . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | 8  |
| I.3 | Summary of Emission Rate Estimates (Q) from the IME/GEOS-FP method and the WRF-LES-Chem/HRRR method, and the associated wind products of both methods. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                         | 15 |
| 2.1 | Summary of research flights of interest during the MethaneAIR campaign over the summer of 2021 . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               | 20 |
| 2.2 | Summary of research flights of interest during the MethaneAIR campaign over the fall of 2022 . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 | 22 |
| A.1 | Specification comparison for MethaneAIR and MethaneSAT . . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       | 82 |
| A.2 | RF04 and RF05 combined best estimate results from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 (RF04) and 3 August 2021 (RF05). The times are in UTC. The emission rates, lower bounds, and upper bounds are in kg/hr. The blinded, blinded lower, and blinded upper columns were reported to the Stanford team as MethaneAIR initial emission estimates on 1 February 2022. The unblinded, unblinded lower, and unblinded upper columns were added after the unblinding process. The upper and lower bound estimates were calculated using the method described in Section 2.3.3. . . . . | 90 |
| A.3 | DI results from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 and 3 August 2021. The times are in UTC. The emission rates, lower bounds, and upper bounds are in kg/hr. . . . .                                                                                                                                                                                                                                                                                                                                                                                                             | 91 |

|     |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                               |     |
|-----|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----|
| A.4 | Original results for all methods from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 and 3 August 2021. The emission rates, lower bounds, and upper bounds are in kg/hr. This table was reported to the Stanford team as MethaneAIR initial emission estimates on 1 February 2022. The times are in UTC. The bounds were calculated using the method described in Section 2.3.3. The SNR is the signal-to-noise ratio. The SL is the scale length of each plume in meters . . . . . | 92  |
| A.5 | Single-blind volume-controlled release data reported by the Stanford team on 1 February 2022. The cr_kgh_CH4_mean90, cr_kgh_CH4_lower90, and cr_kgh_CH4_upper90 represent the 90-second window rolling average, lower, and upper flow rates in kg/hr. The wind speeds are in m/s. The timestamps are in UTC. . . . .                                                                                                                                                                                                                                                          | 93  |
| A.6 | Single-blind volume-controlled release data reported by the Stanford team on 22 March 2023. The release_rate_kgh is the metered rate reported by the Stanford team. The lower_95CI and upper_95CI are the lower and upper bound of the 95th percentile confidence interval of the metered rates. The MAIR is our blinded best estimate of emissions reported to the Stanford team. The MAIR_lower and MAIR_upper are the lower and upper bound of the 95th percentile confidence interval of our estimates. The timestamps are in UTC. . . . .                                | 94  |
| A.7 | Summary table of removed (not detected) RFO4 plumes based on three methods: decision tree (Section A.5.4), 60 pixel threshold, and K-S test. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                          | 107 |
| A.8 | Summary table of removed RFO5 plumes based on three methods: decision tree (Section A.5.4), 60 pixel threshold, and K-S test. . . . .                                                                                                                                                                                                                                                                                                                                                                                                                                         | 107 |

TO MY GRANDMA AND GRANDPA WHO NEVER SAW THE END OF THIS ADVENTURE.

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# 0

## Introduction

GROWING ENVIRONMENTAL CONCERNS HAVE MADE IT IMPERATIVE TO REDUCE GREENHOUSE GAS EMISSIONS. Methane ( $\text{CH}_4$ ) is the second most important anthropogenic greenhouse gas, with more than 80 times the warming potential of carbon dioxide ( $\text{CO}_2$ ) in the first 20 years after its release<sup>29 13</sup>. Due to its shorter atmospheric lifetime and higher thermal infrared absorbing efficiency, the reduction of methane emissions offers an attractive option for near-term mitigation of



greenhouse gas emissions<sup>44</sup>. Since the oil and gas (O&G) industry accounts for more than  $25 \pm 10\%$  of anthropogenic methane emissions, reducing O&G methane emissions is an effective strategy for reducing greenhouse gas emissions with the added benefit of reduced loss of valuable natural gas<sup>38,13,24</sup>. Identifying and quantifying methane emissions over large regions with high accuracy is crucial for achieving this goal.

Motivated by these concerns, several remote sensing instruments designed to image high concentrations of methane very close to point sources (“point source imagers”) have been introduced in the last decade, including hyperspectral airborne systems such as AVIRIS-NG of Carbon Mapper<sup>16</sup> (not explicitly designed for methane measurement), Ball Aerospace’ Methane Monitor<sup>1</sup>, Kairos<sup>42</sup>, Bridger Photonics<sup>22</sup>, and satellites including PRISMA<sup>18</sup>, the Earth Surface Mineral Dust Source Investigation (EMIT)<sup>49</sup>, Sentinel-2<sup>54</sup>, and WorldView-3<sup>48</sup> (Sentinel-2 and WorldView-3 were also not designed for methane measurements), plus a satellite interferometer, GHGSat<sup>21</sup>. There are also area mapping satellites, satellites that can capture regional-scale methane emissions around the globe, such as the Greenhouse Gases Observing Satellites (GOSAT)<sup>35</sup> with sparse pixels of 10 km diameter pixel resolution globally, TROPospheric Monitoring Instrument (TROPOMI)<sup>55</sup>, with the capability to map the entire globe daily at a much lower spatial resolution (5 km x 7 km) and The Methane Remote sensing Lidar Mission (MERLIN)<sup>11</sup> at 200 km by 200 km resolution. Nevertheless, limited by their resolution, global mapping satellites cannot identify the locations of small methane emissions from O&G operations. Sources too small to detect individually may constitute a significant fraction of all methane emissions in US O&G production<sup>33</sup> although the relative contributions of small and large sources to total emissions are uncertain<sup>41</sup>.

The Environmental Defense Fund (EDF) and its wholly-owned subsidiary, MethaneSAT LLC, have developed a satellite mission, MethaneSAT, to fill the gap between these remote sensing approaches. MethaneAIR is an aircraft-based precursor of MethaneSAT, using very similar spectroscopy, for developing and validating MethaneSAT algorithms<sup>9,46</sup>. Both instruments were de-

signed to achieve a wide spatial swath with fine spatial resolution and high precision compared to existing remote sensing instruments. MethaneSAT launched on March 4, 2024. It is designed to have the capability to quantify methane emissions at regional scales (10 km - 100 km), including diffuse emissions, as well as detailed resolution of point sources. MethaneAIR, deployed periodically since 2019, can also provide high-resolution large-spatial scale methane emission detection and quantification as a stand-alone instrument. While the algorithms should be transferable between the two, adjustments will be needed to match MethaneSAT's coarser resolution, higher detection limit, and broader spatial coverage compared to MethaneAIR data (see the table below for specification comparisons).

**Table 1:** Specification comparison for MethaneAIR and MethaneSAT

| Specification                         | MethaneAIR   | MethaneSAT |
|---------------------------------------|--------------|------------|
| O <sub>2</sub> passband (nm)          | 1237–1319    | 1249–1305  |
| O <sub>2</sub> dispersion (nm/pixel)  | 0.08         | 0.06       |
| O <sub>2</sub> spectral FWHM (nm)     | 0.23         | 0.18       |
| CH <sub>4</sub> passband (nm)         | 1592–1678*   | 1598–1683  |
| CH <sub>4</sub> dispersion (nm/pixel) | 0.10         | 0.08       |
| CH <sub>4</sub> spectral FWHM (nm)    | 0.28         | 0.24       |
| Field of view (°)                     | 23.7         | 21.3       |
| Cross-track pixel <sup>†</sup> (m)    | ~ 5 at 12 km | ~ 108      |
| Along-track pixel <sup>†</sup> (m)    | ~ 25         | ~ 400      |
| Point spread function (pixels)        | 2.5          | 1.8        |
| Single pixel SNR <sup>‡</sup>         | ~ 95         | ~ 190      |

\*MethaneAIR uses an InGaAs detector with reduced QE beyond ~ 1660 nm

<sup>†</sup>Distance between pixel centers at nadir

<sup>‡</sup>CH<sub>4</sub> band with nominal radiance of  $1.4 \times 10^{13}$  photons cm<sup>-2</sup> nm<sup>-1</sup> sec<sup>-1</sup> sr<sup>-1</sup>

This thesis addresses a gap in methane quantification methods by developing a framework that integrates advanced technologies from MethaneAIR and MethaneSAT instruments. The goal is to enhance accuracy and reliability in methane quantification from discrete and identifiable sources. In the first chapter, I introduce quantification frameworks specifically designed for scalability and

adaptability. These frameworks are well-suited for various sectors responsible for methane emissions, aligning effectively with the diverse characteristics of such emissions. The subsequent chapter centers around the performance evaluation of the quantification techniques, particularly focusing on MethaneAIR measurements during blinded controlled release experiments. This evaluation provides insights into the efficacy of the proposed frameworks. Following that, the thesis explores potential applications of the quantification techniques and how they can be effectively applied to MethaneSAT, broadening the scope of their utility. Through application and theoretical analysis, this thesis aims to advance our understanding of methane emissions and advocate for environmentally responsible human practices.

I found that (1) quantification algorithms developed in the first and second chapters provide a strong foundation for advancing methane quantification techniques for future satellites. (2) The controlled release experiment shows that our instruments and algorithms are working as designed, paving the path for application for the MethaneSAT (3) From a set of Observing System Simulation Experiments (OSSEs), we have a better sense of what to expect from the MethaneSAT in terms of quantification performance and detection limits. All of the findings prepare us for the point source quantification operation of MethaneSAT, a satellite intended to revolutionize the world of methane monitoring.

*So since all models are wrong, it is very important to  
know what to worry about; or, to put it in another way,  
what models are likely to produce procedures that work in  
practice (where exact assumptions are never true)*

George Box

# 1

## Methane Quantification Methods

AT THE ONSET OF THIS PROJECT, there were scant observations to tell us what methane plumes looked like at the scale to be observed by the instruments we designed. Sources of methane can be small or gigantic. How can we know what they would look like at the given resolution of our instruments?

## 1.1 INTRODUCTION

This chapter is based on the collaboration with Prof. Luis Guanter & Dr. Itziar Irakulis Loitxate from Valencia Polytechnic University, Spain. At the end of 2021, my advisor and I had been thinking about how to quantify methane emissions for a while, but we didn't have a case to prove that our developed methods worked. One day, we got an email from Luis regarding a potential collaboration. Although we couldn't directly verify our estimates through this opportunity, it gave us a head start in looking at methane plumes from various products.

Before delving into the specifics of our work and its outcomes, it's crucial to contextualize the landscape of methane quantification methods. Many existing methods to estimate tracer emissions from point sources apply Gauss's divergence theorem, including the divergence integral (DI) approach<sup>16</sup>, mass flux<sup>7</sup>, or cross-sectional flux methods<sup>53</sup>. These methods assume that the flux across an enclosing surface around the source equals the integral of flux divergence over the same enclosing surface. In the limit of perfect information and resolution, this is not an assumption but a fact. In practice, however, the accuracy of these methods is limited by the accuracy of the wind profiles used, the spatial resolution and precision of methane observations, knowledge of the vertical distribution of methane and boundary conditions, and the influence of nearby methane sources.

In scenarios where the resolution is coarse (up to at least  $10 \times 10 \text{ km}^2$ ) but the signal-to-noise ratio is high, the source pixel method emerges as a viable option<sup>20</sup>. This method is based on observations very close to the point emission, allowing the observer to neglect information from the plume downwind where turbulent motions may lead to complex morphology.

More recent approaches such as integrated mass enhancement (IME) and machine learning<sup>53,23</sup> combine the measured excess mass of methane near a source, compared to inflow concentrations, with an effective wind velocity flushing the excess downwind, to obtain an emission estimate (discussed in detail below). These methods work best with a fine spatial resolution ( $\leq 1 \text{ km}$ ) and high

measurement sensitivity (column precision  $\leq 10\%$ ). However, cases with multiple point sources in close proximity to one another or very low or high wind speeds are still challenging for all the methods.

This chapter focuses on how we quantified methane emissions of the sources observed by two Chinese Advanced Hyperspectral Imagers (AHSI) onboard China's Gaofen-5 (GF5) and ZY1 satellites using existing and new techniques. This is a significant step for the later chapters, in which I focused on the MethaneAIR and MethaneSAT instruments. The details of the two satellites are followed.

**Table 1.1:** Summary of GaoFen-5 Satellite's specifications

| Technical Parameter    | CH <sub>4</sub>                   |
|------------------------|-----------------------------------|
| Central wavelength(um) | 1.65                              |
| Band width(um)         | 1.642-1.658                       |
| Spectral resolution    | 0.27cm <sup>-1</sup>              |
| SNR                    | 250 @ 0.3                         |
| Radiation calibration  | 5% (relative, ~2%)                |
| Size                   | 790mm (X) x 690mm (Y) x 575mm (Z) |
| Field of view          | IFOV<10.3km@708km                 |
| Sample                 | 5, 7, 9-points                    |
| Observation mode       | nadir(mainly)/glint               |
| Weight                 | 109kg                             |
| Data transfer rate     | 30Mbps                            |

**Table 1.2:** Summary of ZY-1 Satellite's specifications

| Technical Parameter      | Hyperspectral Camera                                                                   |
|--------------------------|----------------------------------------------------------------------------------------|
| Mission life             | 5 years                                                                                |
| Weight                   | 1840 kg                                                                                |
| Launch time              | ZY-1 02D: September 12, 2019                                                           |
| Orbit                    | Sun-synchronous, 10:30 am descending<br>778 km altitude, 98.5° inclination node, angle |
| Sensor bands             | Spectral Range: 0.4 $\mu\text{m}$ - 2.5 $\mu\text{m}$<br>VNIR: 76 bands SWIR: 90 bands |
| Resolution (at nadir)    | 30 m                                                                                   |
| Locational accuracy      | 50 m CE90 (w/o GCPs)                                                                   |
| Dynamic range at imaging | 12 bits                                                                                |
| Swath width (at nadir)   | 60 km                                                                                  |
| Onboard storage          | 2 Tb                                                                                   |
| Revisit capacity         | 5 days                                                                                 |

## 1.2 METHODS

Our methods build upon the previous Integrated Mass Enhancement (IME) algorithm Varon et al. <sup>53</sup> that we adapted to take advantage of the instrument's high spatial resolution. We used the Weather Research Forecast (WRF) model version 4.5 in Large Eddy Simulation (LES) mode combined with geographical and HRRR meteorological fields to simulate eddy-scale winds and vertical mixing. Unlike Varon et al. <sup>53</sup>'s study that was based on idealized LES without specifying specific meteorological data of the sites of interest, our study utilized realistic LES with a high-resolution meteorological data, providing important information for source estimation at the scales measured by the instruments of interest. We also used the wind data from the LES to estimate the emission. The details of the IME and ratio methods are as follows.

## LARGE-EDDY SIMULATION BASED APPROACHES

When the idealized LES only allows us to initialize the boundary conditions of the domain of interest, our realistic LES approach allows us to utilize High-Resolution Rapid Refresh (HRRR) meteorological data. In this chapter, we used the Weather Research Forecast Model driven by HRRR in the Large-Eddy Simulation mode (WRF-LES-HRRR) to simulate plumes at the source of interest. The simulated plumes exhibit stochastic behavior in response to eddy-scale winds associated with boundary layer turbulence, providing quantitative information on the uncertainty due to these processes. The WRF-LES code we used was first developed to simulate CO<sub>2</sub> transport in the US Upper Midwest and Indianapolis areas<sup>26,17</sup>. Our WRF-LES model can take GRIB2 files of the meteorological data of interest (e.g., HRRR, European Centre for Medium-Range Weather Forecasts, and Global Forecast System data) and static geographical data as inputs. For this study, we used HRRR winds to drive the WRF-LES combined with the highest resolution static geographical data available on the official WRF website<sup>2,45</sup>. Given the knowledge of point source locations, we nested (one-way) the domains starting from the native HRRR resolution of  $3 \text{ km} \times 3 \text{ km}$  to  $111 \text{ m} \times 111 \text{ m}$  grid cells by nesting the domains three times at a 3:1 ratio each time. The innermost domain is  $103 \text{ pixels} \times 103 \text{ pixels}$  or  $11.44 \text{ km} \times 11.44 \text{ km}$ . There are 49 vertical layers, with the highest resolution near the surface and coarser towards the top of the atmosphere. We used the Noah Land-Surface Model scheme for the WRF surface option, 1.5 order TKE closure (3D) for the eddy coefficient option of the innermost domain, and Mellor–Yamada–Nakanishi–Niino (MYNN)-Eddy Diffusivity-Mass Flux (EDMF) scheme<sup>30,31</sup>. The closure in the boundary layer limits effective eddy resolution to  $50 \text{ m} - 100 \text{ m}$ ; by limiting our grid to  $111 \text{ m}$ , the run time for the model is tractable, and the simulation of the plumes is presumably realistic on a spatial scale approaching MethaneAIR resolution. We ran the simulations for the days of interest, starting 5 hours before the first observation on each day and ending half an hour after the last observation of interest.



The center of the innermost domain was set to align with the emission source location. When multiple sources are clustered in close proximity, within plus or minus  $1/3$  the domain size from the center (approximately 3.5 km for most of our innermost domains), those sources can be placed in the same model run without expanding the domain size. If the sources spread out more than  $1/3$  of the domain size, we can expand the domain sizes accordingly with additional computation time. Our WRF-LES-HRRR writes innermost domain outputs every minute. The outputs include all the model parameters and a specific methane concentration for every point in the domain.

We identified and isolated the plume from each point source by defining enhancement of the plume above a threshold, 1.5 standard deviations of the inflow values above the median value of the inflow values, as part of the plume of interest (see Figure 2.2B for an example of an isolated plume and Figure A.15 for an example of emission rates as a function of thresholds), setting values below that values to NA. Since we have full knowledge of the emission rate underlying the plume from each LES scene, we could, in principle, determine the “effective wind speed” ( $U_{\text{eff}}$ , the turnover rate for the mass in the plume; Varon et al. <sup>53</sup>) that would give the best fit to the observed plume shape for each encounter, and then estimate the emission rate from the ratio of the mass of the observed plume to the mass of the LES plumes times the nominal emission rate given to the LES model. We used this method, called the Ratio Method (Appendix 1.2), in Irakulis-Loitxate et al. <sup>19</sup> where the resolution of the plume images was close to the LES resolution. However, here, the LES spatial resolution is coarser than MethaneAIR resolution, so we modified the Integrated Mass Enhancement (IME) method of Varon et al. <sup>53</sup> to adapt to our conditions, as described below.

**MODIFIED INTEGRATED MASS ENHANCEMENT** The concept of *IME* emission starts with the computation of the total mass enhancement in a defined plume. The *IME* is the integrated mass

enhancement of the plume and is defined as

$$\text{IME} = \sum_{j=1}^N \Delta\Omega_j A_j. \quad (1.1)$$

where  $\Delta\Omega_j$  is the mass enhancement per pixel, defined as the total column of methane enhancement relative to the inflow concentration, and  $A_j$  is the area of each pixel  $j$ . To obtain the emission rate, Varon et al. <sup>53</sup> used the following equation

$$Q = \frac{U_{\text{eff}}}{L} \text{IME}, \quad (1.2)$$

where  $Q$  is the emission (kg/hr),  $L$  is the scale length defined as the  $\sqrt{A}$ , and  $A$  is the area of the plume ( $m^2$ ). We multiply the  $\text{IME}$  by the turnover rate ( $U_{\text{eff}}/L$ ). The effective wind speed  $U_{\text{eff}}$  is a function of  $\log(U_{10}) + 0.6$ , where  $U_{10}$  is the wind speed at the 10m height.

The Varon et al. <sup>53</sup> IME method used  $U_{10}$  from operational meteorological products, whereas our modified approach (mIME) uses the 10 m root-mean-square wind from each LES realization specifically run for the case of interest, averaged over  $\pm 15$  minutes from the sampling time. The relationship between  $U_{10}$  and  $U_{\text{eff}}$  is given by a set of empirical coefficients (Appendix A.3.1; <sup>53</sup>), reflecting the similarity of the root-mean-square winds in our LES runs to the winds from the large ensemble of (idealized) LES simulations in Varon et al. <sup>53</sup>. Since our LES winds simulate the eddy-scale component applicable to each plume, our simulations estimate the variability of the IME at the time of each observation, providing the uncertainty estimate for each event.

**THE RATIO METHOD** We developed a new method based on WRF-LES-HRRR outputs, assuming that the LES plumes represented the actual plumes in both areas and concentration distributions within the plume. This assumption should hold if the models accurately represent atmospheric transportation of the given plume of interest. At a given location and time, the probabilistic

distribution of methane plumes should be similar at any emission rate. The inferred emission rates should linearly depend on the magnitude of concentrations, which can be written as

$$\frac{Q_{unknown}}{Q_{known}} = \frac{IME_{unknown}}{IME_{known}}. \quad (1.3)$$

$IME_{known}$  refers to the values from the LES simulations with a nominal emission rate (usually 988 kg/hr). Thus, the emission rates of the observed plumes are a function of the nominal emission rate of the simulated plumes multiplied by the ratio of the total masses of the observed plumes to the simulated plumes. The relationship can be expressed as

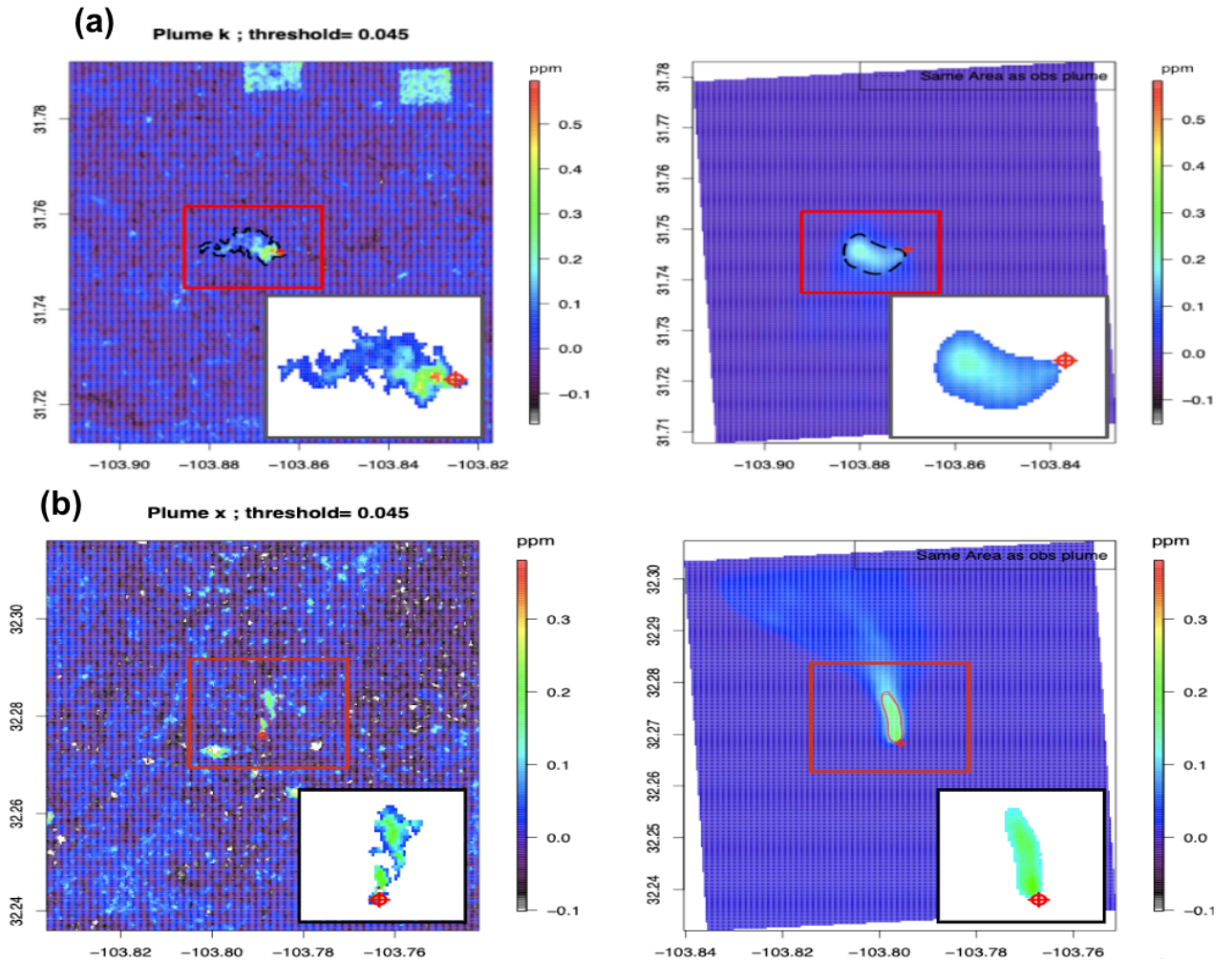
$$Q_{unknown} = Q_{known} \cdot \frac{IME_{unknown}}{IME_{known}}. \quad (1.4)$$

Our ratio method was first used in <sup>19</sup>. It is applicable to very large sources with plumes large enough to be well represented by the 111 m  $\times$  111 m resolution. This limitation arises from the method's dependence on simulated winds that must statistically represent the real atmospheric conditions to yield accurate results.

Since LES cannot capture the instantaneous shape of a snapshot of the observed plume due to large stochastic variations of the atmosphere, we use a range of snaps across time to represent the observed plumes for these two LES methods. We took the standard deviation of the emission rates calculated from 7 snapshots across a 30 minutes interval (15 minutes before and 15 minutes after the retrieval), 5 minutes apart from one another, to avoid autocorrelation effects. For the MethaneAIR data from RFo4 and RFo5 flights, only a few plumes were large enough to be assessed with the ratio and this method is not used in our post-unblinding analysis.

### 1.3 RESULTS FOR GF5 AND ZY1 SATELLITES

We provided independent estimates of emission rates for several plumes (designated “k” and “l” from GF5 and “x” from ZY1)<sup>19</sup>. The WRF model was coupled to 4-D meteorological fields from NOAA’s High-Resolution Rapid Refresh (HRRR) model at the times and locations of the observed plumes. HRRR has a spatial resolution of 3 km x 3 km and is updated hourly, a much finer resolution than the GEOS-FP model used for the IME estimations of emission rates calculated by our collaborators. Our WRF-LES-HRRR model domain was 8 km x 8 km (grid cell 111 m x 111 m), and model plumes were interpolated onto the satellite pixels (30 m x 35 m). We assumed a 988 kg/hr nominal emission rate in each model run. Emissions were scaled by matching the total mass of excess CH<sub>4</sub> in equal areas as follows: The area of each observed plume was defined objectively using the “contourLines” function of R to determine the contour enclosing the threshold for detection of  $\Delta\text{XCH}_4$  (= 0.045 ppm, i.e. 2.5% of total XCH<sub>4</sub>, based on the variance of the retrievals of the satellite images) (see Figure 1.1). The local background was defined within 1 km of the emission source. We then computed a scale factor to match the area-integrated  $\Delta\text{XCH}_4$  of the observed plume (subtracting the local background) with the area-integrated  $\Delta\text{XCH}_4$  of the modeled plume (in equal areas; see Fig X). Results were insensitive to the choice of threshold due to the sharp edges of the plumes. As shown in the figures, the WRF-LES-HRRR model produces realistic plumes with distributions of XCH<sub>4</sub> closely aligning between the model and observations. The resulting emission rate estimates do not use any of the analyses of the IME/GEOS-FP in Table 1.3. The independence of these two approaches provides a rigorous test of the IME estimates in the main text, including the spatial form of the identified plumes and the uncertainties associated with errors in meteorological inputs and model structure. Confidence intervals for the WRF-LES-HRRR estimates were obtained from the t-statistic of seven realizations sampled every 5 minutes, starting 15 minutes before and ending 15 minutes after the satellite overpass.



**Figure 1.1:** Observed (left) and modeled (right) plumes of (a) plume k on August 2, 2020, and (b) plume x on November 1, 2019. The modeled plume was generated by using WRF-Chem-LES with topography and one-way coupling to full HRRR meteorology. We used the "contourLines" function in R to identify the enclosing area with a detection threshold of  $\text{XCH}_4$  (threshold = 0.045, ppm, i.e. 2.5% of total  $\text{XCH}_4$ ), defining the dashed black line in the main image (red box). The selected plume is in the solid white box at the bottom right corner. The red sun-cross symbols indicate the source locations.

Fig 1.1 Observed (left) and modeled (right) plumes of (a) Plume k on 2020-02-08 and (b) Plume x on 2019-11-01. The modeled plume was generated by using WRF-Chem-LES with topography and one-way coupling to full HRRR meteorology. We used the "contourLines" function in R

to identify the enclosing area with a detection threshold of  $\Delta\text{XCH}_4$  (threshold = 0.045, ppm, i.e. 2.5% of total  $\text{XCH}_4$ ), defining each plume the dashed black line in the main image (red box). The selected plume is in the solid white box at the bottom right corner. The red symbols indicate the source locations. (c) Winds observed by the Automated Surface Observation System (ASOS) at the Winkler Regional Airport (KINK, about 60 km E of Plume k, at the same elevation) on 2020-02-08 (black points), GEOS-FP winds at the closest grid to KINK (green asterisk), and HRRR winds at the closest grid to KINK (red plus).

The emission rates estimated for the observed plumes k and l (see Table 1.3 and Figure 1.1) were lower by about 50% than the estimates in Table 1.3, whereas plume x was higher by 50%. The WRF-LES-HRRR model verifies very large emission sources, with central values for the estimated rates lying just outside the estimated confidence intervals for the IME/GEOS-FP approach as indicated in Table 1.3.

**Table 1.3:** Summary of Emission Rate Estimates (Q) from the IME/GEOS-FP method and the WRF-LES-Chem/HRRR method, and the associated wind products of both methods.

| Plume | IME/GEOS FP<br>Emission Rate<br>Estimate (kg/h) | IME/HRRR<br>Emission Rate<br>Estimate (kg/h) | LES<br>Emission Rate<br>Estimate (kg/h)) | Time<br>(UTC) | HRRR<br>Winds<br>(m/s) | ASOS<br>at KINK<br>(m/s) | GEOS-FP<br>Winds<br>(m/s) |
|-------|-------------------------------------------------|----------------------------------------------|------------------------------------------|---------------|------------------------|--------------------------|---------------------------|
| k     | 1630 ± 430                                      | 1364 ± 307                                   | 690<br>[660, 840]                        | 19:00         | 0.68                   | 0                        | 0.76                      |
|       |                                                 |                                              |                                          | 20:00         | 1.03                   | 1.2                      | 2.06                      |
|       |                                                 |                                              |                                          | 21:00         | 1.98                   | 2.91                     | 2.88                      |
| l     | 2910 ± 900                                      | 2267 ± 583                                   | 1420<br>[1360, 1450]                     | 19:00         | 1.37                   | 0                        | 0.76                      |
|       |                                                 |                                              |                                          | 20:00         | 1.38                   | 1.2                      | 2.06                      |
|       |                                                 |                                              |                                          | 21:00         | 1.34                   | 2.91                     | 2.88                      |
| x     | 2120 ± 830                                      | 1314 ± 420                                   | 3180<br>[2880, 3460]                     | 19:00         | 1.14                   | 0                        | 3.92                      |
|       |                                                 |                                              |                                          | 20:00         | 2.27                   | 4.12                     | 4.02                      |
|       |                                                 |                                              |                                          | 21:00         | 3.22                   | 4.63                     | 3.81                      |

Wind speeds at the source locations appear to be one of the major contributors to the differences between estimates from WRF-LES-HRRR and IME/GEOS-FP. We examined HRRR and

GEOS-FP winds at a nearby airport station (Winkler, TX, Figure A.5.2). On 2020-02-08, HRRR winds and Winkler observations were lower than GEOS-FP at the plume sites at the overpass time (20Z), contributing to the difference in estimated emissions in Table 1.3. A similar pattern also existed on 2019-11-01, just at 20Z. Compared to the reported data from the nearby airport, HRRR winds were closer to the observation. However, because the winds in both WRF-LES-HRRR and IME/GEOS-FP are model products, we cannot know if HRRR data are more accurate than GEOS-FP in general.

#### 1.4 CONCLUSION

Our investigation into high-resolution data from Chinese satellites and the development of quantification methods for methane emissions from identified sources has provided valuable insights, as published in Irakulis-Loitxate et al.<sup>19</sup>. We've gained an understanding of the pivotal role of wind patterns and recognized the limitations inherent in our approaches. We developed new quantification methods based on LES, the mIME and the ratio methods, as well as error estimates for methane point sources. While these methods depend on the accuracy of LES results, they provide alternative approaches for quantifying methane emissions and can be utilized in future validation attempts. This chapter serves as a strong foundation for addressing quantification challenges in upcoming chapters, offering a head start in devising strategies for enhancement and refinement.

*No amount of experimentation can ever prove me right; a single experiment can prove me wrong.*

Albert Einstein

# 2

## Controlled Release Experiments

HOW CAN WE VERIFY OUR RESULTS? This chapter highlights the outcomes derived from the MethaneAIR flights conducted in 2021 and 2022, with a specific emphasis on validating emission estimates for O&G sources. These flights marked some of the initial efforts to estimate methane emissions from MethaneAIR retrievals. The unveiling of the results carried a weight of stress. However, we were relieved to discover remarkable congruence between the estimated emission rates and



the actual emissions released. These experiments played a pivotal role in advancing our comprehension of MethaneAIR’s limitations, providing us with preliminary insights into MethaneSAT’s capabilities.

## 2.1 INTRODUCTION

We carried out a single-blind, volume-controlled release experiment to verify the methods for quantifying methane concentrations and point source emissions using MethaneAIR data. The ground team (the Stanford team) for this experiment released methane at various metered emission rates from a site near Midland, TX, in the summer of 2021 without sharing the emission rates with the MethaneAIR team. The MethaneAIR team repeatedly flew over the site on two different days, and estimated emission rates of methane from a known source location without prior information on actual emission rates or measured wind speeds, and submitted the results to the Stanford team. The Stanford team then revealed the volume-controlled release rates. The MethaneAIR team then made minor adjustments to their retrieval algorithms and quality control framework (“decision tree”) to eliminate false positive cases. This procedure is similar to that used in prior remote sensing blinded-volume controlled release efforts including Sherwin et al.<sup>42</sup>, Frankenberg et al.<sup>16</sup>, Sherwin et al.<sup>43</sup>, and Johnson et al.<sup>22</sup>. The experiment was repeated in 2022 at a site in Southern Arizona.

We validated our results by (1) using the single blinded-volume controlled release experiment and (2) comparing our modified integrated mass enhancement (mIME) and DI methods when applied to uncontrolled sources in the Permian Basin. Our DI method also introduces new features to the application of the divergence integral adapted for our airborne mission, as detailed later in this chapter.

## 2.2 DATA

### 2.2.1 METHANEAIR RESEARCH FLIGHTS

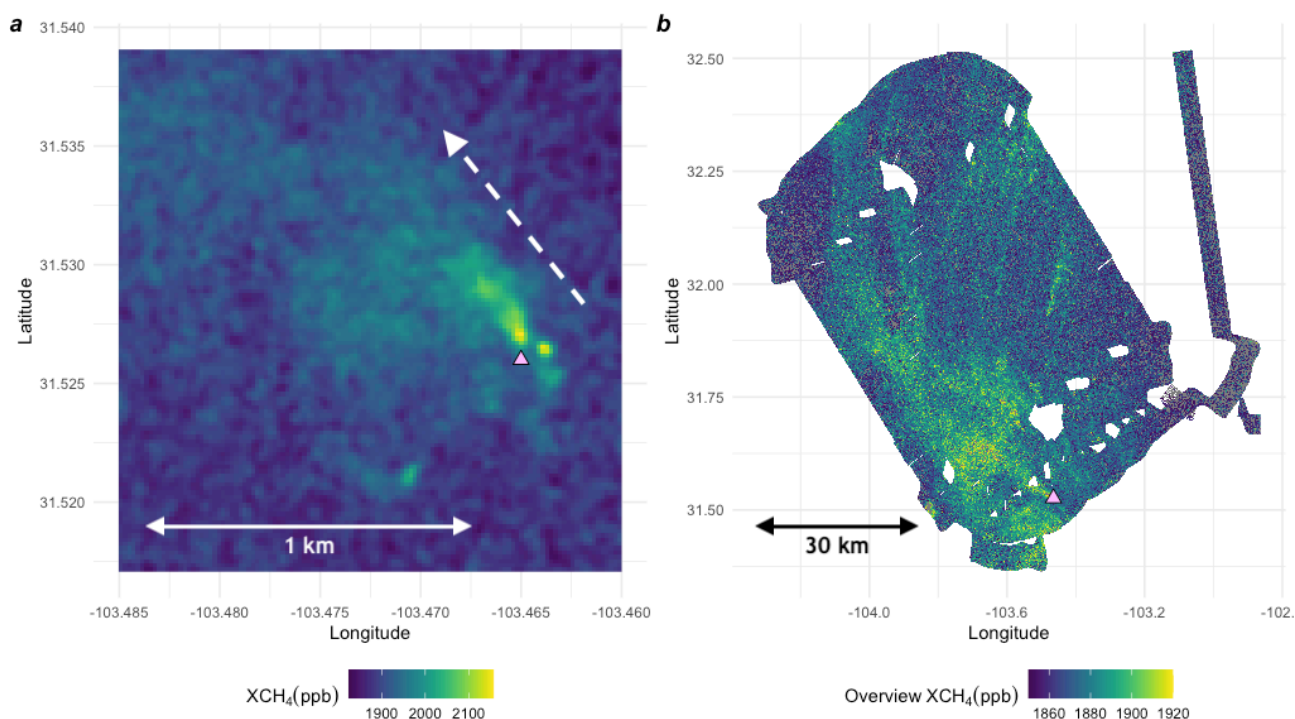
In addition to the controlled-volume experiments, we report here MethaneAIR research flights focused on the Permian Basin, straddling the Texas-New Mexico border, and the nearby Midland-Odessa areas where O&G activities are abundant (see Table 2.1). Methane from nearby sources can confound the identification and quantification of emissions from a particular source, and data from these flights helps us to understand the impact of adjacent sources on our quantification of emissions from point sources in the real world. All the MethaneAIR flights originated from the Rocky Mountain Metropolitan Airport in Broomfield, CO, using the NSF/NCAR GV HIAPER Aircraft (UCAR/NCAR - Earth Observing Laboratory, 2005). Research flights RFo4 and RFo5 targeted the volume-controlled releases, plus validation by our flight over an EM27/SUN solar viewing spectrometer in East Colorado. The plane repeated overflights of the release site from around 12960 m altitude, at approximately 20-minute intervals, for up to 6 hours while the crew on the ground manipulated the release rates. Methane emitted from nearby O&G activity interfered with the observations on at least 3 overpasses. Here, we show the results of 9 and 12 cloud- and interference-free images for RFo4 and RFo5, respectively. The flight tracks can be found in the supplement (FigureA.37, FigureA.38)

Flights RFo6 and RFo7 aimed to map the Delaware sub-basin of the Permian oil and gas field. The strategy for mapping the areas of interest is to create repeated tracks over the target areas with partial overlaps (i.e., area mapping). The overlapped segments act as buffers of missing data, allowing us to observe the changes in the column-averaged dry-air mole fraction of methane ( $\text{XCH}_4$ ) over time.

**Table 2.1:** Summary of research flights of interest during the MethaneAIR campaign over the summer of 2021

| Research Flights | Dates         | Targets                                       |
|------------------|---------------|-----------------------------------------------|
| RFo4             | 30 July 2021  | Blinded-volume controlled releases & EM27/SUN |
| RFo5             | 3 August 2021 | Blinded-volume controlled releases & EM27/SUN |
| RFo6             | 6 August 2021 | Delaware Sub-basin                            |
| RFo7             | 9 August 2021 | Delaware & Midland Sub-basins                 |

During the survey of the Delaware Sub-basin of the Permian Basin in flight (RFo6), MethaneAIR covered more than 9,000 square kilometers in 2.2 hours. An example of what MethaneAIR observed from a source of methane is shown in Figure 2.1 (a). This source was measured during RFo6 and identified as the MiVida Gas Processing Plant in Barstow, TX. When the flight tracks are overlapped, we can combine all the data segments and create a mosaic map of the entire area of interest as shown in Figure 2.1 (b). The MethaneAIR data has one caveat: the basin map was not measured simultaneously, unlike the single snapshot captured by MethaneSAT. The spatial and temporal fluctuations in methane concentrations must be considered. Currently lacking real MethaneSAT data for comparison, the magnitude of differences can only be presumed to correlate with meteorological conditions. Additional investigation is needed.



**Figure 2.1:** Examples of the MethaneAIR data on 6 August 2021 (a) MiVida Gas Processing Plant (pink triangle) from RF06 segment 10 (approximately 2km x 2km image) and (b) mosaic image (75 km x 120 km) of the entire RF06 scene from all the segments including the segment shown in Figure 2.1. The prominent gap areas in this picture were not imaged during the overlapping turns of the pattern. The white dashed line indicates the approximate wind direction at the processing plant site. Full flight track information can be found in A.7.

In 2022, we conducted another single-blind volume-controlled experiment over two research flights near Phoenix, AZ, flight RF01E and RF03E. The flight tracks can be found in the supplement (FigureA.39, FigureA.40). We obtained results of 11 and 13 cloud- and interference-free images for RF01E and RF03E, respectively.

**Table 2.2:** Summary of research flights of interest during the MethaneAIR campaign over the fall of 2022

| Research Flights | Dates           | Targets                                       |
|------------------|-----------------|-----------------------------------------------|
| RF01E            | 25 October 2022 | Blinded-volume controlled releases & EM27/SUN |
| RF03E            | 29 October 2022 | Blinded-volume controlled releases & EM27/SUN |

### 2.2.2 METHANEAIR DATA RETRIEVALS

From our cruise altitude of 12960 m above ground ((13850 above sea level), the across track distance between pixel centers is 5 m and swath of about 4.5 km or 863 pixels. The along-track sampling has centers  $\sim 25$  m apart. The MethaneAIR  $\text{XCH}_4$  data were retrieved from the 1.65- $\mu\text{m}$  band, with  $\text{CO}_2$  as the retrieval proxy for the optical path, using the Smithsonian Planetary Atmosphere interface to VLIDORT (SPLAT-VLIDORT) radiative transfer code<sup>4</sup>. We use  $\text{CH}_4$ ,  $\text{CO}_2$ , and  $\text{H}_2\text{O}$  prior profiles from the TCCON GGG2020 website for the retrieval<sup>50,51,56</sup>. Our multi-layer methane retrieval algorithm is based on the  $\text{CO}_2$  proxy method. We computed high-resolution spectra for methane columns comprising the paths from space to the ground and the ground to the aircraft ( $\approx 12$  km). We account for the influence that topographic variations will have due to the profiles of methane and carbon dioxide using the averaging kernel for our sensor. The tropopause height variations may influence the methane columns due to less methane in the stratosphere. The influence may play a role at some MethaneAIR altitudes and will certainly affect the MethaneSAT data since the satellite is above the atmosphere. Since most *excess* methane lies within the boundary layer (below the aircraft), we assume the averaging kernel for anomalies applies to the lowest kilometers. The averaging kernel is slightly larger near the surface than in the upper atmosphere.

MethaneAIR utilizes two Offner spectrometers (Headwall Photonics) covering specific wavelength ranges and InGaAs detectors (Princeton IR Technologies). We employ the  $\text{CO}_2$  retrieval proxy method in the 1.65  $\mu\text{m}$   $\text{CO}_2$  band and the 2.13  $\mu\text{m}$   $\text{CH}_4$  band. The  $\text{CO}_2$  proxy is known for its robustness under moderate aerosol conditions<sup>15,14,3,34</sup>, with the main errors due to surface albedo

differences between the CO<sub>2</sub>/CH<sub>4</sub> fit windows<sup>35</sup>. Large aerosol concentrations will be flagged by the cloud routines.

Based on the engineering flights, the albedo and aerosol dependence for MethaneAIR data are markedly lower than other methane sensors, given that during the campaign, observations were often over regions blanketed by haze from long-range transport of smoke from fires in the Western United States and Canada. The size distribution may be small enough for the aerosol optical depth to be insignificant at 1600 nm since a large fraction may be in drier air in the free troposphere. The sign and magnitude of albedo bias will be scene-dependent, determined by the prior profile bias and additional light-path modifications induced by aerosol scattering. The detailed discussion can be found in Chan Miller et al.<sup>4</sup>.

The XCH<sub>4</sub> data were later projected onto a 10 m × 10 m grid; in our report to the blinded experiment, we used a nearest-neighbor gridding method that accounts for sensor oversampling on the ground (Appendix A.3.3), whereas in our post-unblinding (“best”) rendering, we used the “snowflake” gridding approach of Sun et al.<sup>47</sup>. The gridded images were denoised using a 2-dimensional mass conserving Gaussian filter with a half-width at half maximum of 0.85 pixels, yielding denoised XCH<sub>4</sub> 10m × 10m gridded data with the noise of 40 ppb (1σ) at the time of the blinded report and 15-20 ppb (1σ) for the best rendering. The spatial decorrelation length for the gridded, denoised image is ~70m.

## 2.3 METHODS

### 2.3.1 METHANE POINT SOURCE EMISSION QUANTIFICATION

We developed two main methane point source quantification approaches, mIME and DI, both adapted from the literature for application to MethaneAIR data. To test the accuracy of our quantification methods, independent of a plume-finding exercise, we assumed that the source location

was already identified. We note that, by assuming that source locations were identified, we have a lower chance of having false positives.

## DIVERGENCE INTEGRAL METHOD

The DI method is based on Gauss' divergence theorem applied to surfaces enclosing the source. The surface integral is estimated based on  $XCH_4$  measurements along a rectangle surrounding the source following Eq. A.5 and Eq. A.6:

$$\Phi_{surf} = \sum_{\text{around rect}} (XCH_{4_i} - \langle XCH_4 \rangle_{rect.}) \cdot n_{column} \cdot M_{CH_4} \cdot v_{\text{perpendicular}} \cdot \Delta l + \left\langle \frac{\partial m}{\partial t} \right\rangle \quad (2.1)$$

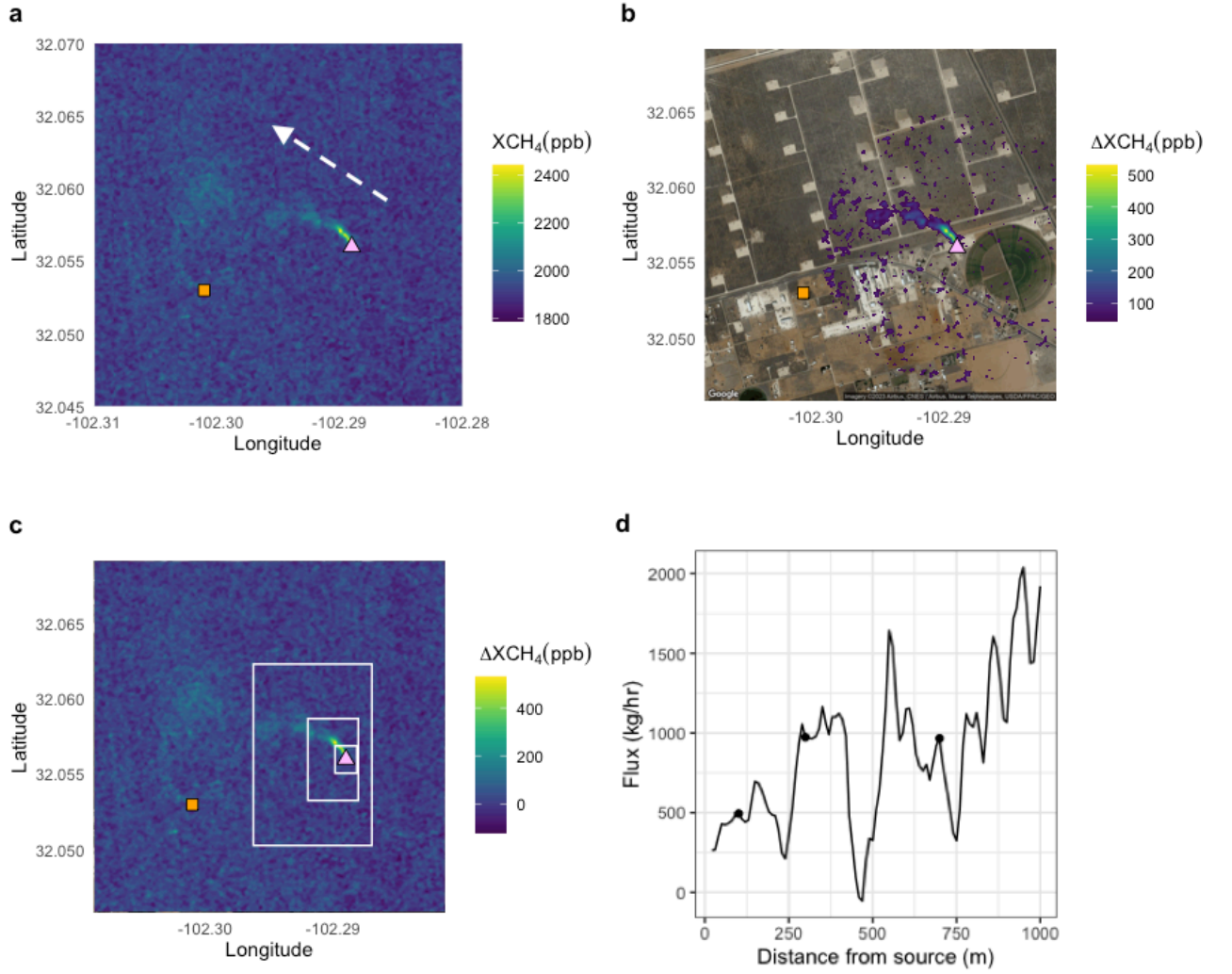
where  $\Phi_{surf}$  is the flux (kg/hr) into the enclosed volume from the land surface,  $XCH_{4_i}$  is an individual measurement along the rectangle,  $\langle XCH_4 \rangle_{rect.}$  is the mean of all measurements along the rectangle,  $\Delta l$  is the distance between successive  $XCH_4$  measurements,  $n_{column}$  is the moles of air in the column based on the surface pressure from HRRR,  $M_{CH_4}$  is the molar mass of methane,  $v_{\text{perpendicular}}$  is the wind speed perpendicular to the sides of the rectangle, and  $\langle \frac{\partial m}{\partial t} \rangle$  is the rate of change of total mass in the enclosed volume ( $\sim 0$  for plume-size volumes). The wind speed was obtained from HRRR but was rotated to match the wind direction of the observed plume using the difference between the major axis of the moment of inertia and the HRRR wind direction (see Appendix A.3.2). Errors in wind direction and wind speed from HRRR will affect the DI method's calculated emissions. We used the wind direction information in the plume image to reduce this potentially large source of error that could flip the sign of the emission estimates.

The flux was calculated for a series of rectangles ("expanding boxes") with the distance from the source to the downwind edge of the rectangle ranging from approximately 100 m to the length of the observed plume (typically 300 - 2000 m). To fully take advantage of the information in the

MethaneAIR image, the size of the rectangle was increased sequentially by one pixel (10 m) in each direction, with the exception of the upwind side, which was increased by a pixel for every fourth rectangle to avoid catching other sources yet still average over the background (Figure 2.2c).

Figure 2.2d shows the calculated fluxes around the unlit flare near the controlled release on 3 August 2021 for individual rectangles as a function of the distance from the source to the downwind edge of the rectangle. Boundary layer eddies break up the plume structure, leading to a buildup of methane concentration in some areas and depletion in others. Since these eddies are not resolved by HRRR, we see oscillations of the computed flux values as a function of distance, on the scale of the associated structures. However, by averaging the flux over the range of distances, we arrive at a robust flux estimate for the source that averages over the eddy-scale variability of the winds. The estimated flux for the individual source is the average flux for all rectangles crossing the plume, in this case, rectangles with edge distances from 100–800 m. (*Note:* The blinded DI emission estimate initially reported to Stanford used the alternate name “Gaussian Integral” for this method).





**Figure 2.2:** a: MethaneAIR image of a blinded volume-controlled release (orange square) and an unlit flare (pink triangle) observed on 3 August 2021. The white dashed line indicates the approximate wind direction at the unlit flare site. b: A sample of the isolated unlit flare plume using the mME method. The isolated plume is superimposed on a ©Google Map 2022 satellite layer. c: Three example rectangles are used to calculate the flux divergence. d: Calculated flux divergence as a function of distance from the source to the downwind edge of the rectangle. Circles indicate the position of the example rectangles are shown in Figure 2.2c. The fluctuations of the apparent flux with distance from the source reflect the influence of eddy scale motion as well as contributions of excess methane from nearby sources. The influence of eddy-scale motions is evident in the oscillation at approximately 250 m intervals, the apparent length scale for eddies at this overpass. The surface flux is estimated by averaging the flux divergence over several eddy scales to average out  $\partial m / \partial t$ . In this example, we averaged the DI from 80 – 700m to avoid influence from other sources nearby that increase the DI beyond 700m from the source.

## MODIFIED INTEGRATED MASS ENHANCEMENT

The modified Integrated Mass Enhancement (mIME) is similar to the approach mentioned in Section 1.2 of Chapter 1, with the only distinction being the subsequent automation of the scripts.

### 2.3.2 WIND VALIDATION

Since the accuracy of the winds directly affects the emission estimates, we examined other sources of wind data to assess the associated uncertainty. We compared our modeled WRF-LES-HRRR winds to wind observations from nearby airports as well as ground-based wind measurements from an instrument at the controlled release site. The comparisons suggest that WRF-LES-HRRR winds are comparable to ground-based wind measurements near the site while the larger-scale or nearby airport winds generated larger errors.

## GROUND WIND VALIDATION

Wind speeds at 10 meters above ground level was measured by a Gill Instruments WindSonic 60 2-dimensional ultrasonic anemometer. The anemometer was placed on a 4 m pole-mounted tripod, secured to the steel flooring of a JLG 400 s telescopic boom lift bucket. The boom lift bucket is designed for a vertical lift while keeping the platform level, and fine adjustments to its height could be made. The anemometer was placed roughly 40 m S-SE from the releasing stack. The data were withheld from the MethaneAIR team before the first data comparison. We later used measured winds from this site to check our estimations from WRF-LES-HRRR.

## AUTOMATED SURFACE OBSERVING SYSTEMS WIND VALIDATION

Automated Surface Observing Systems (ASOS) provide meteorological measurements at airports in the United States, including wind surface observations. The data are reported continuously every

minute and released to the public within two weeks<sup>52</sup>. ASOS winds can be used as quality checks for wind products used in MethaneSAT or MethaneAIR emission estimates when ground-based measurements at the source are not available. The ASOS resolution is 1 kt or 0.5 meter per second, limiting its utility under low wind conditions.

We compared the ASOS data from nearby airports to the average WRF-LES-HRRR winds near the source. The time series of the winds from different products are shown in Figure A.13. The purple crosses represent ASOS winds from the Midland International Air & Space Port (MAF). Because the locations were slightly different, we only look at the variation of the winds within the window of interest. When the variations of the ASOS winds and the WRF-LES-HRRR winds are from the same distribution (two-sided t-test p-value is greater than 0.05), the WRF-LES-HRRR winds are trusted. For the days of blinded-volume controlled release experiments, all the p-values were greater than 0.05 and the LES winds were used in all cases.

### 2.3.3 ERROR ANALYSIS

Since resampling the same view of a plume multiple times is not feasible due to the changing meteorological conditions and flow rates, we used the Monte Carlo simulation principle to calculate the confidence intervals of the mIME estimates. Based on the original observation, we generated a thousand synthetic observations. To simulate a synthetic observation, we sampled  $XCH_4$  pixels of the observed inflow with replacement  $n$  times, where  $n$  is the number of the original inflow pixels. The new resampled inflow mean and standard deviation values were used as a new background and a threshold. After applying the new threshold to the field of  $XCH_4$ , we constrained the extent of the new plume using the original area. A new mass was calculated from the new plume. For the winds, we randomized the mean inflow winds by sampling from seven LES snapshots taken at 5-minute intervals within 30 minutes of the retrieval. We calculated the mIME estimate for each resampled set of values.

Finally, we calculated the confidence interval (the 2.5<sup>th</sup> and 97.5<sup>th</sup> percentile of the emission estimates) from the 1000 resampling trials.

Therefore, our mIME confidence intervals include the variances from the LES winds, instrument, and measurement errors for this particular set of MethaneAIR determination of emission rates.

For the DI method, we used a t-test on the set of fluxes obtained for boxes of different sizes expanding in the downwind direction from the source to estimate the 95% confidence interval of the fluxes. Schneising et al.<sup>39</sup> used a similar approach but only varied the position of the downwind side of the rectangle, leaving the other sides fixed. By changing the position of all sides of the rectangle, we average over different clean segments and reduce errors based on concentration fluctuations outside of the plume (unlike our application, Schneising et al.<sup>39</sup> were able to average over a longer time series enabling them to average out this uncertainty, but we have only one snapshot). We use this method on a much different length scale, as Schneising et al. calculated basin-level emissions, on the scale of 400 km, compared to our goal of quantifying individual emitters with plumes spanning 0.4 to 10 km.

#### 2.3.4 TARGETED EMISSIONS SOURCES

During the MethaneAIR campaign in the Permian over the summer of 2021, we applied mIME and DI approaches to obtain the emission rates, validated the winds, and estimated the errors for the blinded volume-controlled releases of methane and other uncontrolled methane sources in the area.

#### BLINDED-VOLUME CONTROLLED RELEASE EXPERIMENT

Controlled release testing was conducted to evaluate the quantitative performance of MethaneAIR. We conducted repeated passes over the metered methane point source during controlled release in-

tervals. The MethaneAIR team was aware of the release location but not of the release rate of the methane source nor whether any release was happening at a given overpass. Stanford University organized the controlled release experimental campaign near Gardendale, Texas ( $32.053^{\circ}$  N,  $102.301^{\circ}$  W) in July-August 2021, sampled on two days of the campaign. A suite of other teams in controlled release testing, including multiple aerial teams<sup>37</sup> and satellite teams<sup>40</sup>. MethaneAIR participated in the campaign on July 30 and August 3, 2021.

MethaneAIR was in an earlier stage of development than other tested systems, having never obtained any scientific data prior to sampling the controlled release of methane (limited engineering data had been obtained in 2019). Due to this novelty, a slightly modified testing agreement was created with a collaborative publication plan in lieu of independent publication results by the Stanford team, as was performed for other teams. Other protocol changes include: (1) MethaneAIR submitted three sets of estimates using different algorithms and selected which was considered to be best before unblinding (2) MethaneAIR requested that information about wind direction and the presence or absence of meaningful wind speeds be provided, which Stanford sent to Harvard during the experiment seven times. This information was intended solely to inform the aircraft's flight trajectory, which was not, in fact, modified from the filed flight plan during the testing.

The limitations of the 2021 experiment include (1) a small range and low flow rates of the controlled release values (all less than 1000 kg/hr) compared to larger than 1000 kg/hr releases observed by other participants, (2) the release sites were shifted between 3 m and 5.25 m altitude and (3) the number of blinded-volume controlled releases measured was limited to 21 points on two days. The experiment was, however, fully blinded — the metered emission values were not revealed to the MethaneAIR team until after initial emission estimates from MethaneAIR using the mIME and DI methods were reported to the Stanford team, including all overpasses where the IME method detected an  $\text{XCH}_4$  plume above a 1.5 standard deviation threshold within one kilometer from the releasing site. An unplanned limitation was imposed by significant methane emissions from sources

close to the releasing point, including an unlit flare with a high emission rate located at  $32.056^{\circ}$  N,  $102.288^{\circ}$  W,  $\sim 1$  km to the east. This suggests that sources within 1 km may impact the ability to quantify emissions. Emissions from this source were detected on all passes of the blinded-volume controlled release experiment. MethaneAIR has higher sensitivity and a large pixel size (10m x 10m or 20m x 20m, depending on the gridding choices) than other airborne sensors. The methane from this and other extraneous emission sources sometimes interfered with MethaneAIR observations, especially for the DI.

After unblinding the 2021 experiment, we observed the second set of single-blind volume controlled releases near Phoenix, AZ ( $32.821^{\circ}$  N,  $111.786^{\circ}$  W) on 25 & 29 October 2022. Stanford's 2022 instruments have better accuracy than those used in the 2021 experiment<sup>12</sup>. With better precision in the instruments, the Stanford team reported the 2022 emission rates as 60-second means instead of 90-second means in 2021.

## EMISSION RATES FOR MISCELLANEOUS METHANE SOURCES IN THE DELAWARE BASIN

Many miscellaneous methane sources were detected during the MethaneAIR research flights RFo6 and RFo7. We compared the two emission estimates, mIME and DI, and report here both values. We manually identified the scenes with high methane concentrations and co-located with any O&G infrastructures or reported leaks. The sources from RFo6 and RFo7 enabled us to estimate methane emissions from 9 overpasses of the MiVida gas plant ( $31.524^{\circ}$  N,  $103.467^{\circ}$  W, Bartow, TX) over two days. We discovered and quantified a very large pipeline leak in New Mexico on RFo7.

## 2.4 RESULTS AND DISCUSSION

### 2.4.1 BLINDED VOLUME-CONTROLLED RELEASE EXPERIMENT (2021)

For two testing days, MethaneAIR collected 21 data points at the Arizona site. We included emission estimates based on the mIME, the DI, and the Ratio methods presented in Table A.4 as the original results submitted to Stanford by MethaneAIR on 1 February 2022 (before unblinding the metered release volumes). Based on the decision tree workflow in Appendix A.5.4 using both mIME and DI results, a set of blinded best-estimate emissions was submitted to Stanford as the original submission. In this original blinded submission, one emission estimate of the 21 was made using the mean of DI and mIME, and the rest were made using mIME only. After the initial report of the blinded best-estimate data, we decided to use only the mIME method for the consistency of all the data points. No emission estimates were modified post-unblinding. However, some edge cases were flagged via the decision tree. Both blinded best estimate and post-unblinding results are presented in Table A.2. Only post-unblinding mIME results are presented in Figure 2.3. For references, the post-unblinding DI results are presented in Table A.3. The post-unblinding mIME and DI results are also presented in Figure A.6 and Figure A.7 as bar plots representing the emissions and color-coded following the decision tree. In addition, the post-unblinding DI estimates are presented in Figure A.9 as an analogous plot of Figure 2.3 similar to the blinded best-estimate results that are presented in Figure A.8.

After the Stanford team provided the metered emission rates from the experiment, we designed an algorithm that could reliably reject mIME estimates that were false positives or below the detection threshold for a point source. We also developed a more comprehensive bootstrapping approach to account for uncertainty from background concentrations and thresholding in addition to the winds. In the second set of results, this filtering algorithm designated “below detection limit” six emission passes from the dataset; the quantified emissions were slightly changed after unblinding

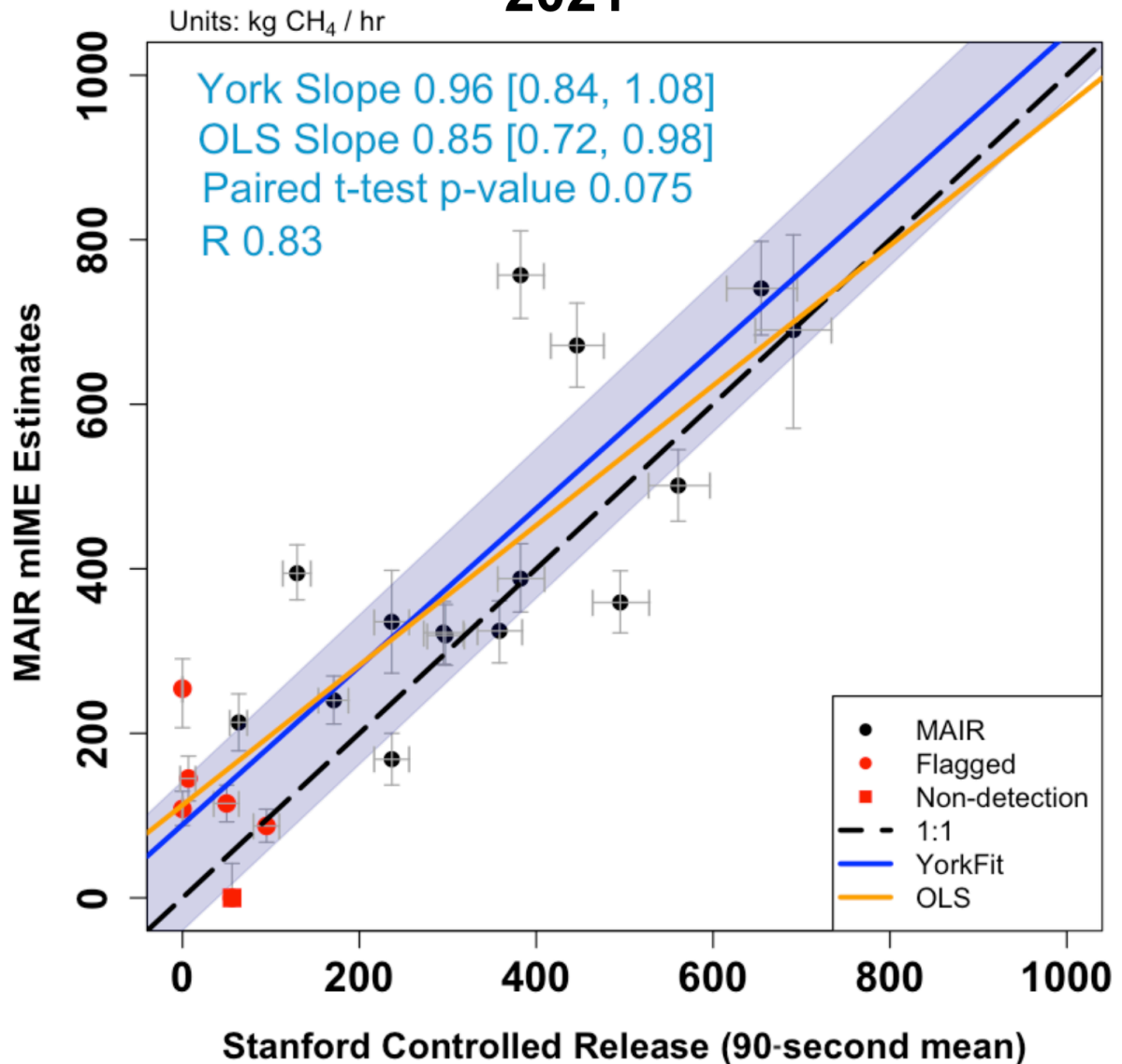
due to the new bootstrapping results. One flagged data point: MethaneAIR estimated the emissions to be above 200 kg/hr when the metered emission is below 200 kg/hr; this observation suggested that, when emissions are below the detection limit, the estimated emissions are uncertain and can be overestimated. These flagged and no-detection data points were not removed before we applied the OLS or York regressions. Based on this comparison, we determined that the detection limit for the MethaneAIR 1X1 retrievals using the mIME method was approximately 200 kg/hr under the conditions of this experiment, which included interference from nearby sources. This value is preliminary because it is based on a small sample size, for two days, in contaminated areas; future work is needed to fully characterize the detection limit (such as using methods presented in Conrad et al. <sup>8</sup>).

Figure 2.3 shows methane flux rates determined by the MethaneAIR mIME method compared to the controlled release rates from the Stanford team from RFo4 and RFo5 flights on 30 July and 3 August 2021, respectively (all the plumes identified and used here are shown in Appendix A.6), including the minor adjustments after unblinding and designating with red circles the false positives in the blinded submission. The red square designates the false negative in the blinded submission. The York regression slope (Appendix A.3.4, MethaneAIR (y-axis) vs blinded-volume controlled releases (x-axis), is  $0.96 (\pm 0.12, 95\% \text{ confidence interval})$ , using error estimates from our bootstrap (y-axis, mIME emission rates) and from the Stanford team (x-axis, reported metered flows). The intercept is 88 kg/hr ( $\pm 27 \text{ kg/hr}$ ). Since the uncertainties in the controlled release rates were not negligible in 2021, the York slope provides the maximum likelihood estimate of the relationship between controlled release and emissions obtained from analyzing MethaneAIR data. We compare this fit to the ordinary least square (OLS) regression results, often used to analyze this type of experiment, where the slope is  $0.85 (\pm 0.13, 95\% \text{ confidence interval})$ ,  $R$  of 0.83, and the intercept is 113 kg/hr ( $\pm 45 \text{ kg/hr}$ ). The limitations of this experiment include (1) a small range of blinded-volume controlled release values (all less than 1000 kg/hr) and (2) a limited number of blinded volume-controlled releases measured ( $N = 21$ ). The excellent agreement between the fluxes calculated by ap-



plying the mIME method to MethaneAIR data and the true controlled release rates provide strong support for the validity of our  $\text{XCH}_4$  retrievals and flux calculations, subject to those limitations.

# MAIR Controlled Release Validation 2021



**Figure 2.3:** Volume-controlled release experiment results after unblinding. The results presented in this figure are based entirely on the mIME method. The black circles represent the post-unblinding estimates plotting against the reported metered emissions. The red square represents no detection. The red circles represent data flagged as below the detection limit (A.16). The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from the resamples. The orange is the Ordinary Least Squares (OLS) fit. The p-value of 0.075 from the paired t-test between the estimated emissions and Stanford emissions suggests that we cannot reject the null hypothesis that the population mean of the differences is 0 with a 95% confidence interval.

We conducted paired sample t-tests to determine whether the mean difference between the reported emissions from the Stanford team and the estimated emissions is different than zero. Our null hypothesis is that there is no difference between the mean observed and estimated emissions. The test was performed three times for three sets of estimated emissions based on three different wind products: WRF-LES-HRRR winds, HRRR winds, and measured winds at the release site (See Figure A.14 for the comparison). The p-values are 0.17, 0.041, and 0.31, respectively. We cannot reject the null hypothesis of no true mean difference between the reported emissions and the estimated emissions derived from WRF-LES-HRRR or observed winds. However, the 0.0408 p-value for HRRR winds suggests that there is likely to be a true mean difference between the reported emissions and the estimated emissions. We infer that the WRF-LES simulated winds improve the mIME method compared to directly using HRRR winds in the IME function. Within our limited data, the mIME method works equally well using WRF-LES-HRRR winds or measured local winds.

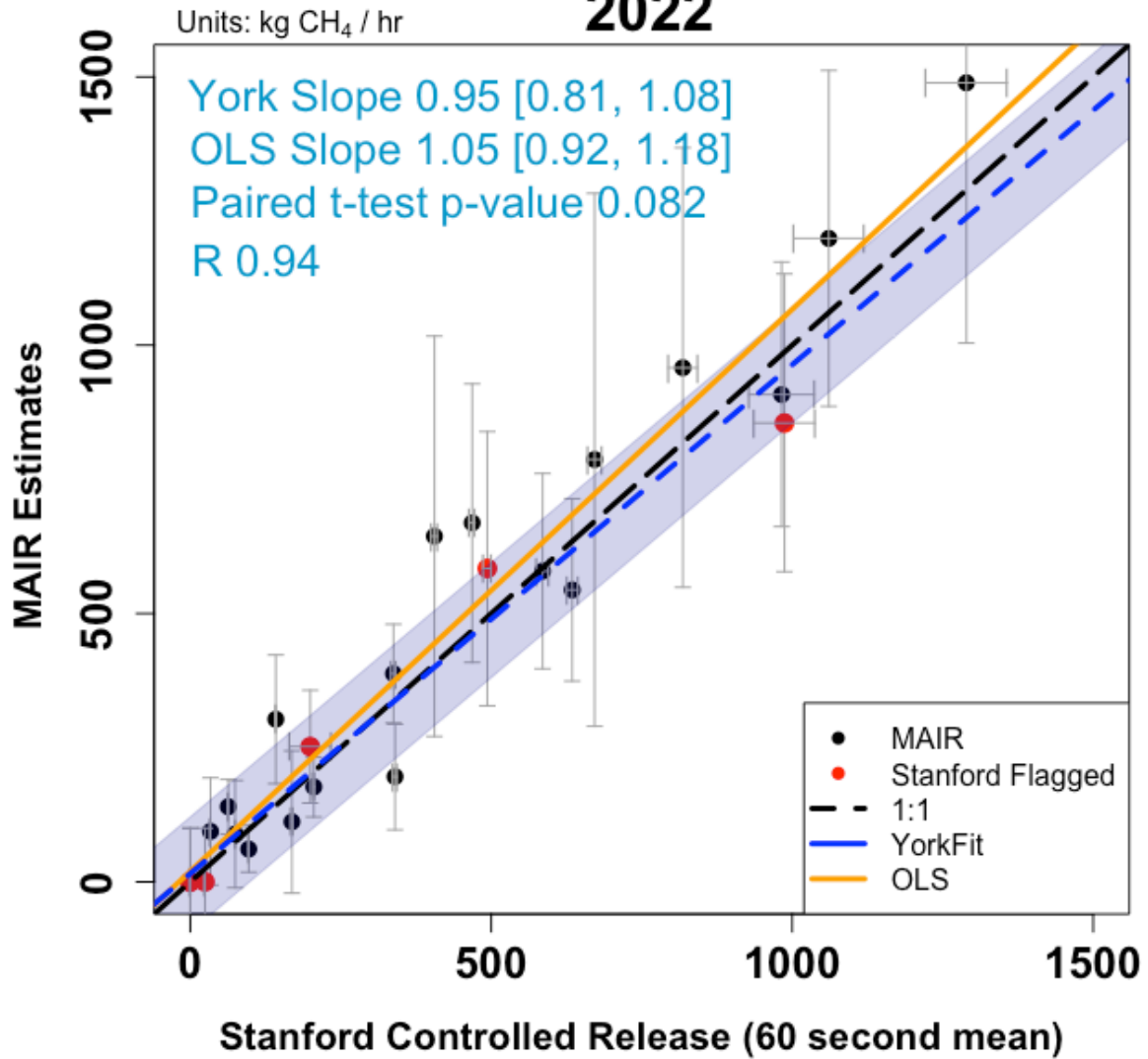
#### 2.4.2 BLINDED VOLUME-CONTROLLED RELEASE EXPERIMENT (2022)

For two testing days in 2022, MethaneAIR collected 24 data points. We included emission estimates based on both the mIME, the DI, and the best estimate (the average between the two methods) as results submitted to Stanford by MethaneAIR on 22 March 2023 (prior to unblinding the metered release volumes).

Figure 2.4 shows methane flux rates determined by the average of the MethaneAIR mIME and DI methods compared to the controlled release rates from the Stanford team from RForE and RForE flights on 25 and 29 October 2022, respectively, including the low-quality estimates designating with red circles in the blinded submission. As a result, the confidence intervals are much larger than the confidence interval in 2021, which was exclusively determined by the mIME method.

# MAIR Controlled Release Validation

## 2022

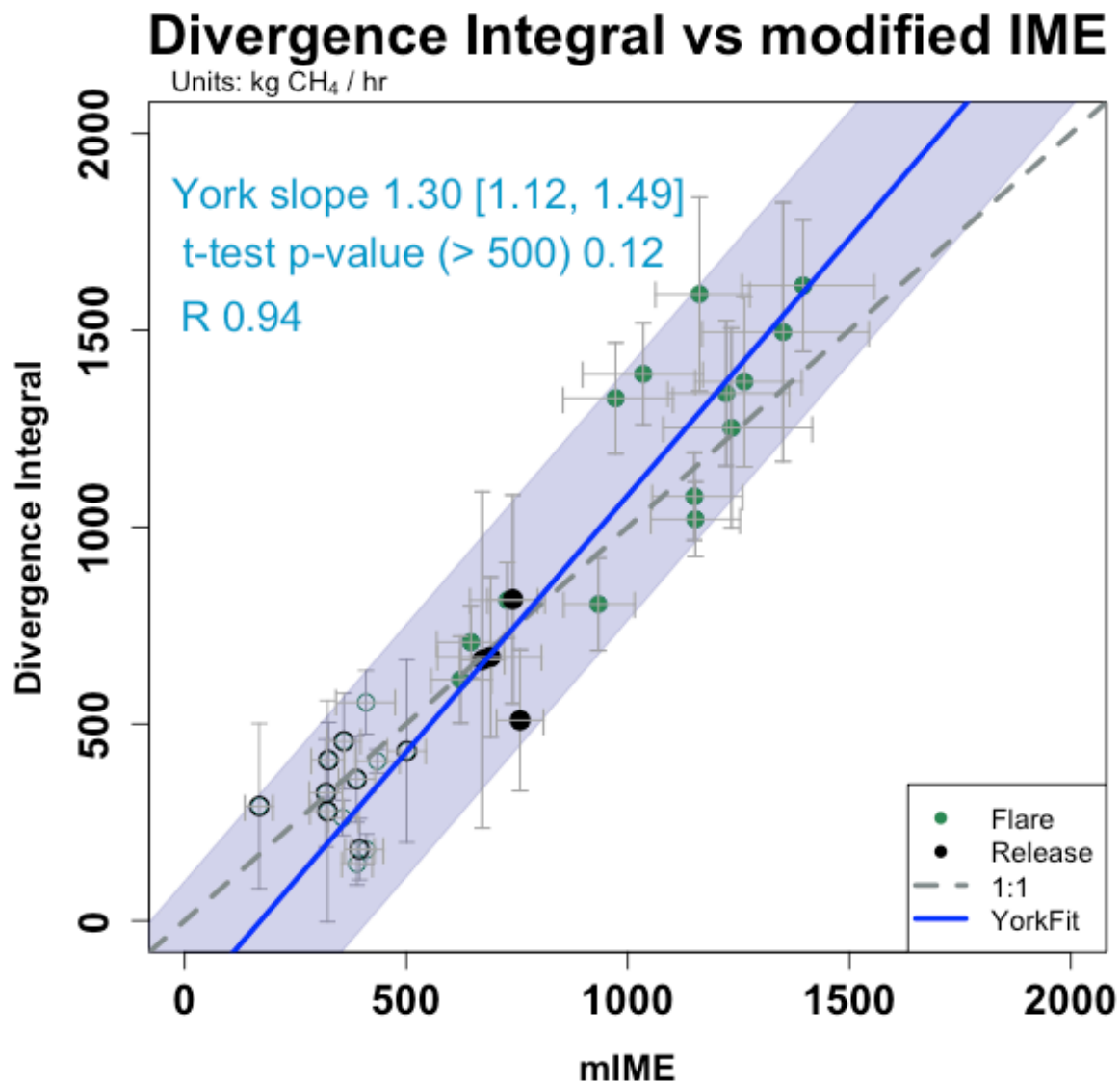


**Figure 2.4:** Volume-controlled release experiment results after unblinding. The results presented in this figure are based on the average between the mIME and the DI method. The black circles represent the post-unblinding estimates plotting against the reported metered emissions. The red circles represent data flagged as low-quality data points by the Stanford team. MethaneAIR flags none of these data points. The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from the resamples. The orange is the Ordinary Least Squares (OLS) fit. The p-value of 0.082 from the paired t-test between the estimated emissions and Stanford emissions suggests that we cannot reject the null hypothesis that the population mean of the differences is 0 with a 95% confidence interval.

### 2.4.3 CONFIDENCE LEVELS OF THE EMISSION ESTIMATES

Beyond the confidence flags from the decision tree (Appendix A.5.4) and attempts to understand the methane contamination of the scenes of interest by methane from nearby sources (Appendix A.5.5), we also evaluated the validity of our emission estimates by plotting the methane emission rates from both methods from all the sources within the same plot (Figure 2.5). This agreement between the two methods has a slope of 1.30 [1.12, 1.49],  $R = 0.94$ , and  $N = 42$  for emissions exceeding 500 kg/hr. The t-test p-value of the two methods is greater than 0.05 and suggests that we cannot reject the null hypothesis that the estimates from these two methods came from the same distribution. Since the two methods agree for the blinded-volume controlled releases and the flare sources when the estimated emission rates are greater than 500 kg/hr, we infer that both methods provide valid emission estimates when emission rates are greater than 500 kg/hr. For rates below 500 kg/hr, both methods detect the sources down to 250 kg/hr, but the DI overestimates the rates due to the influence of nearby sources.

The mean percentage error of the methods should also be similar to those from the blinded-volume controlled release experiment (around 30%), given that uncertainty from both methods came largely from the influence of eddy-scale variability. The mIME simulation captures the variation over time in the simulations. The DI averages the flux information across various distances, capturing spatial variability due to eddies.



**Figure 2.5:** mIME vs DI estimated emissions for all valid cases from all the research flights of interest in 2021. Using the algorithms presented here and meteorological fields from HRRR, MethaneAIR observations of XCH<sub>4</sub> enabled accurate estimation of methane emission from point sources within 30% errors, close to zero bias, and a detection limit of 200 kg/hr. The black circles represent the blind volume controlled release mIME estimates plotting against the DI emission estimates. The green circles represent the unlit flare mIME estimates plotting against the DI emission estimates. The circles representing emission estimates under 500 kg/hr are open. The circles representing emission estimates over 500 kg/hr are solid. The blue solid line is the post-unblinding York fit. The blue shaded area represents a 95% confidence interval of the York fit from resampling. The paired t-test between the Divergence Integral and the mIME emissions ( $p = 0.12$ , rate > 500 kg/hr) indicates no statistically significant difference between the population means for the two methods.

## 2.5 CONCLUSIONS

Our estimates of methane emissions from the blinded volume-controlled release experiment agreed well with the metered flow rates. The 2021 comparison between the MethaneAIR estimates and the known rates provides a slope of 0.96 [0.84, 1.08] with an R of 0.83. The 2022 comparison offers a slope of 0.95 [0.81, 1.08] with an R of 0.94. The results suggest a 30% mean percentage error and a detection limit of 200 kg of methane per hr at 12960 m cruise altitude above ground (13850 m above sea level). The results validate the emission estimates for methane point sources from oil and gas infrastructure using XCH<sub>4</sub> data from the MethaneAIR imaging spectrometer, based on blinded-volume controlled release experiments and comparing two algorithmic approaches for images of uncontrolled sources. Our study supports the application of our emission estimation algorithms to the upcoming MethaneSAT satellite after adjusting for the lower spatial resolution and higher signal-to-noise ratio, given appropriate testing and validation of the satellite. This study is limited by the number of collected data (21 data points on two days, 43 points on four days) from two locales; further evaluations in a variety of conditions are needed to establish detection limits and accuracy more fully.

As shown in Chulakadabba et al.<sup>5</sup>, this study tackles the challenge of quantifying the uncertainty of emissions due to the effects of eddy-scale variability on remote sensing observations. Our methods explicitly estimate the influence of eddies on the values derived for each emission source of interest. The *mIME* captures the variation across simulated cycles. The DI averages the flux information from the XCH<sub>4</sub> image across spatial scales that capture plume size and shape modulation by eddy scale motions. From our limited data, our *mIME* and the original application of IME by Varon et al.<sup>53</sup> perform equally well, provided that local wind information is available. Our *mIME* performs better than an IME using only mesoscale-scale winds from HRRR. Thus, the *mIME* provides an alternative to local wind data, which is typically not available, and it also produces error

estimates associated with the wind variability near the source of interest.

We conclude that the new MethaneAIR imaging spectrometer supports accurate emission rate estimates for high emitting methane sources above 200 – 500 kg/hr detection limit when using the Integrated Mass Enhancement algorithm of Varon et al.<sup>53</sup> modified for our system (*mIME*) or when applying our spatially oversampled application of the divergence integral. The sensor and analysis methods described here provide the greenhouse gas emission quantification community with improved approaches to consider the variability from the eddies. They establish the foundation for applying MethaneAIR and future MethaneSAT data to identify and quantify methane emissions to the atmosphere.

However, uncertainty remains regarding whether our instruments have detection limits that are sufficiently low and emission estimates that are sufficiently accurate to make effective contributions to the broader goal of reducing methane emissions. The distribution of methane emissions sizes is a topic of active debate. Some argue that numerous small methane leaks are the main contributors to overall emissions<sup>33,36</sup>, while others suggest that only a small fraction of large leaks account for the majority of emissions<sup>10</sup>. Despite this uncertainty, we are confident that our instruments, as demonstrated in this chapter, will offer valuable insights for better understanding methane emissions overall.



*I did not want to publish anything until I found something that could be verified experimentally.*

Sir Geoffrey Ingram Taylor

# 3

## Observing System Simulation Experiments

THE OBSERVING SYSTEM SIMULATION EXPERIMENT (OSSE) is a study to estimate the impacts of proposed designs of observing instruments of interest. Since the MethaneSAT satellite was just launched on March 4, 2024, it is impossible to get real data from the satellite during my Ph.D. As a result, I introduced a series of OSSEs that will allow us to predict what to expect from MethaneSAT.

### 3.1 INTRODUCTION

In this chapter, I compare methane emission approximations derived from the WRF-LES simulation analysis with the model's known emission rate. Using the mIME, IME, and DI approaches, I estimate emissions from MethaneAIR data at a resolution of 10 meters by 10 meters and compare to emission rates estimated when those MethaneAIR data are mapped onto MethaneSAT grids with a MethaneSAT noise model. I also introduce the Geostatistical Inverse Modeling (GIM) concept for point source estimation, resembling the ratio method discussed in Chapter 1, normally used for non-point sources.

The rationale behind computing methane emission approximations for concentrations derived from Observing System Simulation Experiments (OSSEs) is to predict methane source quantification capabilities expected from MethaneSAT. Running the WRF-LES-HRRR on a domain of tens of kilometers enables the generation of a four-dimensional simulation (representing three-dimensional space and time) of the area of interest. This simulation offers a comprehensive representation of the system. In contrast to direct observations by MethaneAIR or MethaneSAT, which lack complete knowledge of the sources, our simulation enables us to determine emission rates accurately. Reprojecting the WRF-LES outputs onto MethaneAIR and MethaneSAT resolutions allows us to simulate the data expected to be obtained by our satellite. Through the application of quantification methods to the simulation, we can assess the performance of our algorithms and instruments. Similar to Chapters 1 and 2, the mIME, IME, and DI techniques are applied to the WRF-LES simulation to gain insight into point source emissions. We apply DI and GIM to area sources, defined as sources that cannot be assigned to specific point sources.

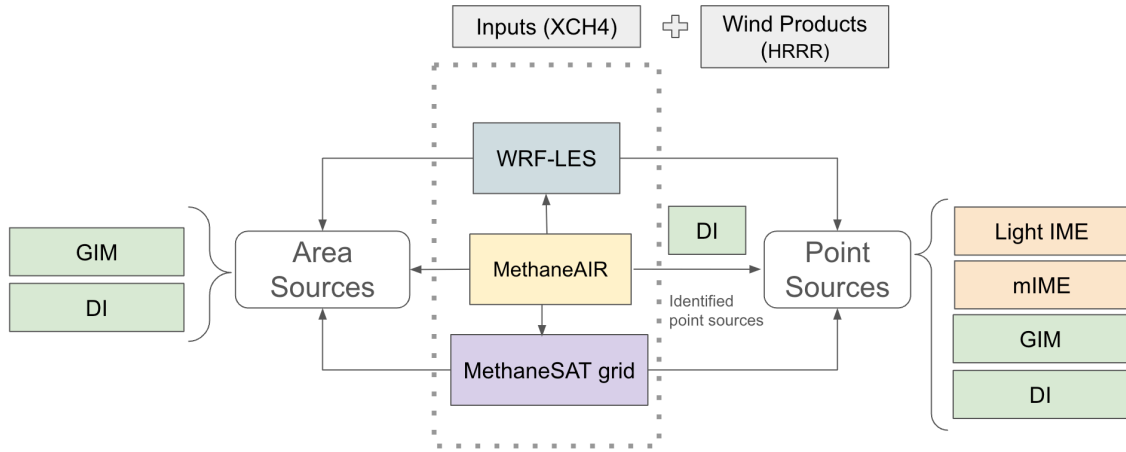
The GIM, initially used for surface flux estimation of atmospheric trace gases, has evolved into a widely utilized framework for emission quantification, with adaptations tailored to specific case requirements Michalak et al.<sup>28</sup>. Dr. Joshua Benmergui of MethaneSAT LLC is developing MethaneAIR

and MethaneSAT's GIM framework. Based on the MethaneAIR and MethaneSAT's GIM framework, I focus on understanding how we can obtain point source emission estimates directly from a GIM and evaluate the GIM's performance on OSSE for area source estimation.

To summarize, the experiments in this chapter are centered on several key areas of OSSE. Firstly, I estimate the quantification limits of MethaneSAT based on WRF-LES simulations and MethaneAIR data. Secondly, I develop prototypes for generating point source emission estimates using the GIM framework. Thirdly, I utilize the OSSE concept to gain insights into the performance of the GIM. Lastly, I establish connections between methane quantification methods across various methane source types. These investigations are essential for accurately identifying and allocating error budgets to MethaneAIR and MethaneSAT's methane emission sources.

## 3.2 METHODS

In this section, I will go over all the aspects of our OSSE represented in Figure 3.1. Each subsection will go into detail regarding the methods and data used to create the analyses represented in the figure.



**Figure 3.1:** The summary of the OSSE framework. MethaneAIR flight data provide the setting for WRF-LES OSSE runs and are image-processed to simulate gridded MethaneSAT data.

### 3.2.1 SCENES OF INTEREST FOR THE OSSE

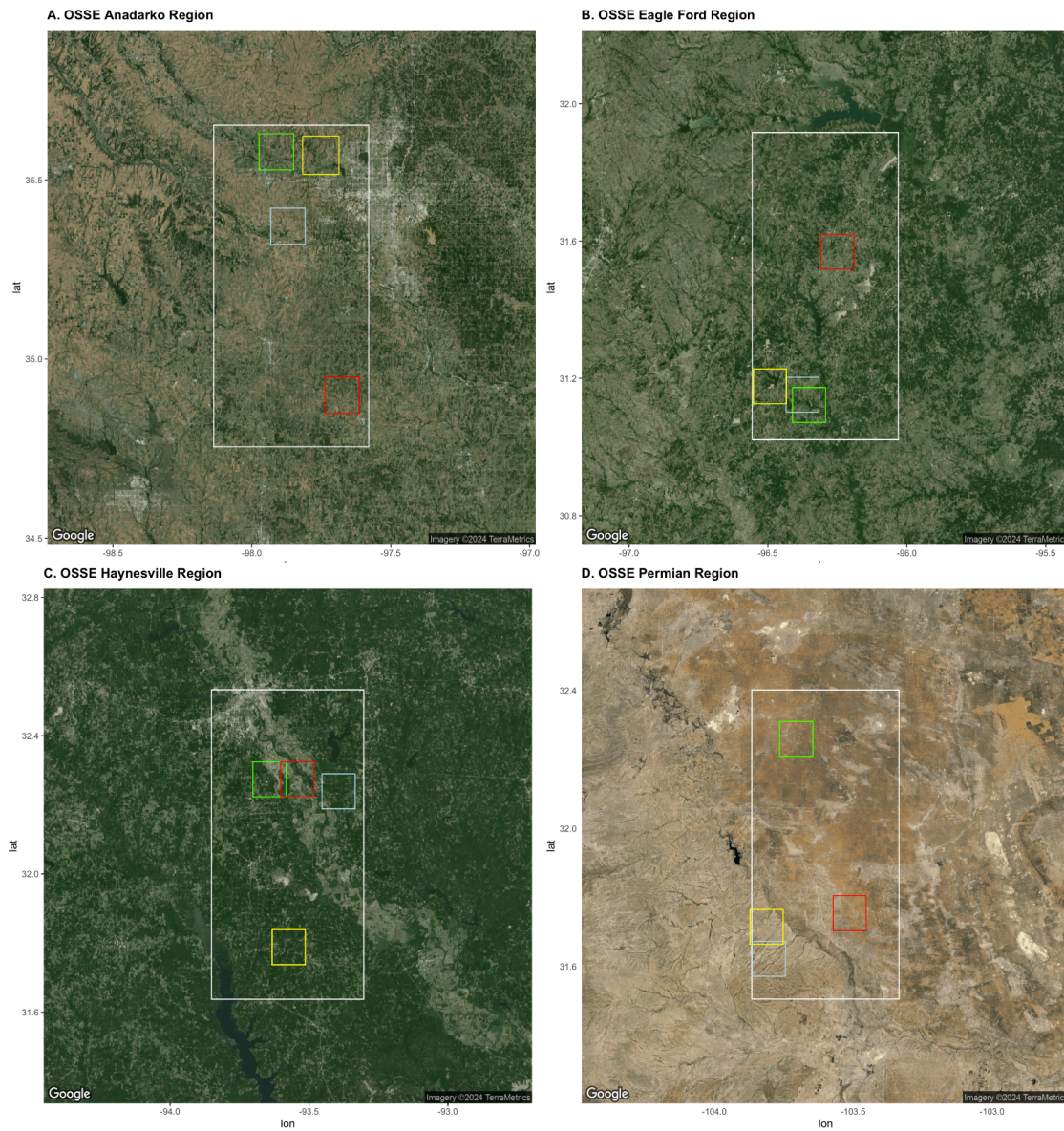
In this chapter, we looked at two sets of data. One is a set of actual data MethaneAIR from various sites, and the other is WRF-LES data at four scenes subsetted from all the MethaneAIR flights.

For the MethaneAIR data, we look at flights RFo1E, RFo3E, RFo4, RFo4E, RFo5, RFo6, RFo7, RFo8, MX002, MX003, MX004, MX007, MX012, MX013, MX015, MX017, MX018, MX022, MX023, MX024, MX025, MX026, MX027, MX030, MX031, MX034, MX035, MX036, MX037, MX039, MX042, MX043, MX044, MX045, MX046, MX048, MX049, MX050, MX051, MX052, MX059, MX060, MX061, MX062, MX063, and MX064.

For the WRF-LES-HRRR data, the four scenes based on MethaneAIR flights are Anadarko (MethaneAIR-X 59, 7 October 2023), Eagle Ford (MethaneAIR-X 3, 2 June 2023), Haynesville (MethaneAIR-X 17, 27 June 2023), and Permian (MethaneAIR RFo6, 6 August 2021). We chose these sites based on important O&G basins among the MethaneAIR flights and days with various meteorological conditions.

For each site, I ran a 50km by 100km WRF-LES-HRRR run at 200 m by 200 m resolution and

four 10km by 10km WRF-LES run at 111m by 111m resolution for the four plumes of interest of a given site. The high-resolution runs are within the red, green, blue, and yellow boxes situated within MethaneAIR flights segments.



**Figure 3.2:** OSSE targets: A. Anadarko (MethaneAIR-X 59, 7 October 2023), B. Eagle Ford (Methane-X 03, 2 June 2023), C. Haynesville (MethaneAIR-X 17, 27 June 2023), and D. Permian (MethaneAIR RF06, 6 August 2021).

In the 50km by 100km, I have 20 tracers representing different interesting scenarios potentially for an OSSE. The 20 tracers are the following: six points source, one EDGAR inventory emission, one square source, one line, one Gaussian noise, one evenly distributed binary, one flat, one gradient, one temporal (on and off emission source in the center), and six Gaussian scale lengths from 3.125km to 6.25km, 12.5km, 25km, 50km, and 100km.

### 3.2.2 THE QUANTIFICATION LIMITS

CAN WE USE WHAT WE HAVE AND INFER POINT SOURCE QUANTIFICATION LIMITS OF METHANE-SAT? In this section, I would like to focus on the point source quantification experiment of the OSSE. Although we could approximately determine MethaneAIR quantification limits based on the MethaneAIR data<sup>4</sup>, we could only guess what it would be like for MethaneSAT based on the specifications when designing and calibrating the instruments. I proposed a way to utilize the WRF-LES-HRRR (similar to what I used in the previous chapters) to simulate methane plumes from various locations in the OSSE domain and use the plume to test the ability to quantify point source emission from the GIM framework. I used the mIME, the IME, and the DI methods discussed in earlier chapters to assess methane emissions across the OSSE scenes. I bootstrapped the emission estimates to get the confidence intervals of the results. I compared the baseline emission rates with the projected emission rates under varying noise conditions. This comparative analysis was conducted three times, each with MethaneSAT resolutions of 1x1, 2x1, and 4x1 pixel<sup>2</sup>, respectively. The resolutions can be translated to roughly 110m by 400m, 220m by 400m, and 440 by 400m, respectively, at the nadir.

By looking at the emission rates as a function of noise, I can observe an increase in the coefficient of variation (CV), which is the ratio of the standard deviation ( $\sigma$ ) to the mean ( $\mu$ ) at the given noise level.

$$CV = \frac{\sigma}{\mu} \quad (3.1)$$

Once the coefficient of variation is more than 0.3 for three noise incremental steps in a row, I defined that noise level as the critical value. The critical value can be interpreted in two different ways. The first way is that the critical value suggests the simple estimate of the MethaneSAT quantification limit at the given nominal emission rate. The second way is that the critical value suggests the simple estimate of the MethaneSAT quantification limit at a particular signal-to-noise ratio. This concept can be generalized and applied broadly for MethaneSAT quantification limits at particular signal-to-noise ratios, regardless of the actual emission rates. This analysis can also draw a connection to Chan Miller et al.<sup>4</sup>'s paper, where he estimated the quantification limits of the MethaneSAT instruments as a function of signal-to-noise ratios and solar zenith angles.

## LIGHT IME

In order to make the IME quantification techniques more efficient, we decided to use the regular IME or light IME, where we simply pull out the large-scale winds and plug them into the IME equation<sup>53</sup>.

Although I have shown in A.5.2 that the IME with large-scale winds provides statistically different estimates than mIME or IME with observed surface winds, the differences are still within 90% confidence interval.

### 3.2.3 PROTOTYPES OF THE GIM FRAMEWORK

Moving on to the OSSE experiment and its GIM application, we can use the OSSE framework to bridge the gap between point source and area source quantification.

This chapter introduced a point source quantification method based on the GIM framework.

GIMs use various types of information data to predict how pollutants are distributed. GIMs differ from more conventional inverse methods by not relying on specific prior emission estimates. The model then adjusts these predictions based on actual observations in the atmosphere to estimate the emission rates that best match the predicted and observed  $\text{XCH}_4$ . GIMs also predict patterns in emissions that may not be present in the available initial predictor datasets.

Based on the concept of inverse modeling,

$$z = b(r, s) + v \quad (3.2)$$

For our study,  $z$  represents a vector of observations with dimensions  $n \times 1$ , while  $s$  is an  $m \times 1$  “state vector” derived from observation of the surface fluxes targeted for estimation. The vector  $r$  includes supplementary parameters crucial for the transport model function  $b(s, r)$ . The difference between the model and the observed data is denoted by the vector  $v$ .

This error includes the measurement error linked to data collection and any random numerical or conceptual inaccuracies related to the assessment of the function  $b(s, r)$ .

In situations where the function  $b(s, r)$  is linear with respect to the unknown  $s$ , as is evident in linear transport models, such as the one to be utilized here, it can be expressed as

$$b(r, s) = Hs \quad (3.3)$$

$H$  denotes a known  $n \times m$  matrix, acting as the Jacobian that characterizes the sensitivity of observations  $z$  to surface fluxes  $s$  (specifically,  $H_{i,j} = \partial z_i / \partial s_j$ ).

STILT is a dispersion model developed from the HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model. STILT focuses on backward-in-time dispersion simulations to identify the source regions of receptors with several key features such as vertical interpolation algorithm for meteorological input files, improved boundary layer height estimation, enhanced turbulence



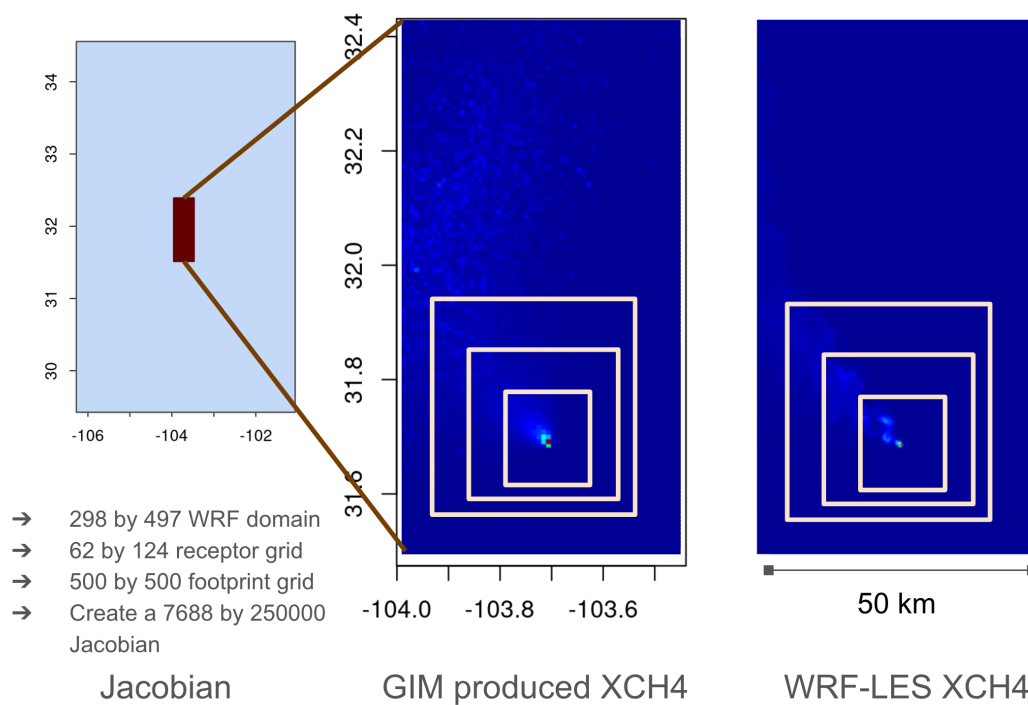
parameterization, dynamic vertical Lagrangian time scale, sophisticated dispersion algorithms, and additional convection schemes compared to traditional HYSPLIT<sup>27</sup>.

We ran a backward Stochastic Time-Inverted Lagrangian Transport (STILT) model to construct our GIM framework's Jacobian ( $H$ ). The Jacobian was created by combining individual STILT footprints into column-averaged footprints. We first decomposed the existing footprint files congruent with the receptor list. Then, we integrated the footprints for each column and assigned them to the Jacobian, which is the key for our point source quantification.

For our GIM framework, our ultimate goal is to minimize the following term.

$$s = H^{-1}(z - v) \quad (3.4)$$

Another question you might raise is how to prevent double counting. We eliminated the methane emission around the sources of interest by simulating the  $XCH_4$  due to each point source and subtracting the point sources from the  $XCH_4$  map and quantifying those point sources separately. The remaining  $XCH_4$  map can later be processed through the GIM approach.



**Figure 3.3:** Illustration of the Jacobian domain compared to the GIM produced XCH<sub>4</sub> and WRF-LES XCH<sub>4</sub> outputs.

### 3.2.4 EMISSION ESTIMATES FROM THE GIM

We could get two types of output from the Jacobian we constructed: emission estimates for point sources and XCH<sub>4</sub> of the entire scenes.

For the point source estimate, we multiply the Jacobian by a vector of a unit emission, which would result in a predicted XCH<sub>4</sub> of the plume (Figure 3.3). By calculating the ratio between the observation (in this case, the WRF-LES-HRRR XCH<sub>4</sub>) and the estimated XCH<sub>4</sub> from the Jacobian, we obtain the ratio between the observed and the input nominal unit emission rates. After multiplying the unit emission rate by the ratio, we can obtain the estimated emission rates of the point sources of interest.

Instead of multiplying the Jacobian by a vector of a unit emission rate, the  $XCH_4$  of the entire scenes can be calculated by multiplying the Jacobian (with matrix size equal to the number of receptors by the number of the footprint grids) by a vector of the emission rates. The emission rates are the same as the ones that we used to initialize the WRF-LES-HRRR runs. For the non-OSSE scenarios, the emission rates would either come from prior emission rates or be the target we want to optimize.

#### TESTING THE JACOBIAN-PRODUCED $XCH_4$ FOR THE POINT SOURCES

Inputs: A unit point at the location of interest ( $e$ ) and the Jacobian ( $H$ ).

Process: To make the Jacobian computationally feasible, I need to reproject the unit point from 200m by 200m to the 800m by 800m grid (a factor of four), matching the Jacobian's resolution derived from STILT runs. For this experiment, I used  $XCH_4$  data from the WRF-LES-HRRR results instead of the MethaneAIR and MethaneSAT plume data. We then multiply the Jacobian by a unit emission vector, resulting in a predicted  $XCH_4$  of the plume ( $XCH_{4GIM}$ ). The calculation is shown in Eq. 3.5. By calculating the ratio between the observation (in this case, the WRF-LES-HRRR  $XCH_4$ ) and the estimated  $XCH_4$  from the Jacobian, we obtain the ratio between the observed and the input nominal unit emission rates. After multiplying the unit emission rate by the ratio, we can obtain the estimated emission rates of the point sources of interest. The calculation is shown in Eq. 3.6. This approach is similar to the ratio method discussed in Chapter 1 (Eq. 1.4)

Outputs: Emission estimates of the plumes ( $Q_{GIM}$ )

$$XCH_{4GIM} = H \cdot e \quad (3.5)$$

$$Q_{GIM} = \frac{XCH_{4observation}}{XCH_{4GIM}} \quad (3.6)$$

## INVERSE $\text{XCH}_4$ FOR THE ENTIRE SCENES

Inputs: A vector of the emissions (E) and the Jacobian (H)

Process: I aggregated the emission inputs of the entire scene (E) from the WRF-LES resolution at 200m by 200m to the resolution of the Jacobian at 800m by 800m. I multiplied the Jacobian by the emission inputs and obtained the predicted  $\text{XCH}_4$  from the GIM ( $\text{XCH}_{4\text{GIM}}$ ). The calculation is shown in Eq. 3.7. To verify the  $\text{XCH}_{4\text{GIM}}$  in this OSSE, I compared  $\text{XCH}_4$  data from the WRF-LES results instead of using the real MethaneAIR and MethaneSAT data.

Outputs: Estimated  $\text{XCH}_4$  for the entire domain from the Jacobian ( $\text{XCH}_{4\text{GIM}}$ )

$$\text{XCH}_{4\text{GIM}} = H \cdot E \quad (3.7)$$

## 3.3 RESULTS

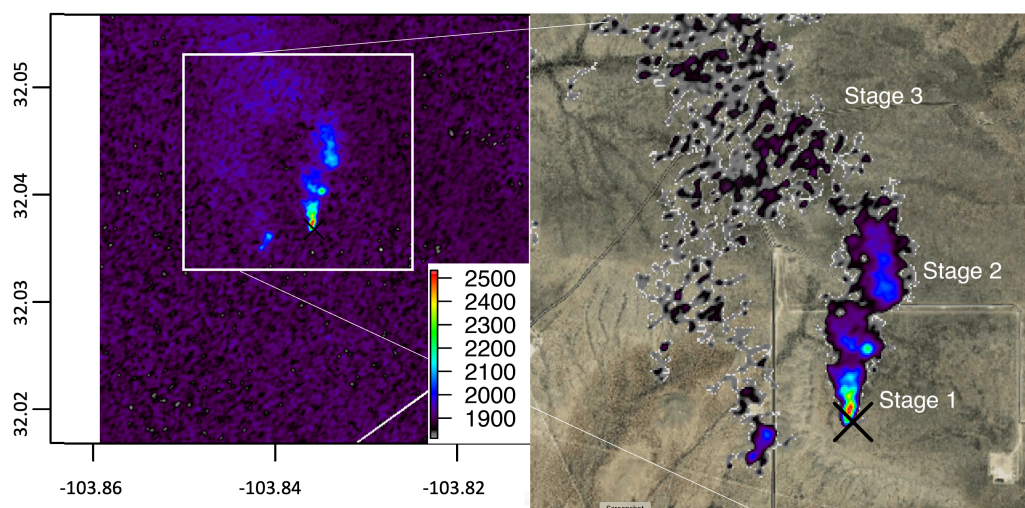
### 3.3.1 METHANEAIR OBSERVATIONS

Before we look at the results, I would like to share some of my observations. Although these cannot be quantified objectively yet, these subjective observations can guide us through future attempts to understand the results better.

#### EFFECT OF PLUME MORPHOLOGY

By looking at the MethaneAIR plumes, I observed the nature of the methane plumes and classified the stages of the plumes into three categories. The first section is near the source where the plume has not yet mixed with boundary layer eddies at an observable rate. The second section is when the turbulence started kicking in, and we started to see the effects of the large eddies on the plume and the expansion to the eddy scale. The plume starts to become dissected. The third stage

is when the plume is fully extended into the boundary layer and starts dispersing horizontally in the atmosphere. This part of the plume is no longer confined and prolate (plume-like). When we quantify methane emissions from a point source, we need to define the extent of the plumes and keep these three stages of plumes in mind. Many point source sensors are most sensitive to the first stage. MethaneAIR is most sensitive to the second stage. As we transition from MethaneAIR to MethaneSAT resolutions, we lose sensitivity when the plume does not fill the larger pixels or fills just a few pixels. Because MethaneSAT has low noise (20 - 30 ppb rms), it is likely to be more sensitive to the third stage compared to the first or the second stages.

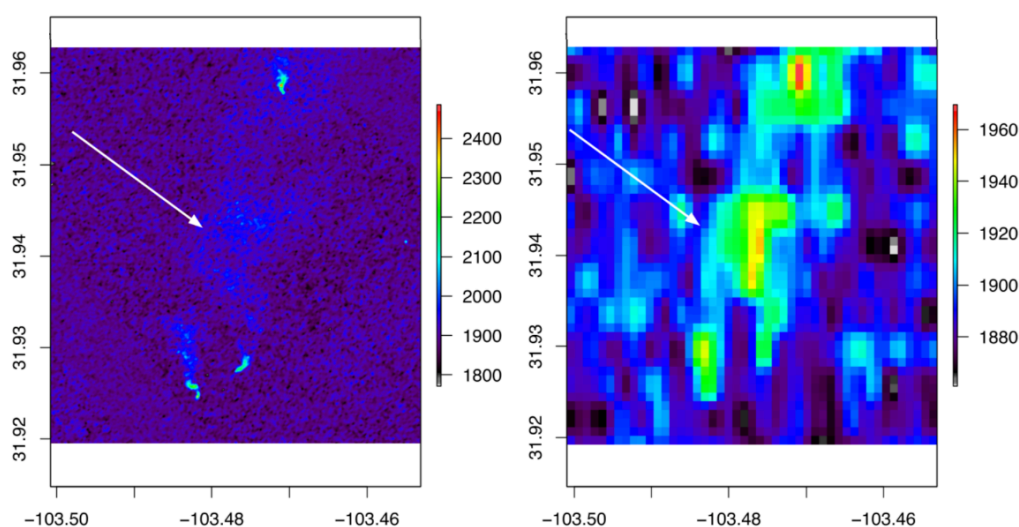


**Figure 3.4:** Example of scene data with large (3000 kg/hr) and small (300 kg/hr) emission sources Permian Basin – MethaneAIR Flight RF07, Aug 9, 2021. This observed plume suggests three stages depending on the mixing level and distance from the source.

## EFFECTS OF CLUSTERED POINT SOURCES

During our analysis of MethaneAIR data, we observed that a minimum of 10% of the plumes aggregated within a 5 km by 5 km area. Figure 3.5 provides an example, showcasing clustered methane sources in the Permian region captured by MethaneAIR RF06 on August 6, 2021 (left) and their

projection onto the MethaneSAT grid (right). While individual plumes on the MethaneAIR grid are separated, distinguishing them becomes challenging once projected onto the MethaneSAT grid without presuming consistent wind directions and limiting the distance between plume fragments to consider them as a single plume. Even with such assumptions, effectively applying the quantification method poses challenges, as the DI method requires specific distances for the growing box technique. If the growing box distance exceeds the distance between plumes, there's a risk of incorrectly treating multiple plumes as a single source when observed by the MethaneSAT.

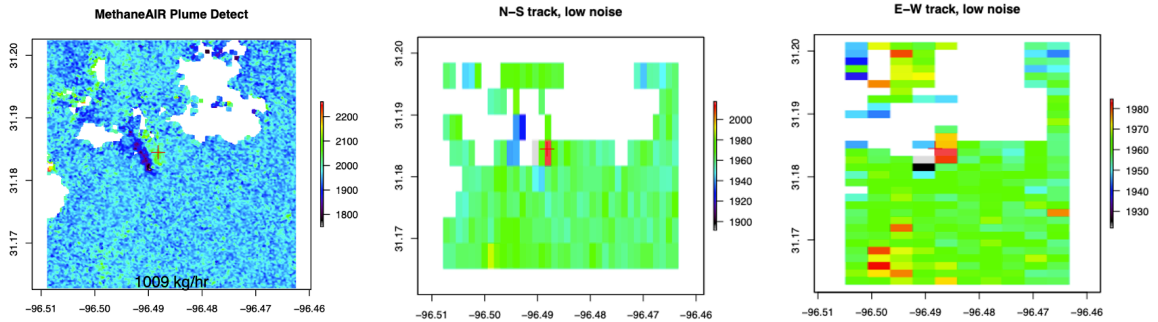


**Figure 3.5:** Example of clustered methane sources in the Permian from MethaneAIR RF06 on August 6, 2021 (left) and projected MethaneAIR data onto the MethaneSAT grid (right) with 20 ppb rms noise.

## EFFECTS OF CLOUDS

Clouds can interfere with our observations when they are above the sources of interest. When clouds are present in the scene, our instrument cannot measure methane under the clouds or in cloud shadows. We can still quantify partially cloud-covered plumes with MethaneAIR's fine spatial resolution. However, identifying or quantifying such plumes may be impossible once projected

on the MethaneSAT grid. Figure 3.6 is an example of cloud effects on the plume. This example comes from MethaneAIR-X flight number 3 on June 1st, 2023. The estimated emission from DI at the MethaneAIR resolution is 1009 kg/hr (left). Once the plume is projected on the MethaneSAT grid, it is impossible to identify or quantify the plume regardless of the horizontal (middle) or vertical (right) cross-track orientation. The plumes in the middle and the right plots are projected MethaneAIR onto the MethaneSAT 1x1 grid without additional noise being imposed on them. However, they are considered low noise and not noise-free since MethaneAIR data contain systematic and random noise.

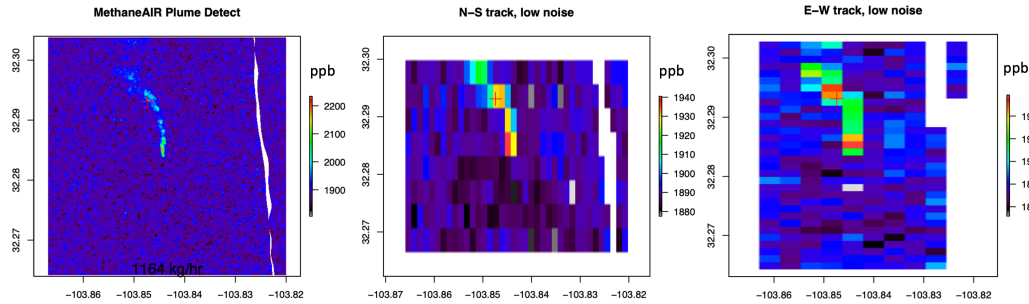


**Figure 3.6:** Example of cloud effects on the plume. This example comes from MethaneAIR-X flight number 3 on June 1st, 2023. The estimated emission from DI at the MethaneAIR resolution is 1009 kg/hr (left). Once the plume is projected on the MethaneSAT grid, it is impossible to identify or quantify the plume regardless of the horizontal (middle) or vertical (right) cross-track orientation. The dark plume is CO<sub>2</sub> from a power plant, and the methane plume is from a natural gas liquifaction plant (Ethan Manennin, private communication).

## EFFECTS OF PIXEL ORIENTATION

For square pixels, the orientation might not have a large effect on the quantifiability of the plumes. However, the orientation is significant for MethaneSAT observation since the native pixel at 1x1 is rectangular. If the cross-track direction is perpendicular to the wind, the plumes will look longer than they actually are. If the cross-track direction shares the same direction as the wind, the plumes

will look wider than they actually are. Figure 3.7 is an example of pixel orientation effects on the shape of the plumes. This example comes from MethaneAIR-X flight number 24 on July 19th, 2023. The estimated emission from DI at the MethaneAIR resolution is 1164 kg/hr (left). Once the plume was reprojected on the MethaneSAT grid, the emissions were  $594 \pm 481$  kg/hr when the cross-track direction was horizontal across the scene (middle) and  $1381 \pm 796$  kg/hr when the cross-track direction was vertical across the scene (right). The plumes in the middle and the right plots are projected MethaneAIR onto the MethaneSAT 1x1 grid low noise similar to Figure 3.6.



**Figure 3.7:** Example of pixel orientation effects on the shape of the plumes. This example comes from MethaneAIR-X flight number 24 on July 19th, 2023. The estimated emission from DI at the MethaneAIR resolution is 1164 kg/hr (left). Once the plumes were reprojected on the MethaneSAT grid, the emissions were  $594 \pm 481$  kg/hr when the cross-track direction was horizontal across the scene (middle) and  $1381 \pm 796$  kg/hr when the cross-track direction was vertical across the scene (right).

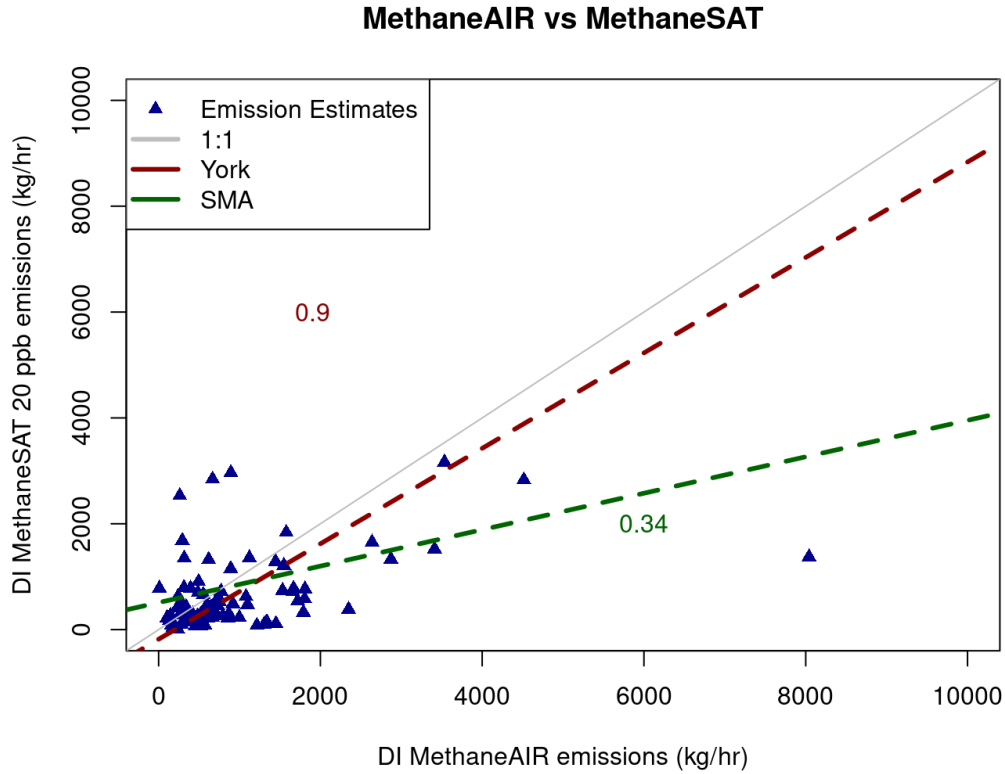
### 3.3.2 REPROJECTION OF METHANEAIR ONTO THE METHANESAT GRID

I analyzed all the plumes detected in MethaneAIR flights and plotted the DI estimated emissions for MethaneAIR resolutions alongside estimates derived from reprojecting MethaneAIR data onto MethaneSAT resolutions (Figure 3.8). The projected MethaneSAT XCH<sub>4</sub> values were determined based on MethaneSAT’s design specifications, featuring a resolution of approximately 110 m by 400 m and a predicted noise level of 20 - 40 ppb. Out of the available 388 MethaneAIR plumes, we could quantify 112 reprojected MethaneSAT plumes, accounting for roughly 30%. The York re-



gression indicates a slope of  $0.94 \pm 0.13$  and an intercept of  $-207 \pm 98$ , while the SMA regression suggests a slope of  $0.97 \pm 0.14$  and an intercept of  $421 \pm 88$ . These results imply that MethaneSAT is likely to capture around 30% of the plumes observed by MethaneAIR. Given the consistent relationship between emission estimates, there's potential to mitigate the emission estimate bias of MethaneSAT through further studies.

An attempt was made to compare the estimates of reprojected MethaneAIR data onto the MethaneSAT grid using the light IME method. However, this approach failed to estimate the emissions of most plumes due to MethaneSAT's coarse resolution and the limited number of pixels covered by each plume.

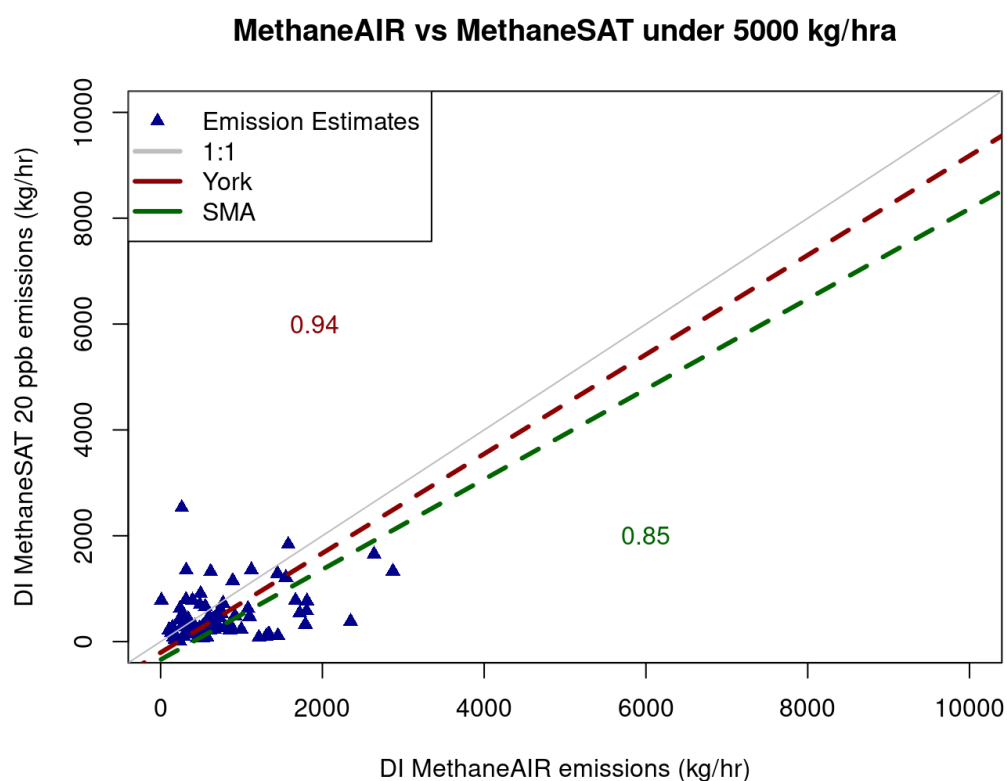


**Figure 3.8:** DI results based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis). The York regression suggests a  $0.94 \pm 0.13$  slope and  $-207 \pm 98$  intercept. The SMA regression suggests a  $0.97 \pm 0.14$  slope and  $421 \pm 88$  intercept.

I investigated the relationship further by constraining the emission rates up to 5,000 kg/hr. The relationship is also tighter if we only look at the plumes under 5,000 kg/hr (Figure 3.9). The York regression suggests a  $0.90 \pm 0.11$  slope and  $-181 \pm 90$  intercept. The SMA regression suggests a  $0.53 \pm 0.08$  slope and  $799 \pm 80$  intercept.

By looking at this experiment closely, this observation suggests that the plumes could not be quantified due to clouds and missing data. When plumes have emissions of more than 5,000 kg/hr, we have the tendency to underestimate their emissions due to the large spreading nature of the large plumes. When I changed the noise to 40 ppb, the difference between MethaneAIR and Methane-

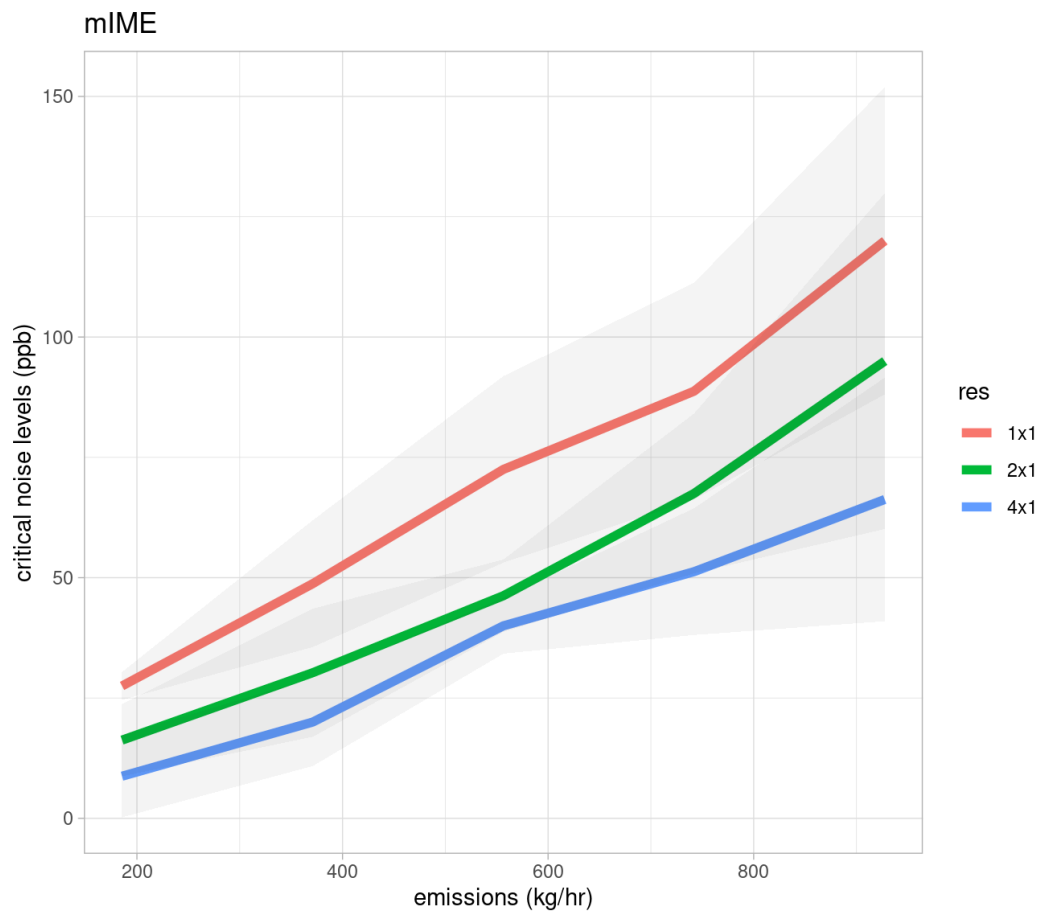
SAT estimates was even more diverged (A.2 and A.3). The plots show a conservative quantification limit of MethaneSAT 1X1 resolution to be around 500 kg/hr or higher at 20 ppb noise and 1000 kg/hr or higher at 40 ppb noise. Since we expect MethaneSAT's noise to be between 20 ppb and 40 ppb, this simple reprojecting method suggested a quantification limit of 500 - 1,000 kg/hr for MethaneSAT at 1X1 resolution under favorable conditions (no interference from clouds or nearby emissions).



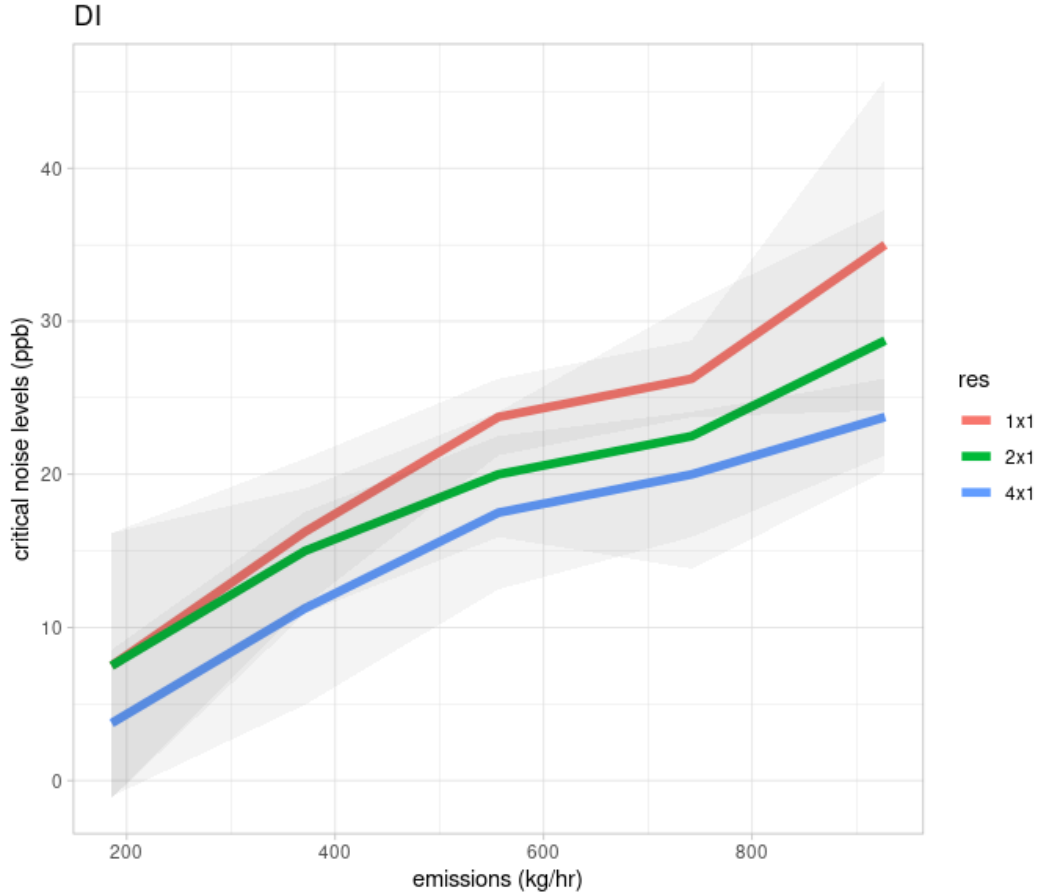
**Figure 3.9:** DI results under 5,000 kg/hr based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis). The York regression suggests a  $0.90 \pm 0.11$  slope and  $-181 \pm 90$  intercept. The SMA regression suggests a  $0.53 \pm 0.08$  slope and  $799 \pm 80$  intercept.

### 3.3.3 THE QUANTIFICATION LIMITS BASED ON CRITICAL NOISE LEVELS (OSSE)

I varied the nominal emission rates from 186, 371, 557, 742, and 928 kg/hr and added Gaussian noise from 5 ppb to 200 ppb for a given nominal emission rate at a 5 ppb increment. I repeated this for three MethaneSAT resolutions: 1x1, 2x1, and 4x1. Figure A.4 is an example of the nominal emission rate of 928 kg/hr. After visually inspecting the emission estimates, I calculated the CVs for each case and identified the critical noise level as shown in Figure A.5. In figure 3.10 and 3.11, I showed the relationship between the critical noise levels as a function of emissions based on the mIME and the DI methods. From the plots, if I assume the critical noise levels represent the quantification limits of MethaneSAT, the conservative quantification limits of the MethaneSAT given 30 ppb of noise are 200, 400, and 500 kg/hr for mIME at the resolutions of 1x1, 2x1, and 4x1, respectively. The quantification limits are 800, 900, and 1100 kg/hr for the DI at resolutions of 1x1, 2x1, and 4x1, respectively.



**Figure 3.10:** The plot shows the critical noise values (ppb) as a function of nominal emission rates (kg/hr) based on mIME. The green line represents the critical values for the 1x1 MethaneSAT grid. The red line represents the critical values for the 2x1 MethaneSAT grid. The blue line represents the critical values for the 4x1 MethaneSAT grid.



**Figure 3.11:** The plot shows the critical noise values (ppb) as a function of nominal emission rates (kg/hr) based on DI. The green line represents the critical values for the 1x1 MethaneSAT grid. The red line represents the critical values for the 2x1 MethaneSAT grid. The blue line represents the critical values for the 4x1 MethaneSAT grid.

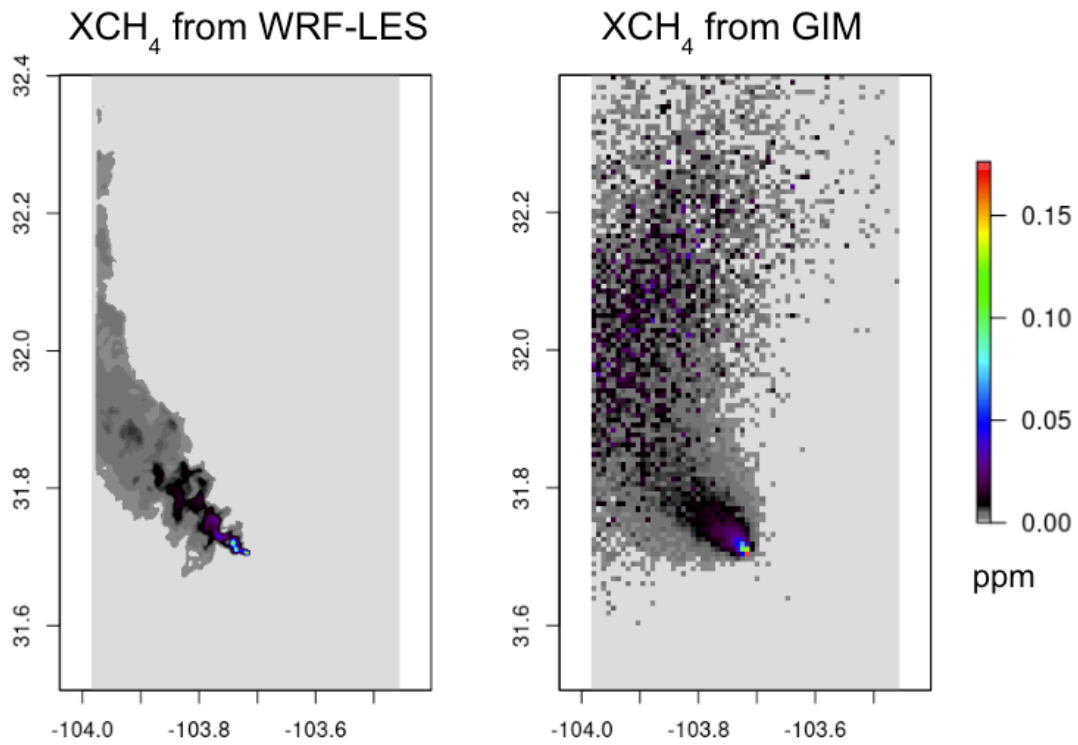
After looking at these OSSE results, we have two sets of quantification limit estimates from the projection in the previous section and the OSSE in this section. The quantification limits for DI at 1x1 in this section (750 kg/hr to 1200 kg/hr) overlap with those from the reprojection section (500 kg/hr to 1000 kg/hr). This agreement gives confidence in our results. However, the quantification limit from the mIME based on our WRF-LES OSSE is around 100 to 300 kg/hr, which is relatively low compared to the DI estimates. This observation suggests that our WRF-LES OSSE framework might underestimate the system errors. By adding only the Gaussian noise to the data,

we cannot fully represent the influence of the noise and background interference, and we do not account for spreading of the plume at the WRF resolution (200m). As a result, I would suggest the best conservative estimate of the point source quantification and quantification limits from these two experiments would come from the DI results. By combining the reprojection and the OSSE ranges, we got the quantification limit for the MethaneSAT at a 1x1 resolution of around 750 kg/hr to 1200 kg/hr.

#### 3.3.4 THE GIM OUTPUTS

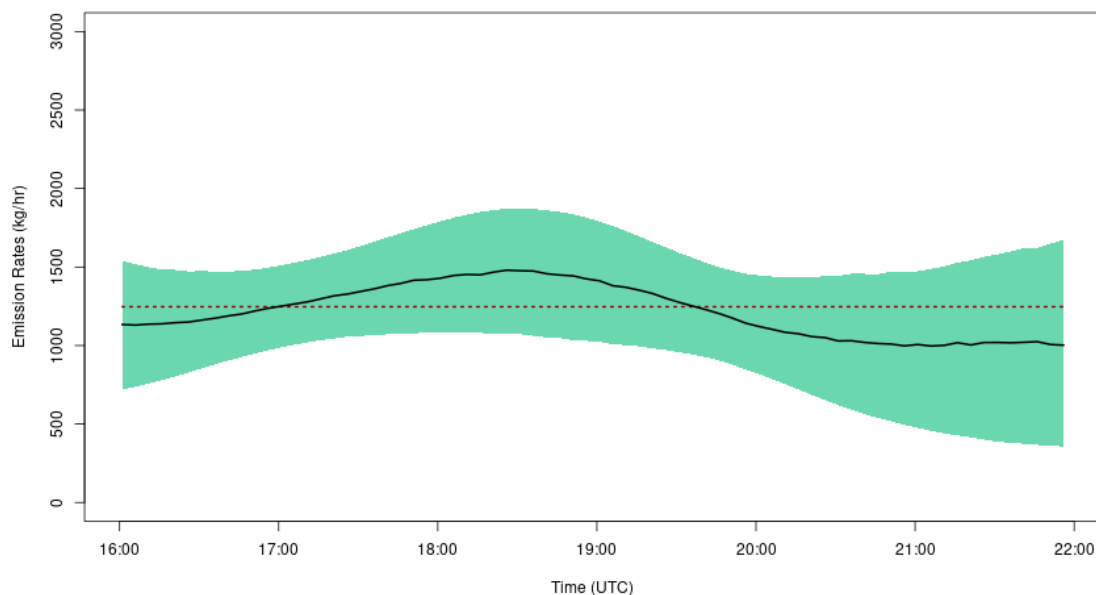
The upcoming segments of this chapter will focus on the outcomes derived from the prototypes of our OSSE framework's applications and explore its potential integration with the GIM framework. After creating a Jacobian matrix from STILT and identifying the region of interest, we can create examples of the entire scene and the point source estimates from the GIM. Instead of comparing the GIM results with real MethaneAIR or MethaneSAT data, we can compare the results with WRF-LES-HRRR results.

## INVERSE $\text{XCH}_4$ FOR THE POINT SOURCES



**Figure 3.12:** The left figure is the XCH<sub>4</sub> from the WRF-LES-HRRR. The right figure is the XCH<sub>4</sub> from the GIM Point Source Quantification, reconstructing the plume from the Jacobian of the WRF field. The source strength has been optimized by minimizing the observed (left) and reconstructed (right).

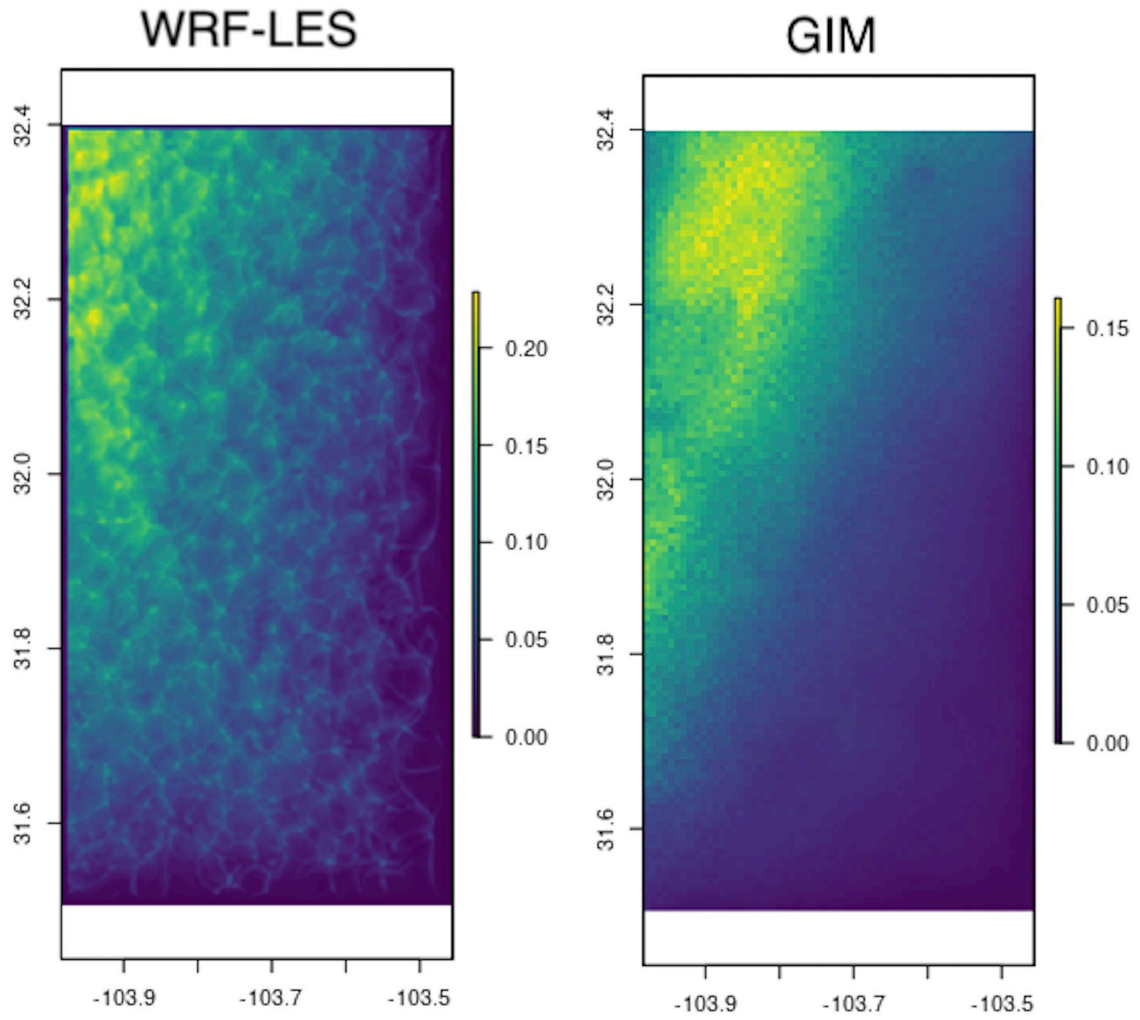




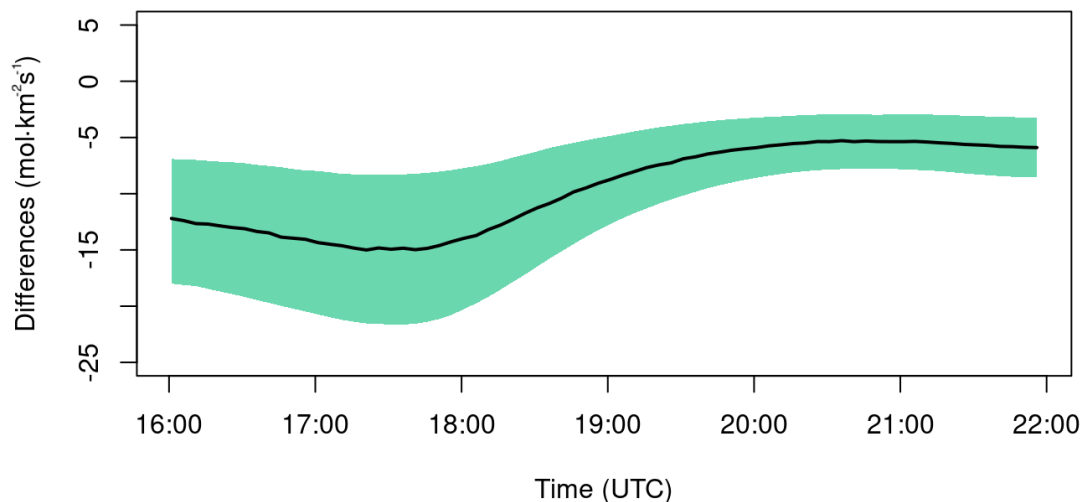
**Figure 3.13:** This figure shows a time series of mean quantified emission from point sources based on the GIM framework as a function of time in the simulation. We found consistent emission estimates of 1247 kg/hr, similar to the nominal emission rate.

I first tried to estimate the point source emission rates based on the GIM framework and compared the results to the nominal emission of 1247 kg/hr. A snapshot of the GIM-generated  $\text{XCH}_4$  field compared to the WRF-LES  $\text{XCH}_4$  field is shown in Figure 3.12. Since the WRF-LES took hours to spin up the simulation, I only averaged the results between 16:00 - 20:00 (6 hours after the starting time) and got the emission estimate of 1247 (1177, 1362) kg/hr. The entire time series of the estimated emission from the GIM can be found in Figure 3.13. This prototype suggests the feasibility of the GIM for point source estimates, a close to “free” point source byproduct of the GIM with only a few multiplications and summation away. This could be a shortcut allowing us to estimate the point source emissions without running the other point source quantification algorithms. However, additional studies are needed to understand this method’s limitations better.

## INVERSE $\text{XCH}_4$ FOR THE SCENES



**Figure 3.14:** The left figure is the  $\text{XCH}_4$  from the WRF-LES based on a random Gaussian noise background of  $1.0 \pm 0.1$  kg/hr per pixel. The right figure is the  $\text{XCH}_4$  from the GIM with the same emission field.



**Figure 3.15:** The mean difference between the GIM and WRF-LES from various OSSE runs. Since WRF-LES has more details from the eddies, the magnitudes are higher than the patterns suggested by the GIM. Overall, the difference is around  $-9.7$  ( $-14.0, -5.3$ )  $\text{mol} \cdot \text{km}^{-2} \cdot \text{s}^{-1}$ .

I later tried to recreate a random Gaussian noise background of  $1.0 \pm 0.1$  kg/hr per pixel ( $25 \pm 2.5$  kg/hr per  $\text{km}^2$ ) based on the GIM framework and compared the results to the WRF-LES-HRRR simulations (Figure 3.14). In Figure 3.15, I combined the results between 16:00 - 20:00 UTC from Gaussian noise and Gaussian correlation length backgrounds and got the mean difference of around  $-9.7$  ( $-14.0, -5.3$ )  $\text{mol} \cdot \text{km}^{-2} \cdot \text{s}^{-1}$ . Since the maximum value of the WRF-LES background is around  $23 \text{ mol} \cdot \text{km}^{-2} \cdot \text{s}^{-1}$ , we expect underestimation of the peak could be around 42 (23, 60) %

These two prototypes show that the GIM estimated  $\text{XCH}_4$  for the entire scene or the point source tends to spread out more depending on the resolution of the GIM. So far, if we believe that the WRF-LES represents the actual atmosphere, our results are promising indicators of the GIM performance. The GIM framework should work with the following caveats and limitations. We have made several assumptions, including the noise being Gaussian and the albedo effects being

removed completely. Types of meteorological products we can access might also affect our results. In this experiment, we used the HRRR wind product to drive the WRF-LES and the GIM. By sharing the same meteorological product, our results are not independent. If the meteorological product we use doesn't represent the region of the atmosphere that is of interest to us well, the difference between the GIM and the actual activities in the atmosphere will be greater.

By verifying the GIM's ability to work for both point and area sources, we show that, at least in principle, calculating the GIM for the entire area can be expedited by using the GIM Jacobian and computing point source emission rates as above instead of relying on the DI or mIME. This is a crucial step in bridging the point source and area source quantification of MethaneAIR and MethaneSAT products.

### 3.4 CONCLUSIONS

Due to limited access to MethaneSAT data, we applied the OSSE concept to help us better understand our quantification methods for point sources and to predict the performance of MethaneSAT. Through direct observation, we could determine the potential sources of errors, including plume morphology, cloud effects, and pixel orientation effects. By projecting MethaneAIR data onto the MethaneSAT grid, with a simplified MethaneSAT noise model, and by running WRF-LES for the OSSE before applying quantification algorithms on the WRF-LES outputs, we come up with quantification limits of the MethaneSAT. The MethaneSAT quantification limits from these two approaches overlap within the range of 750 to 1200 kg/hr. Additionally, we evaluated the performance of the GIM within the OSSE framework. Despite the preliminary nature of our experiments in this chapter, we demonstrate that the GIM shows promise in estimating emissions from both point and area sources, potentially exhibiting lesser emission bias compared to methods like IME or DI. These investigations enable us to anticipate the limitations and performance of

MethaneAIR and MethaneSAT's methane emission point source quantification.

# 4

## Conclusions

This thesis represents a significant step in understanding methane point source emission quantification, particularly for the MethaneAIR and MethaneSAT sensors. Although the instruments are unique, we can learn generally applicable lessons. The high-resolution satellite data in Chapter 1 paved the path for the later chapters, showing the applicability of LES tools and the limitations that led to two other methods for quantitatively estimating emissions from point sources.

By exploring and developing various quantification methods and considering factors such as

source characteristics and surrounding conditions in the first and second chapters, we have enhanced our ability to estimate methane emissions comprehensively and provided the basis for appropriately choosing the quantification approaches depending on the situation. The findings from controlled release experiments in chapter two and detection limit analysis in chapter three provide valuable insights into the performance and limitations of our methods, guiding the development of algorithms for future satellite missions. Additionally, the successful application of these techniques underscores their broader relevance beyond our specific projects, offering valuable tools for methane emission assessment in diverse scenarios.

Overall, this research contributes to a deeper understanding of methane emissions and lays the groundwork for more effective and informed decision-making in mitigating methane's environmental impact.



## Additional Information

### A.1 MICROHISTORY OF THE ANADARKO BASIN

#### A.1.1 THE STORY

Professor Emma Rothschild and Steven Wofsy collaborate through the Salata Institute on a project centered around the utilization of remote sensing data for comprehensive insights into methane release locations. Their initiative seeks to combine micro-histories of methane emission



sites with satellite and aircraft imaging technologies. The objective is to gain a deeper understanding of activities that are pivotal to global greenhouse gas emissions and refine our understanding of methane reduction strategies, considering their social implications. The primary focus will revolve around the data from the MethaneAIR, a remote-sensing instrument measuring methane concentrations across expansive landscapes with outstanding spatial resolution and precision. The collaboration introduced me to micro-histories, a historical genre focused on investigating small units, such as events, communities, individuals, or settlements, closely linked with social and cultural history. The following section contains the draft intended for publication on the Joint Center for History and Economics' website.

#### A.1.2 THE DRAFT

As an environmental engineer by training, I will start writing this microhistory with a hypothesis and a method for this writing piece. Microhistory shall allow us to dive into certain stories in-depth and give us valuable contexts of the larger story. This essay will explore the stories related to a particular site where our instrument, MethaneAIR, detected a large methane leak.

The site was chosen based on cross-referencing to the original 1800 Histories map ([link](#)) discovered by Lauvaux et al., in 2022, published a list of major methane emission sources worldwide. One of the sources was right around the corner from an abandoned drilled and plugged site of Spess Oil Company. The emission was reported to be  $38.8 \pm 17.18$  t/hr. Coincidentally, MethaneAIR observed the same region with a similar estimated emission. The location was at  $(-98.015^{\circ}\text{W}, 35.687^{\circ}\text{N})$ . Anyhow, let's go back in time and look at what happened to this Spess Oil Company drilled and plugged site.

Against the backdrop of the American prairies, the microhistory of a drilled and plugged oil and natural gas well unfolds in the heart of the Anadarko basin in Oklahoma. It was the mid-20th century, and the oil boom echoed through the state, promising prosperity to those who dared to ven-

ture into the exploration of hidden treasures beneath the surface.

Our story begins in Canadian County, where whispers of wealth beneath the soil caught the attention of many. After Frank Spess graduated from Mannford High School in 1942, he assisted his father in the oil and gas fields and, with help from his brother, established his own company, Spess Oil Company.

Since the company's formation, more than 146 Spess Oil Company facilities have been built in Oklahoma [citation]. Drilling rigs rose, striking black gold. Oil and natural gas surged to the surface, passing through the wellheads, pipes, and valves, eventually reaching the extensive pipeline system across the region. Several jobs were created, businesses flourished, and the community thrived.

However, prosperity came with its challenges. Although the basin's production has consistently increased since the 20th century, some wells have to be out of commission due to their lifetimes. After 20 to 30 years, wells complete their production cycles. As the wells' production gradually declined, the decision to plug them became inevitable. The once-symbolic drilling rigs of economic promise were dismantled, and the wellheads were capped. Over 132 facilities were abandoned, with unknown follow-ups.

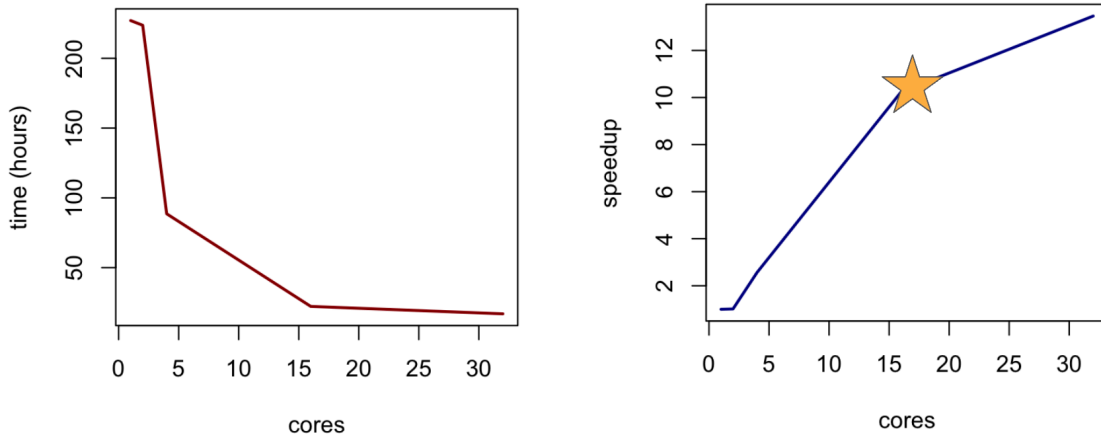
There was not much data after the abandonment of the wells. Still, we know that the abandoned sites were known to contribute significant methane emissions to the atmosphere.

Some of Spess Oil Company's facility sites also face legal challenges. The town of Prague, OK, filed an earthquake damage suit against the company after a 5.7 magnitude earthquake struck near the town in 2011.

While the site has probably still been emitting methane, the microhistory of Spess Oil Company's abandoned site encapsulates the complex interplay between prosperity, environmental impact, and legal entanglements, highlighting the nuanced legacy of oil and history in the United States.

## A.2 WRF BENCHMARKING

In addition to modifying the WRF's Fortran-sourced code, I did a benchmark to optimize the number of WRF's cores so that I could pick the right number for the most efficient performance. The WRF run that I did was for a WRF run in the Permian with an inner domain of  $101 \times 101$  pixel<sup>2</sup>. Each pixel was 111.11 m by 111.11 m.



**Figure A.1:** WRF's runtime as a function of cores (left) and the speedups as a function of cores

## A.3 CALCULATIONS

### A.3.1 MIME EFFECTIVE WIND SPEEDS

The effective wind speeds used in the MIME calculation came from the relationship proposed by Varon et al.<sup>53</sup>,  $U_{eff} = \alpha \cdot \log(10 \text{ m wind}) + 0.6$  where  $\alpha$  is between 0.9-1.1. We took advantage of the LES runs by introducing the LES-specific effective wind  $U_{adaptive, eff}$  for each time step.  $U_{adaptive, eff}$  is defined as  $\frac{Q \cdot L}{\overline{ME}}$ . Because we know all the terms in this equation from the LES, we can calculate the

true  $U_{adaptive, eff}$  that gives the best estimate of  $Q$ .

By calculating the  $U_{adaptive, eff}$  for all the time steps of interest, we end up with multiple pairs of  $U_{adaptive, eff}$  and  $U$ , which can be fitted into a linear regression function of the form  $U_{eff} = a \cdot \log(10 \text{ m wind}) + b$  with unique  $a$  and  $b$  coefficients.

The LES-specific relationship can introduce overfitting results when the data points are limited, and wind speeds are low. If we overfit the relationship, we cannot justify the predicted effective wind speeds and may come up with biased emission estimates. Since the wind speeds during the controlled release experiments were lower than 5 m/s for most of the time, we decided to use the coefficients in <sup>53</sup> for the sake of simplicity.

Though the coefficients <sup>53</sup> derived from idealized LES with varying meteorological conditions are the most comprehensive IME coefficients available, we should be aware of ambient weather and turbulence conditions that may affect the relationship's universality.

### A.3.2 DIVERGENCE INTEGRAL

The divergence integral method is based on the integral form of the continuity equation, which states that the net production of  $CH_4$  inside an enclosing surface,  $\int_V P$ , through an enclosing surface is given by

$$\int_V P = \int_V \vec{\nabla} \cdot c \vec{v} + \int_V \frac{dm}{dt} = 0 \quad (A.1)$$

where  $c$  is the methane concentration,  $v$  is the wind speed, and  $\langle \frac{\partial m}{\partial t} \rangle$  is the change in mass of methane with time within volume  $V$ .  $\int_V P = 0$  since there is no *in situ* production. The flux divergence can be decomposed into two terms:

$$\nabla \cdot cv = v \cdot \nabla c + c \cdot \nabla v \quad (A.2)$$

Conley et al.<sup>6</sup> demonstrate that for wind speeds over 1 m/s and fluxes above 200 kg/hr, the wind divergence term (2nd term in Eq. A.2) near a source is 1% or less of the concentration gradient term (1st term in Eq. A.2); therefore, we can neglect the 2nd term for our application. To isolate the XCH<sub>4</sub> enhancement relative to the surroundings, we subtract the mean column XCH<sub>4</sub> along the rectangle from the divergence term, which does not alter  $\nabla c$ , because  $\nabla \langle c \rangle = 0$

$$c' = c - \langle c \rangle \quad (\text{A.3})$$

$$\nabla c = \nabla (\langle c \rangle + c') = \nabla c' \quad (\text{A.4})$$

where  $c'$  is the XCH<sub>4</sub> enhancement and  $\langle c \rangle$  is the mean XCH<sub>4</sub> around the rectangle. The  $c'$  values in Eq. A.3 equation can be both positive and negative. When there is a positive flux through the surface, the positive  $c'$  values outweigh the negative  $c'$  values. There are some instances where the flux through the surface is negative (i.e., Figure 2.2d, 420m), and those values are averaged into the total flux as well. In most cases, these reflect the influence of boundary layer eddies.

For analysis of the total column data from MethaneAIR in the vicinity of an emission source, we assume that methane concentrations above the planetary boundary layer are equal to unperturbed values outside the bounding surface. Then, the horizontal gradients of XCH<sub>4</sub> arise entirely in the boundary layer, and we may take  $v$  as the pressure-weighted mean boundary layer wind speed and direction.

We then use Gauss's theorem to relate the volume integral to a surface integral around the cuboid:

$$\begin{aligned}
0 &= \int_V v \cdot \nabla c' dV + \left\langle \frac{\partial m}{\partial t} \right\rangle \\
&= -\Phi_{surf} + \oint_{\partial V=S} v \cdot \hat{n} c' dS + \left\langle \frac{\partial m}{\partial t} \right\rangle \\
&= -\Phi_{surf} + \oint_{\partial V=S} v \cdot \hat{n} ((XCH_4 - \langle XCH_4 \rangle_{rect.}) * n_{column} * M_{CH_4}) dS + \left\langle \frac{\partial m}{\partial t} \right\rangle \\
\Phi_{surf} &= \oint_{\partial V=S} v \cdot \hat{n} ((XCH_4 - \langle XCH_4 \rangle_{rect.}) * n_{column} * M_{CH_4}) dS
\end{aligned} \tag{A.5}$$

where  $\Phi_{surf}$  is the flux into the volume from the land surface,  $S$  is the surface enclosing the cuboid,  $\hat{n}$  is an outward pointing unit vector normal to the surface,  $n_{column}$  is the moles of air in the column based on the surface pressure from HRRR and  $M_{CH_4}$  is the molar mass of methane. For a volume the size of a single plume, the term  $\langle \frac{\partial m}{\partial t} \rangle$  should be insignificant, assuming that the plume is in a steady state and no mass is building up in the volume. We assume that the vertical flux through the top of the cuboid at the planetary boundary layer (PBL) height is zero, consistent with neglecting the divergence of the horizontal wind. Thus, the horizontal flux through the surface is balanced by and therefore equal to the flux through the bottom of the cuboid from the surface source,  $\Phi_{surf}$  (kg/hr), we wish to measure. Based on Eq. A.5, the flux from the source is computed from observed  $XCH_4$  as follows:

$$\Phi_{surf} = \sum_{\text{around rect}} (XCH_{4_i} - \langle XCH_4 \rangle_{rect.}) \cdot n_{column} \cdot M_{CH_4} \cdot v \cdot \hat{n} \cdot \Delta l \tag{A.6}$$

where  $\Delta l$  is the distance between successive  $XCH_4$  measurements. We average the fluxes calculated over a sequence of rectangles extending downwind of the source, spanning several eddy scales. The box grows by 1 pixel in each step except for the upwind direction, where it moves over by 1/4 pixel. The upwind boundary was selected to be close to the upwind of the source using the moment of inertia of the plume (see Fig. 2)

### A.3.3 GRIDDING BY NEAREST NEIGHBOR AND PHYSICS-BASED METHODS

The nearest neighbor method for gridding used the R function `binMean` from the `oce` package to place the georeferenced  $\text{XCH}_4$  data onto a predefined  $10\text{m} \times 10\text{m}$  grid. The retrieved  $\text{XCH}_4$  values have a standard deviation of about 80 ppb. Since the point spread function is about 12 m wide across the track, and 35 m along track, the grid of native pixels spatially oversamples the image on the ground. Therefore, we spatially filtered the retrieved  $\text{XCH}_4$  data to create the gridded product in two steps. First, the holes in the initial gridded field were filled using a  $3 \times 3$  averaging matrix. Then we further spatially filtered this field by a Gaussian filter with full width at half maximum of 1.7 pixels before inputting the  $\text{XCH}_4$  values to our point source quantification methods. The resulting oversampled gridded product has a standard deviation of 40 ppb and a spatial correlation length of about 20 m, comparable to the length scale of the point spread function. The nearest-neighbor gridding method was used for the initial (blinded) input from Harvard to Stanford.

The improved estimates after unblinding used the “physics-based” gridding method of Sun et al.<sup>47</sup>.

### A.3.4 YORK REGRESSION

When errors in the observables are present on both the x-axis and the y-axis, it is recommended to use York regression and other equations derived from York to determine the slope, y-intercept, and errors of the least-square estimation solutions. The full equations and derivations can be found on York<sup>57</sup> and York et al.<sup>58</sup>

### A.3.5 MEAN PERCENTAGE ERROR

$$MPE = \frac{100\%}{n} \sum_{t=1}^n \frac{a_t - f_t}{a_t} \quad (\text{A.7})$$

where  $a_t$  is the actual value of the quantity being forecast,  $f_t$  is the forecast, and  $n$  is the number of different times for which the variable is forecast.

## A.4 ADDITIONAL RESULTS

### A.4.1 MISCELLANEOUS METHANE SOURCES

#### UNLIT FLARE

MethaneAIR observed the unlit flare near the release site 21 times with repeated overpasses on 30 July and 3 August 2021. As shown in Figure A.11, the flare emission rates ranged from 500 kg/hr to 2000 kg/hr as calculated by the mIME and DI methods. Based on this comparison and the comparison with the controlled releases, we estimated the detection limit of the DI method to be approximately 500 kg/hr.

#### MIVIDA GAS PROCESSING PLANT

We quantified emissions from a large consistent emitter, the MiVida Gas Processing Plant, in Barstow, TX, 9 times with repeated overpasses on 6 and 9 August 2021. Typical emission rates were 2000 kg/hr with 30% uncertainty estimate based on the controlled releases experiment results (Figure A.12). Several emission points could be identified within the boundaries of this plant.

#### THE PIPELINE LEAK IN NEW MEXICO

We discovered a large pipeline leak at 103.697, 32.366 in New Mexico. MethaneAIR detected the leak on 9 August 2021, not present on the earlier flight on 6 August 2021. Using the mIME and DI methods, we estimate the emission rate to be approximately 5000 kg/hr with 30% uncertainty estimate based on the controlled releases experiment results (Figure A.12).



We attributed the leak to a gathering pipeline emission event at the location, confirmed by emission incident reports to the New Mexico Oil Conservation Division on August 26, 2021, which identified the cause as a rupture at a weld along a gathering pipeline segment<sup>32</sup>. Based on this report, we estimated a methane emission rate of 8200 kg/h over an 18-hour reporting period, assuming 80% methane content in the gathered natural gas. With simple extrapolation of the lower estimate of 5000 kg/hr from the flight on August 9 until the leak was fixed on August 24, the total methane emission could be more than  $1.8 \times 10^6$  kg (1,800 metric tons) over the 15 days.

#### A.4.2 METHANEAIR AND METHANESAT COMPARISON

##### THE SPECIFICATIONS

**Table A.1:** Specification comparison for MethaneAIR and MethaneSAT

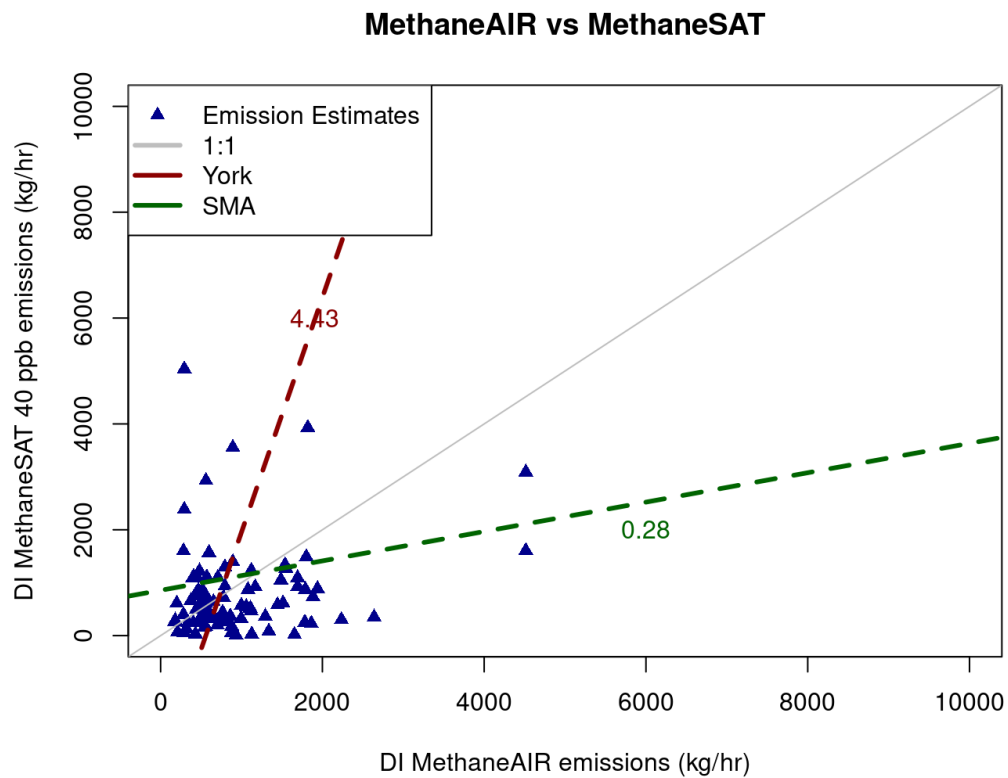
| Specification                         | MethaneAIR   | MethaneSAT |
|---------------------------------------|--------------|------------|
| O <sub>2</sub> passband (nm)          | 1237–1319    | 1249–1305  |
| O <sub>2</sub> dispersion (nm/pixel)  | 0.08         | 0.06       |
| O <sub>2</sub> spectral FWHM (nm)     | 0.23         | 0.18       |
| CH <sub>4</sub> passband (nm)         | 1592–1678*   | 1598–1683  |
| CH <sub>4</sub> dispersion (nm/pixel) | 0.10         | 0.08       |
| CH <sub>4</sub> spectral FWHM (nm)    | 0.28         | 0.24       |
| Field of view (°)                     | 23.7         | 21.3       |
| Cross-track pixel <sup>†</sup> (m)    | ~ 5 at 12 km | ~ 108      |
| Along-track pixel <sup>†</sup> (m)    | ~ 25         | ~ 400      |
| Point spread function (pixels)        | 2.5          | 1.8        |
| Single pixel SNR <sup>‡</sup>         | ~ 95         | ~ 190      |

\*MethaneAIR uses an InGaAs detector with reduced QE beyond ~ 1660 nm

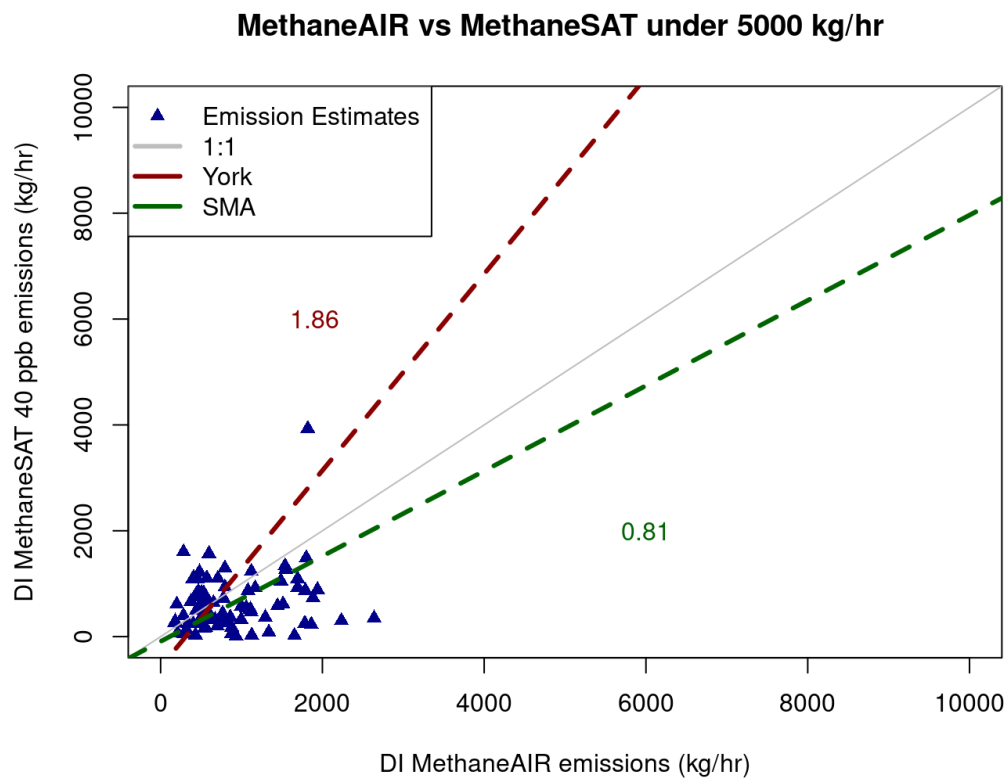
<sup>†</sup>Distance between pixel centers at nadir

<sup>‡</sup>CH<sub>4</sub> band with nominal radiance of  $1.4 \times 10^{13}$  photons cm<sup>-2</sup> nm<sup>-1</sup> sec<sup>-1</sup> sr<sup>-1</sup>

## THE ANALYSIS

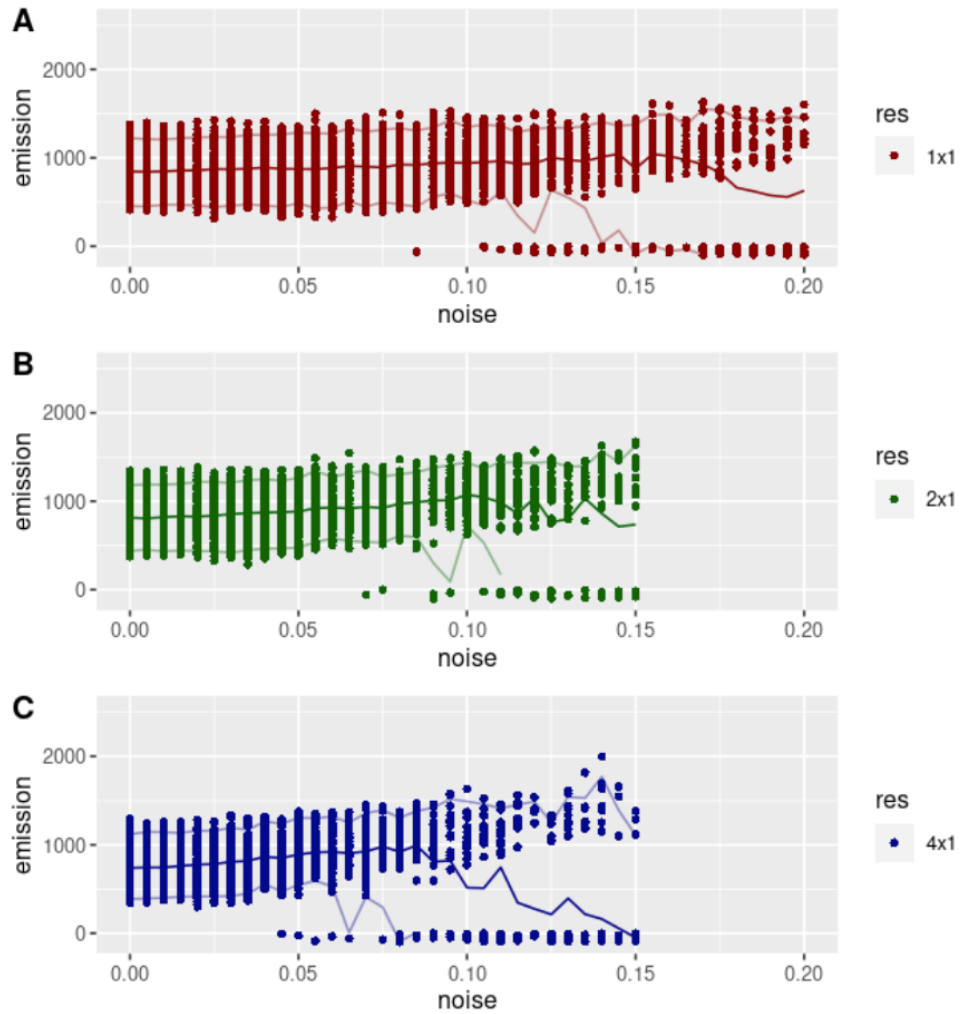


**Figure A.2:** DI results based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis) when the MethaneSAT noise is 40 ppb.

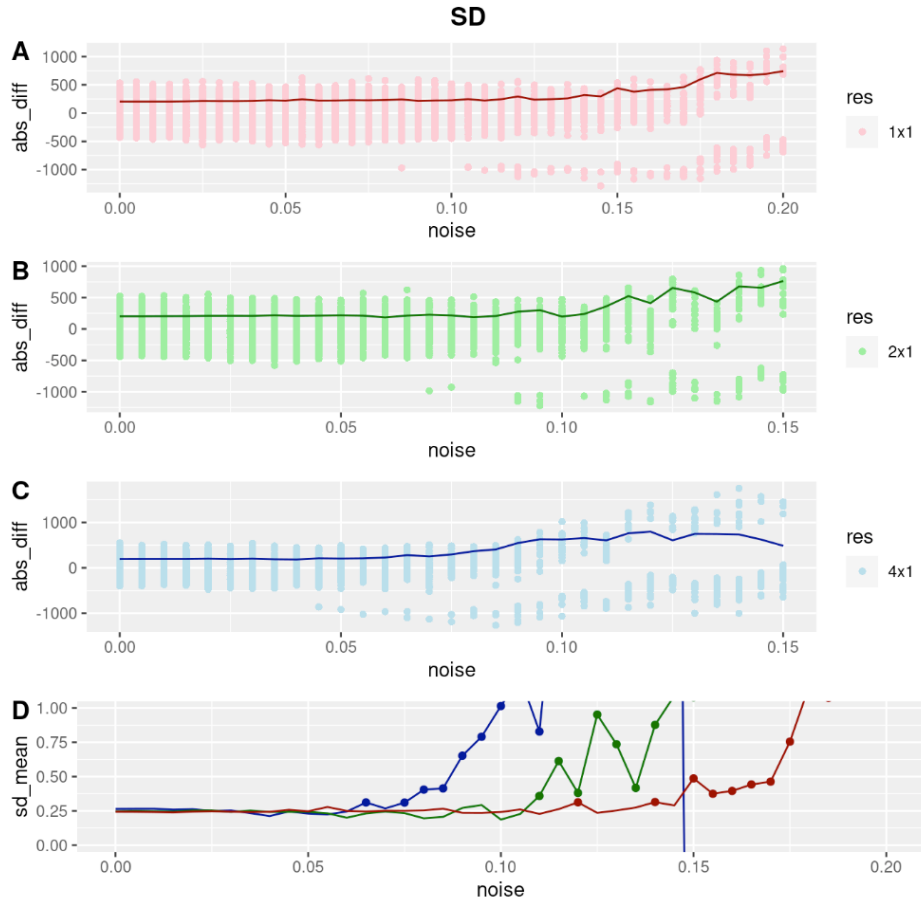


**Figure A.3:** DI results under 5,000 kg/hr based on the MethaneAIR data (x-axis) and projected MethaneAIR data onto the MethaneSAT (y-axis) when the MethaneSAT noise is 40 ppb.

## Emission: 928 kg/hr



**Figure A.4:** The nominal emission for this figure is 928 kg/hr. A. The panel shows the estimated emission using the light IME method as a function of noise at 1x1 MethaneSAT resolution. B. The panel shows the estimated emission using the light IME method as a function of noise at 2x1 MethaneSAT resolution. C. The panel shows the estimated emission using the light IME method as a function of noise at 4x1 MethaneSAT resolution.



**Figure A.5:** The nominal emission for this figure is 928 kg/hr. A. The panel shows the absolute difference between the nominal emissions and the emissions from the light IME method as a function of noise at 1x1 MethaneSAT resolution (red). B. The panel shows the absolute difference as a function of noise at 2x1 MethaneSAT resolution (green). C. The panel shows the absolute difference as a function of noise at 4x1 MethaneSAT resolution (blue). D. The coefficient of variation as a function of the noise.

#### A.4.3 BLINDED VOLUME-CONTROLLED RELEASE EXPERIMENTS

The release rate was measured by a Sierra 640i QuadraTherm meter. Stanford performed an extensive set of meter intercomparison tests to characterize uncertainty in the QuadraTherm 640i meters (Rutherford et al. <sup>37</sup>). Based on these intercomparison tests, a bias of up to 6% can be observed

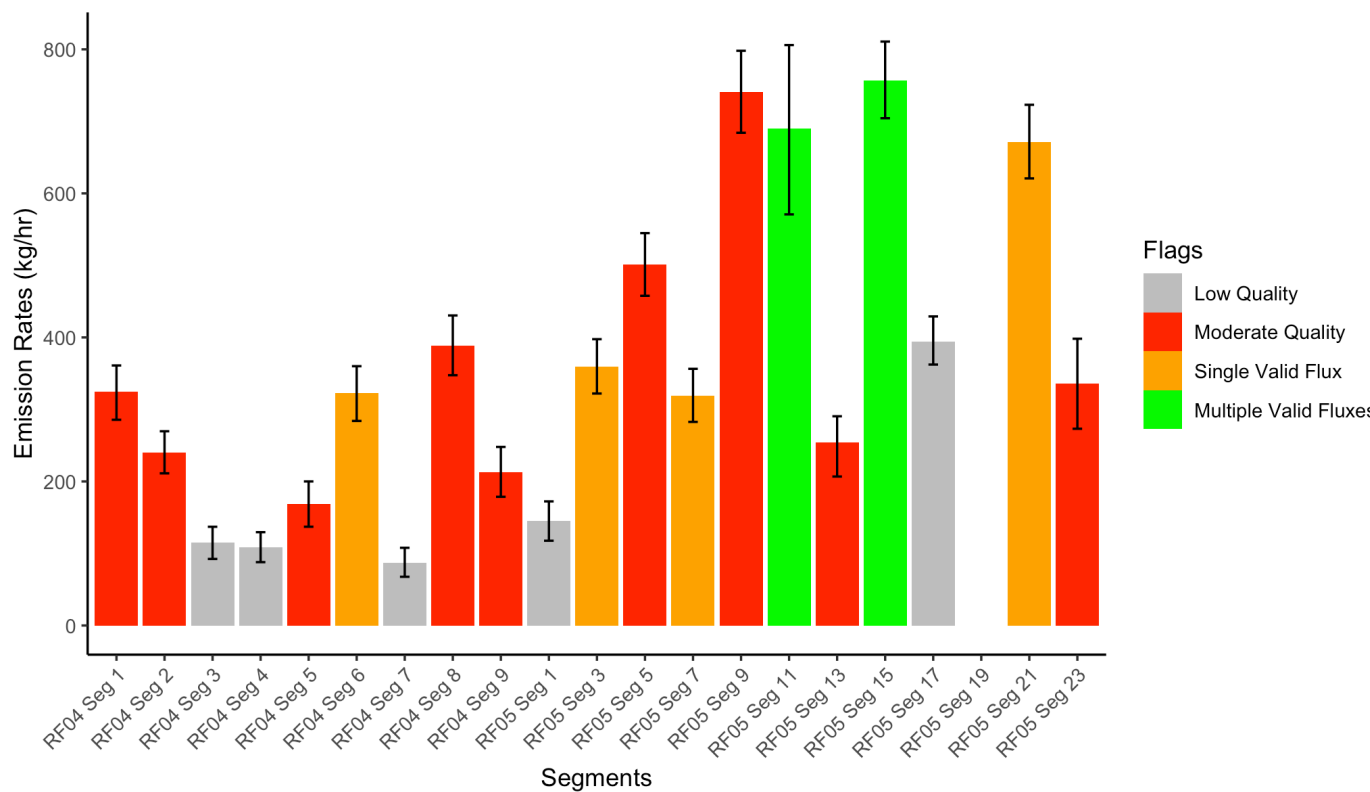
between QuadraTherm 640i meters. Error in metered controlled release volumes is characterized using a Monte Carlo analysis that also accounts for uncertainty in gas composition.

On seven occasions during the missions, the Stanford team informed the MethaneAIR team of the wind direction at the site since winds were light and variable, as reported by nearby ASOS systems. The MethaneAIR team considered reorienting the flight track to capture the plumes due to the wind conditions, but no changes in the flight track were required. On two occasions, Stanford confirmed that local winds were not zero. Although null winds were not desired for this first test of MethaneAIR, since the winds were not zero, the decision was made to complete the surveys as planned.

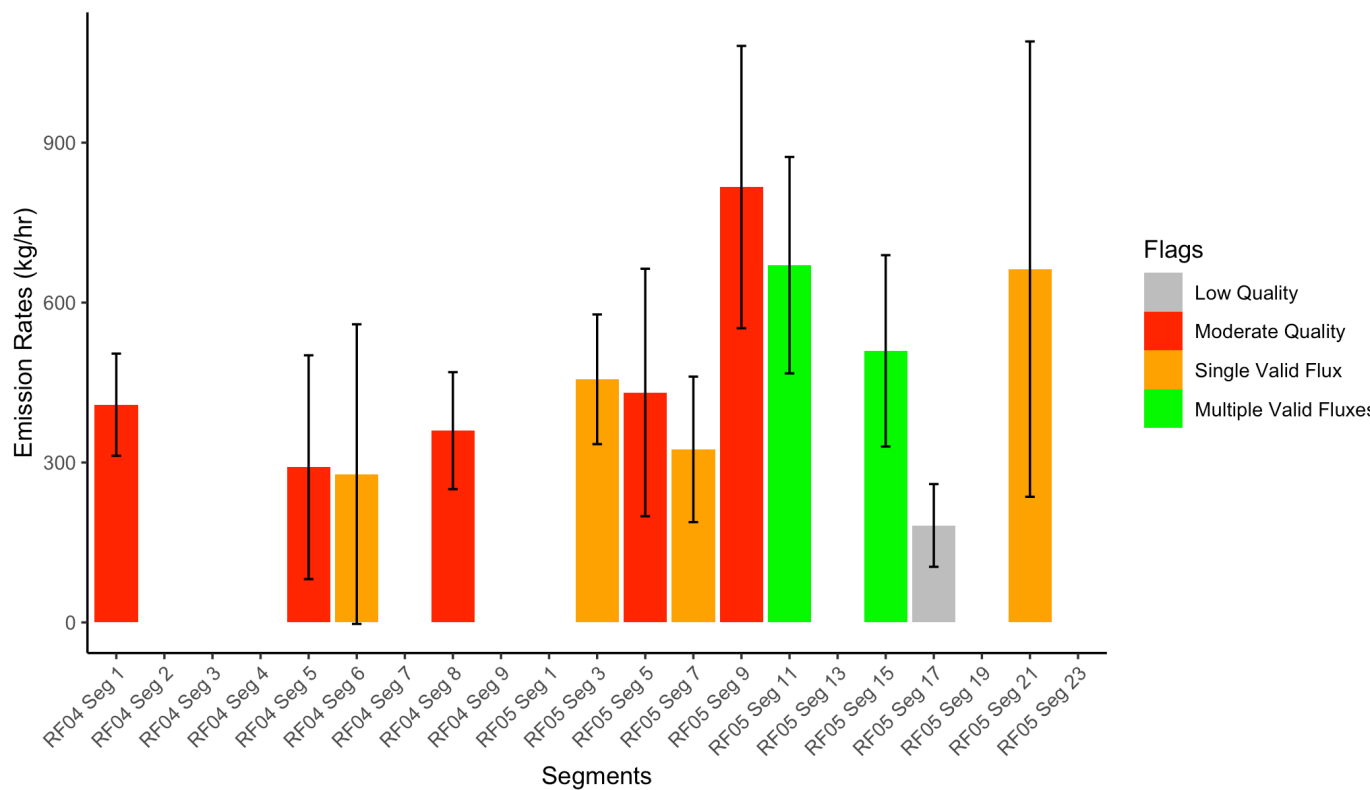
For the analysis, the MethaneAIR team used the York fit, which accounts for uncertainties in both the X and the Y coordinates, instead of the commonly used OLS, which assumes that uncertainties are significant only in the Y coordinate. In this experiment, the uncertainties in the metered flow rates are not negligible, and the error estimates are available from the Stanford team. Hence, the York fit was selected to approximate the desired Maximum Likelihood Estimator for the relationship between the Harvard values and the Stanford emission rates.

The MethaneAIR team conducted tests on the OLS to verify whether the OLS is an appropriate regression choice. The slope of the OLS line relating the Harvard mIME emissions to the Stanford single-blind volume-controlled release rates is 0.78 for the original submission. Based on the heteroskedasticity test, this OLS fit violates the assumptions for linear regression (p-value 0.006). The violation indicates a significant relationship between the residuals and the predictor (Stanford emission rates). The Non-Constant Error Variance test (ncvTest) gave a p-value of 0.14, suggesting violation at the 84% level. These tests support the application of the York fit for the 2021 results.

Based on the 2021 experiment ( $N = 21$ ), the false positive rate is  $\approx 10\%$  and the false negative is  $\approx 5\%$ . These rates can not be generalized to the instrument capability since we have a limited number of data points, and the false positive/negative rates depend on the scene of interest.



**Figure A.6:** Post-unblinding best estimates of methane emissions from the controlled release experiments during RF04 and RF05 using the mIME method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16.



**Figure A.7:** Post-unblinding methane emission estimates from the single-blind volume-controlled release experiments during RF04 and RF05 using the DI method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16.



**Table A.2:** RF04 and RF05 combined best estimate results from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 (RF04) and 3 August 2021 (RF05). The times are in UTC. The emission rates, lower bounds, and upper bounds are in kg/hr. The blinded, blinded lower, and blinded upper columns were reported to the Stanford team as MethaneAIR initial emission estimates on 1 February 2022. The unblinded, unblinded lower, and unblinded upper columns were added after the unblinding process. The upper and lower bound estimates were calculated using the method described in Section 2.3.3.

| Seg | Timestamp (UTC) | Blinded | Blinded Lower | Blinded Upper | Unblinded | Unblinded Lower | Unblinded Upper | Flagged |
|-----|-----------------|---------|---------------|---------------|-----------|-----------------|-----------------|---------|
| 1   | 30/7/21 15:41   | 321.74  | 179.30        | 471.71        | 324.55    | 285.64          | 361.07          | No      |
| 2   | 30/7/21 15:52   | 232.19  | 133.63        | 338.38        | 240.14    | 211.28          | 269.73          | No      |
| 3   | 30/7/21 16:07   | 109.22  | 62.73         | 160.03        | 114.76    | 92.43           | 137.06          | Yes     |
| 4   | 30/7/21 16:22   | 102.76  | 59.80         | 151.54        | 108.16    | 87.96           | 129.54          | Yes     |
| 5   | 30/7/21 16:36   | 152.31  | 90.07         | 222.33        | 168.53    | 137.12          | 200.03          | No      |
| 6   | 30/7/21 16:51   | 225.42  | 99.69         | 392.61        | 322.45    | 284.06          | 360.08          | No      |
| 7   | 30/7/21 17:06   | 82.21   | 33.30         | 136.95        | 87.43     | 67.63           | 107.83          | Yes     |
| 8   | 30/7/21 17:22   | 362.67  | 138.79        | 613.80        | 388.06    | 347.49          | 430.51          | No      |
| 9   | 30/7/21 18:08   | 205.48  | 89.54         | 331.16        | 213.25    | 178.73          | 247.93          | Yes     |
| 1   | 3/8/21 15:53    | 140.85  | 76.46         | 211.86        | 140.85    | 76.46           | 211.86          | Yes     |
| 3   | 3/8/21 16:11    | 344.58  | 189.55        | 512.42        | 344.58    | 189.55          | 512.42          | No      |
| 5   | 3/8/21 16:30    | 472.22  | 280.28        | 682.46        | 472.22    | 280.28          | 682.46          | No      |
| 7   | 3/8/21 16:49    | 299.01  | 197.33        | 415.01        | 299.01    | 197.33          | 415.01          | No      |
| 9   | 3/8/21 17:06    | 694.32  | 537.16        | 859.69        | 694.32    | 537.16          | 859.69          | No      |
| 11  | 3/8/21 17:24    | 648.93  | 521.87        | 791.07        | 648.93    | 521.87          | 791.07          | No      |
| 13  | 3/8/21 17:41    | 243.47  | 166.76        | 326.58        | 243.47    | 166.76          | 326.58          | Yes     |
| 15  | 3/8/21 17:59    | 730.75  | 494.07        | 987.37        | 730.75    | 494.07          | 987.37          | No      |
| 17  | 3/8/21 18:15    | 385.89  | 288.5         | 487.62        | 385.89    | 288.50          | 487.62          | Yes     |
| 19  | 3/8/21 18:35    | NA      | NA            | NA            | NA        | NA              | NA              | No      |
| 21  | 3/8/21 18:53    | 525.67  | 342.93        | 727.14        | 525.67    | 342.93          | 727.14          | No      |
| 23  | 3/8/21 19:11    | 173.21  | 118.33        | 231.18        | 173.21    | 118.33          | 231.18          | No      |

**Table A.3:** DI results from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 and 3 August 2021. The times are in UTC. The emission rates, lower bounds, and upper bounds are in kg/hr.

| Seg | Timestamp (UTC) | Unblinded Estimate | Lower Bound | Upper Bound |
|-----|-----------------|--------------------|-------------|-------------|
| 1   | 30/7/21 15:41   | 408.43             | 312.61      | 504.25      |
| 2   | 30/7/21 15:52   | NA                 | NA          | NA          |
| 3   | 30/7/21 16:07   | NA                 | NA          | NA          |
| 4   | 30/7/21 16:22   | NA                 | NA          | NA          |
| 5   | 30/7/21 16:36   | 291.14             | 81.20       | 501.09      |
| 6   | 30/7/21 16:51   | 278.21             | -2.83       | 559.24      |
| 7   | 30/7/21 17:06   | NA                 | NA          | NA          |
| 8   | 30/7/21 17:22   | 359.74             | 250.00      | 469.49      |
| 9   | 30/7/21 18:08   | NA                 | NA          | NA          |
| 1   | 3/8/21 15:53    | NA                 | NA          | NA          |
| 3   | 3/8/21 16:11    | 456.12             | 334.50      | 577.73      |
| 5   | 3/8/21 16:30    | 431.36             | 199.19      | 663.53      |
| 7   | 3/8/21 16:49    | 324.64             | 188.14      | 461.15      |
| 9   | 3/8/21 17:06    | 816.66             | 551.81      | 1081.50     |
| 11  | 3/8/21 17:24    | 670.18             | 467.17      | 873.18      |
| 13  | 3/8/21 17:41    | NA                 | NA          | NA          |
| 15  | 3/8/21 17:59    | 509.45             | 329.93      | 688.98      |
| 17  | 3/8/21 18:15    | 181.93             | 104.31      | 259.54      |
| 19  | 3/8/21 18:35    | NA                 | NA          | NA          |
| 21  | 3/8/21 18:53    | 662.77             | 235.70      | 1089.84     |
| 23  | 3/8/21 19:11    | NA                 | NA          | NA          |

**Table A.4:** Original results for all methods from the single-blind volume-controlled release experiments (32.053 , 102.301 ) and MethaneAIR Level 3 Gaussian filtered data on 30 July 2021 and 3 August 2021. The emission rates, lower bounds, and upper bounds are in kg/hr. This table was reported to the Stanford team as MethaneAIR initial emission estimates on 1 February 2022. The times are in UTC. The bounds were calculated using the method described in Section 2.3.3. The SNR is the signal-to-noise ratio. The SL is the scale length of each plume in meters

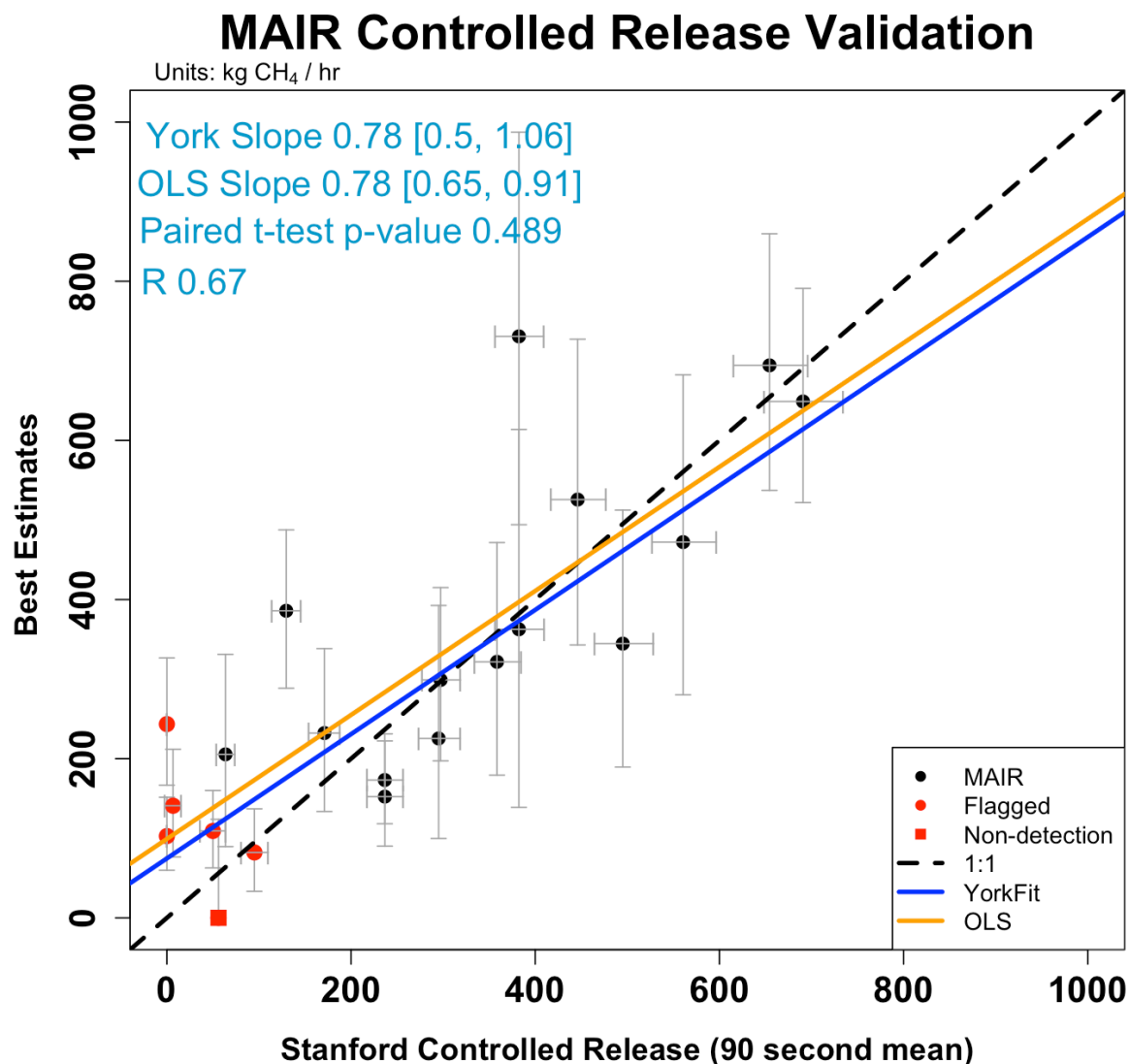
| Seg | Timestamp (UTC) | IME    | IME.l  | IME.u  | Ratio   | Ratio.l | Ratio.u | Gaussian | Gauss.l | Gauss.u | SNR  | SL     |
|-----|-----------------|--------|--------|--------|---------|---------|---------|----------|---------|---------|------|--------|
| 1   | 30/7/21 15:41   | 321.74 | 312.25 | 330.84 | 1267.08 | 1072.46 | 1419.36 | 387.04   | 318.53  | 455.55  | 2.33 | 153.92 |
| 2   | 30/7/21 15:52   | 232.19 | 222.98 | 241.58 | 765.04  | 631.16  | 885.97  | NA       | NA      | 4.44    | 2.45 | 128.82 |
| 3   | 30/7/21 16:07   | 109.22 | 102.16 | 115.46 | 206.19  | 159.37  | 250.17  | NA       | NA      | NA      | 2.19 | 79.98  |
| 4   | 30/7/21 16:22   | 102.76 | 94.56  | 110.81 | 224.24  | 189.35  | 255.61  | NA       | NA      | NA      | 2.18 | 87.16  |
| 5   | 30/7/21 16:36   | 152.31 | 137.82 | 165.12 | 362.74  | 307     | 412.7   | 253.13   | 158.86  | 347.39  | 2.26 | 106.28 |
| 6   | 30/7/21 16:51   | 291.55 | 266.56 | 313.72 | 572.28  | 448.29  | 685.25  | 159.29   | 83.16   | 235.42  | 2.5  | 175.46 |
| 7   | 30/7/21 17:06   | 82.21  | 75.89  | 87.72  | 131.26  | 109.96  | 144.92  | NA       | NA      | NA      | 2.43 | 58.3   |
| 8   | 30/7/21 17:22   | 362.67 | 339.8  | 385.66 | 694.59  | 589.45  | 812.25  | 296.98   | 260.15  | 333.82  | 2.45 | 196.69 |
| 9   | 30/7/21 18:08   | 205.48 | 196.33 | 214.49 | 515.09  | 316.43  | 651.97  | 98.32    | 14.51   | 182.13  | 2.13 | 115.74 |
| 1   | 3/8/21 15:53    | 140.85 | 133.45 | 148.46 | 215.43  | 190.36  | 240.07  | NA       | NA      | NA      | 2.25 | 85.46  |
| 3   | 3/8/21 16:11    | 344.58 | 322.5  | 362.84 | 819.55  | 664.12  | 965.76  | 282.12   | 223.41  | 340.84  | 2.53 | 185.53 |
| 5   | 3/8/21 16:30    | 472.22 | 445.41 | 497.88 | 806.07  | 690.37  | 972.87  | 258.84   | 204.09  | 313.59  | 2.49 | 237.98 |
| 7   | 3/8/21 16:49    | 299.01 | 275.76 | 323.11 | 707.65  | 589.56  | 799.42  | 128.58   | 89.51   | 167.65  | 2.62 | 185.79 |
| 9   | 3/8/21 17:06    | 694.32 | 644.67 | 736.81 | 752.41  | 616.16  | 870.63  | 784.38   | 710.68  | 858.07  | 2.29 | 363.56 |
| 11  | 3/8/21 17:24    | 648.93 | 587.62 | 711.46 | 1130.2  | 784.58  | 1471.68 | 1056.21  | 981.26  | 1131.17 | 2.75 | 203.28 |
| 13  | 3/8/21 17:41    | 243.47 | 229.22 | 256.64 | 932.64  | 759.34  | 1083.86 | 313.8    | 252.26  | 375.35  | 2.02 | 128.49 |
| 15  | 3/8/21 17:59    | 730.75 | 701.77 | 761.88 | 1189.06 | 1078.02 | 1297.29 | 192.44   | 90.55   | 294.34  | 2.52 | 280.44 |
| 17  | 3/8/21 18:15    | 385.89 | 375.8  | 395.69 | 818.72  | 687.49  | 918.82  | NA       | NA      | 3.18    | 2.48 | 199.81 |
| 19  | 3/8/21 18:35    | NA     | NA     | NA     | NA      | NA      | NA      | NA       | NA      | NA      | NA   | NA     |
| 21  | 3/8/21 18:53    | 525.67 | 490.81 | 558.78 | 1242.4  | 1020.6  | 1441.07 | 328.44   | 207.52  | 449.37  | 2.83 | 171.8  |
| 23  | 3/8/21 19:11    | 173.21 | 167.15 | 178.92 | 210     | 178.99  | 234.64  | 505.73   | 447.7   | 563.76  | 2.39 | 53.87  |

**Table A.5:** Single-blind volume-controlled release data reported by the Stanford team on 1 February 2022. The cr\_kgh\_CH4\_mean90, cr\_kgh\_CH4\_lower90, and cr\_kgh\_CH4\_upper90 represent the 90-second window rolling average, lower, and upper flow rates in kg/hr. The wind speeds are in m/s. The timestamps are in UTC.

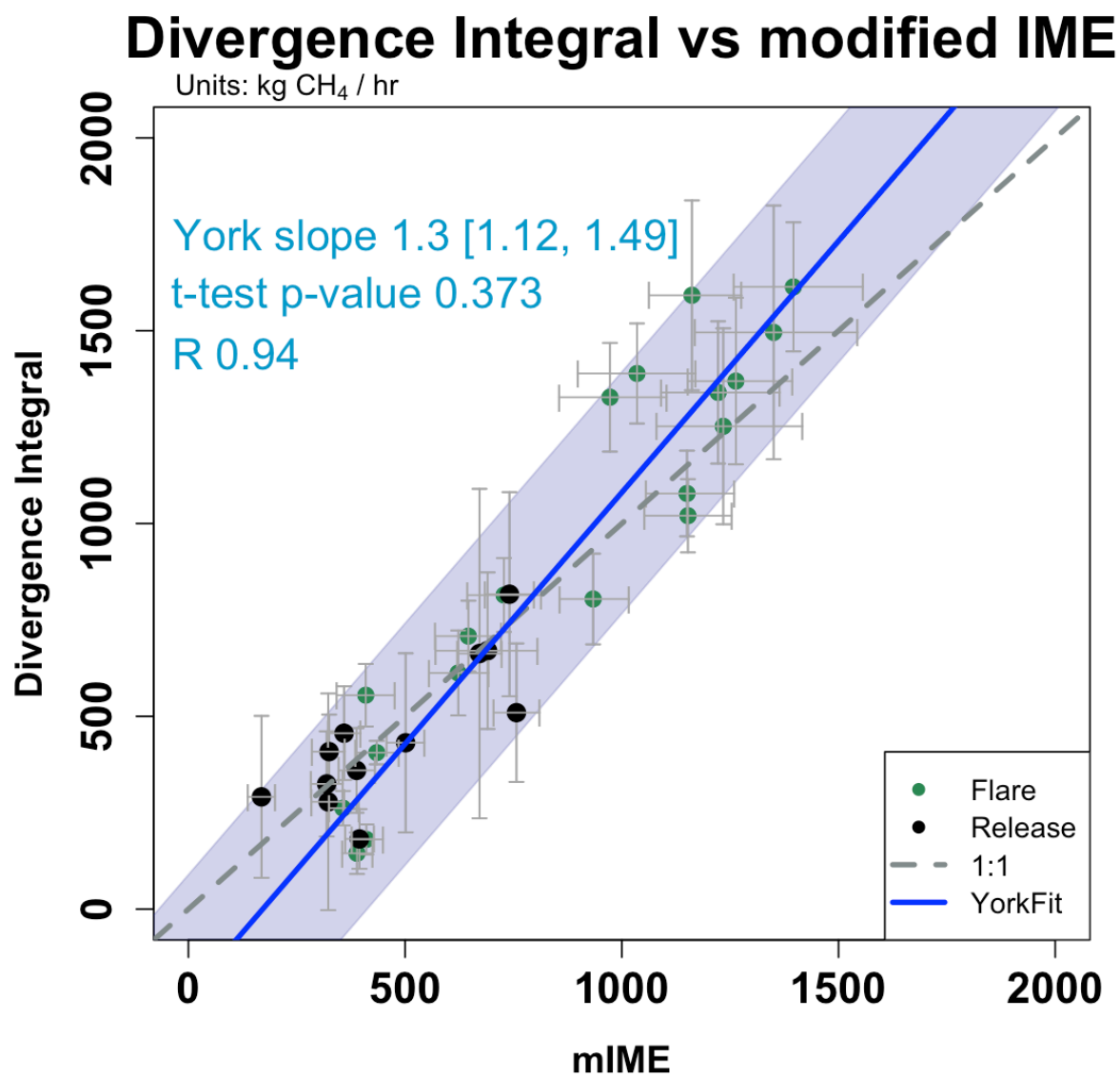
| Seg | Timestamp<br>(UTC) | cr_kgh_CH4<br>mean90 | cr_kgh_CH4<br>lower90 | cr_kgh_CH4<br>upper90 | Winds |
|-----|--------------------|----------------------|-----------------------|-----------------------|-------|
| 1   | 30/7/21 15:41      | 358.48               | 334.29                | 384.75                | 2.8   |
| 2   | 30/7/21 15:52      | 171.08               | 154.32                | 188.05                | 2.71  |
| 3   | 30/7/21 16:07      | 49.98                | 36.06                 | 63.71                 | 2.44  |
| 4   | 30/7/21 16:22      | 0.00                 | 0.00                  | 0.00                  | 2.29  |
| 5   | 30/7/21 16:36      | 236.74               | 217.42                | 256.81                | 2.36  |
| 6   | 30/7/21 16:51      | 295.08               | 273.58                | 318.53                | 2.59  |
| 7   | 30/7/21 17:06      | 95.05                | 80.53                 | 109.95                | 2.68  |
| 8   | 30/7/21 17:22      | 382.25               | 356.59                | 409.77                | 2.54  |
| 9   | 30/7/21 18:08      | 63.65                | 53.99                 | 73.67                 | 3.29  |
| 1   | 3/8/21 15:52       | 6.72                 | -2.07                 | 15.42                 | 2.32  |
| 3   | 3/8/21 16:11       | 495.05               | 464.61                | 528.52                | 2.28  |
| 5   | 3/8/21 16:30       | 560.57               | 527.17                | 596.61                | 2.27  |
| 7   | 3/8/21 16:48       | 297.28               | 277.02                | 318.78                | 2.03  |
| 9   | 3/8/21 17:06       | 654.38               | 615.38                | 695.86                | 1.92  |
| 11  | 3/8/21 17:24       | 690.80               | 648.48                | 734.36                | 2.07  |
| 13  | 3/8/21 17:41       | 0.00                 | 0.00                  | 0.00                  | 2     |
| 15  | 3/8/21 17:59       | 382.28               | 356.57                | 409.38                | 1.64  |
| 17  | 3/8/21 18:15       | 129.63               | 113.98                | 145.39                | 1.87  |
| 19  | 3/8/21 18:35       | 56.05                | 41.93                 | 70.22                 | 2.48  |
| 21  | 3/8/21 18:53       | 446.05               | 417.23                | 476.86                | 2.42  |
| 23  | 3/8/21 19:11       | 236.63               | 217.37                | 256.66                | 2.21  |

**Table A.6:** Single-blind volume-controlled release data reported by the Stanford team on 22 March 2023. The release\_rate\_kgh is the metered rate reported by the Stanford team. The lower\_95CI and upper\_95CI are the lower and upper bound of the 95th percentile confidence interval of the metered rates. The MAIR is our blinded best estimate of emissions reported to the Stanford team. The MAIR\_lower and MAIR\_upper are the lower and upper bound of the 95th percentile confidence interval of our estimates. The timestamps are in UTC.

| Seg | Timestamp<br>(UTC) | release_rate<br>(kgh) | lower_95CI | upper_95CI | MAIR | MAIR<br>lower | MAIR<br>upper |
|-----|--------------------|-----------------------|------------|------------|------|---------------|---------------|
| 1   | 25/10/22 17:15     | 205.23                | 203.92     | 206.55     | 177  | 121           | 232           |
| 2   | 25/10/22 17:32     | 97.04                 | 96.42      | 97.66      | 61   | 18            | 105           |
| 3   | 25/10/22 17:50     | 987.13                | 936.13     | 1038.13    | 855  | 578           | 1133          |
| 4   | 25/10/22 18:09     | 982.50                | 928.47     | 1036.52    | 908  | 662           | 1155          |
| 5   | 25/10/22 18:27     | 0.00                  | 0.00       | 0.00       | 0    | -100          | 100           |
| 6   | 25/10/22 18:47     | 468.33                | 463.82     | 472.85     | 669  | 409           | 928           |
| 7   | 25/10/22 19:07     | 24.42                 | 24.31      | 24.53      | 0    | -100          | 100           |
| 8   | 25/10/22 19:28     | 634.57                | 625.46     | 643.68     | 544  | 374           | 714           |
| 9   | 25/10/22 20:10     | 142.14                | 141.52     | 142.75     | 303  | 183           | 423           |
| 10  | 25/10/22 20:29     | 74.65                 | 74.32      | 74.99      | 89   | -11           | 189           |
| 11  | 25/10/22 20:46     | 63.03                 | 62.78      | 63.29      | 140  | 89            | 191           |
| 1   | 29/10/22 16:25     | 0.00                  | 0.00       | 0.00       | 0    | -100          | 100           |
| 2   | 29/10/22 16:50     | 340.16                | 336.53     | 343.79     | 196  | 97            | 294           |
| 3   | 29/10/22 17:17     | 671.74                | 660.33     | 683.16     | 787  | 290           | 1283          |
| 4   | 29/10/22 17:42     | 405.59                | 400.31     | 410.87     | 644  | 271           | 1017          |
| 5   | 29/10/22 18:07     | 33.61                 | 33.27      | 33.94      | 94   | -6            | 194           |
| 6   | 29/10/22 18:31     | 337.56                | 333.89     | 341.23     | 388  | 296           | 480           |
| 7   | 29/10/22 18:54     | 818.30                | 793.86     | 842.75     | 958  | 549           | 1368          |
| 8   | 29/10/22 19:16     | 1060.77               | 1002.64    | 1118.91    | 1199 | 886           | 1512          |
| 9   | 29/10/22 19:38     | 1289.35               | 1221.55    | 1357.15    | 1489 | 1004          | 1974          |
| 10  | 29/10/22 19:59     | 585.32                | 575.53     | 595.11     | 579  | 397           | 761           |
| 11  | 29/10/22 20:21     | 199.26                | 165.06     | 233.47     | 252  | 147           | 357           |
| 12  | 29/10/22 20:42     | 493.52                | 487.02     | 500.02     | 584  | 328           | 839           |
| 13  | 29/10/22 21:02     | 168.86                | 167.19     | 170.53     | 112  | -21           | 244           |



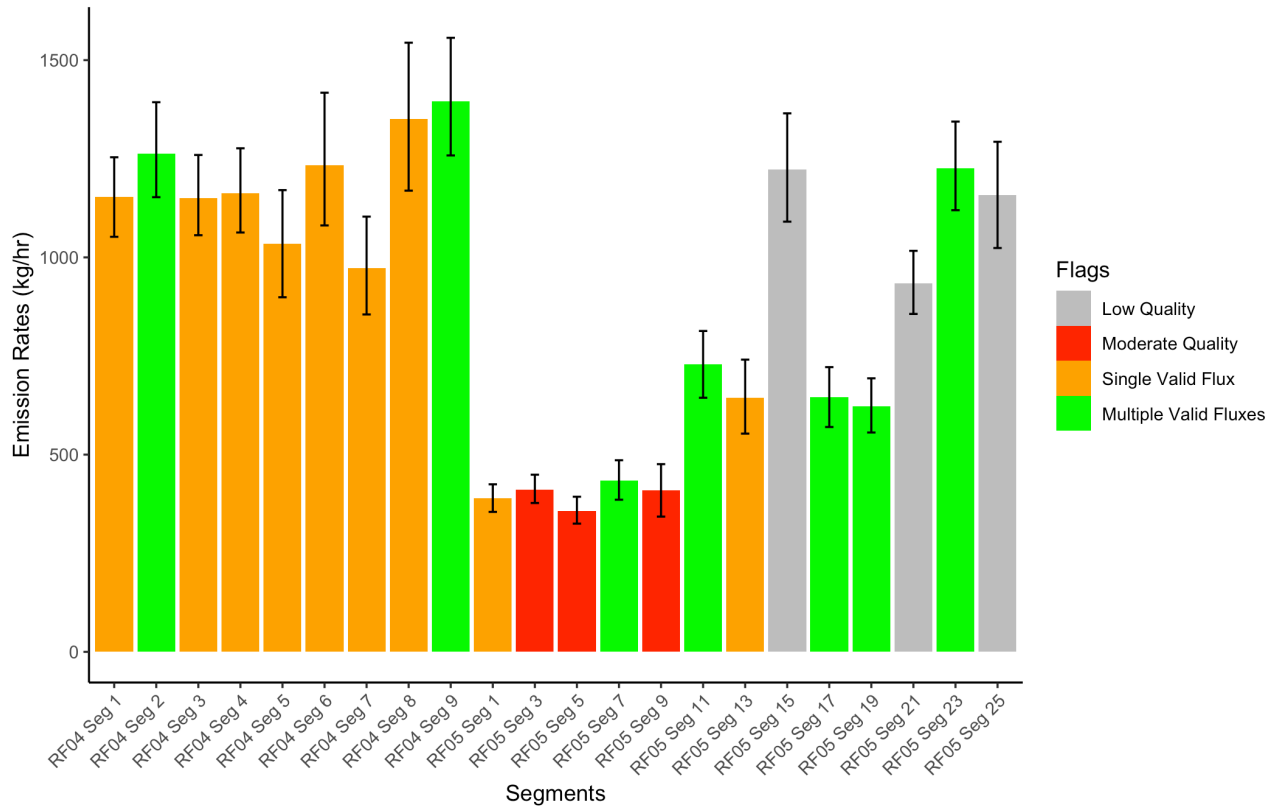
**Figure A.8:** An analogous plot of Figure 3. The results presented in this figure are based on the “best estimates” reported on 1 February 2022. The black circles represent the post-unblinding comparison between the reported and estimated emissions. The red square represents the original comparison, which was later identified as no detection and removed. The red circles represent the flagged data points determined by the decision tree (Section A.5.4). The blue solid line is the post-unblinding York fit. The orange is the Ordinary Least Squares (OLS) fit. The comparisons shown by the York and OLS fits are notional and do not actually pertain to the information exchanged in the unblinding. The errors in the MethaneAIR data had not yet been assessed in detail and, in fact, were overestimated. The MethaneAIR team did not know the metered rates and the associated uncertainties were not documented.



**Figure A.9:** An analogous plot of Figure 3. The results presented in this figure are based on the post-unblinding estimates using the DI results. The black circles represent the post-unblinding comparison between the reported and estimated emissions. The blue solid line is the post-unblinding York fit.

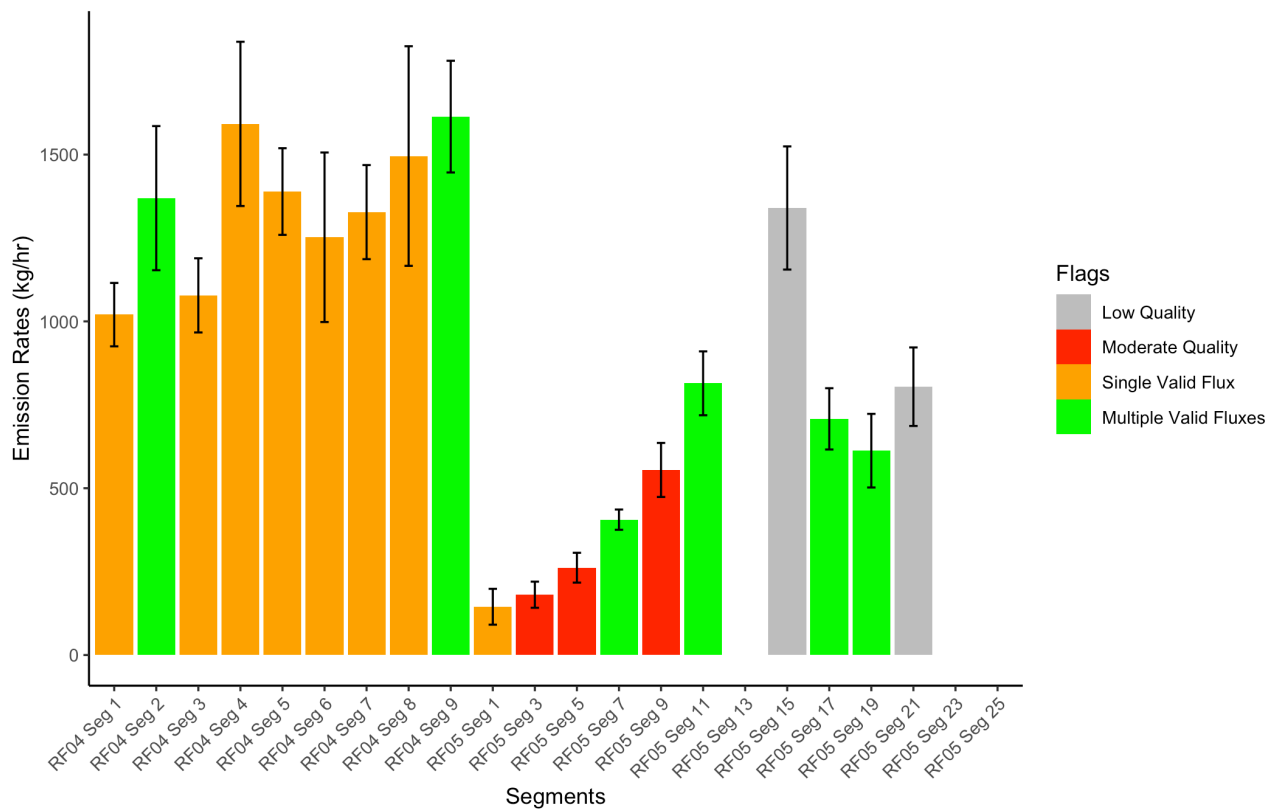
#### A.4.4 UNLIT FLARE

As shown in Figure A.11, the flare emission rates range from 500 kg/hr to 2000 kg/hr.



**Figure A.10:** Methane emission estimates from an unlit flare during RF04 and RF05 using the mME method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16.





**Figure A.11:** Methane emission estimates from an unlit flare during RF04 and RF05 using the DI method. The endpoints are tagged and color-coded according to the decision tree in Figure A.16.

#### A.4.5 MVD & PIPELINE

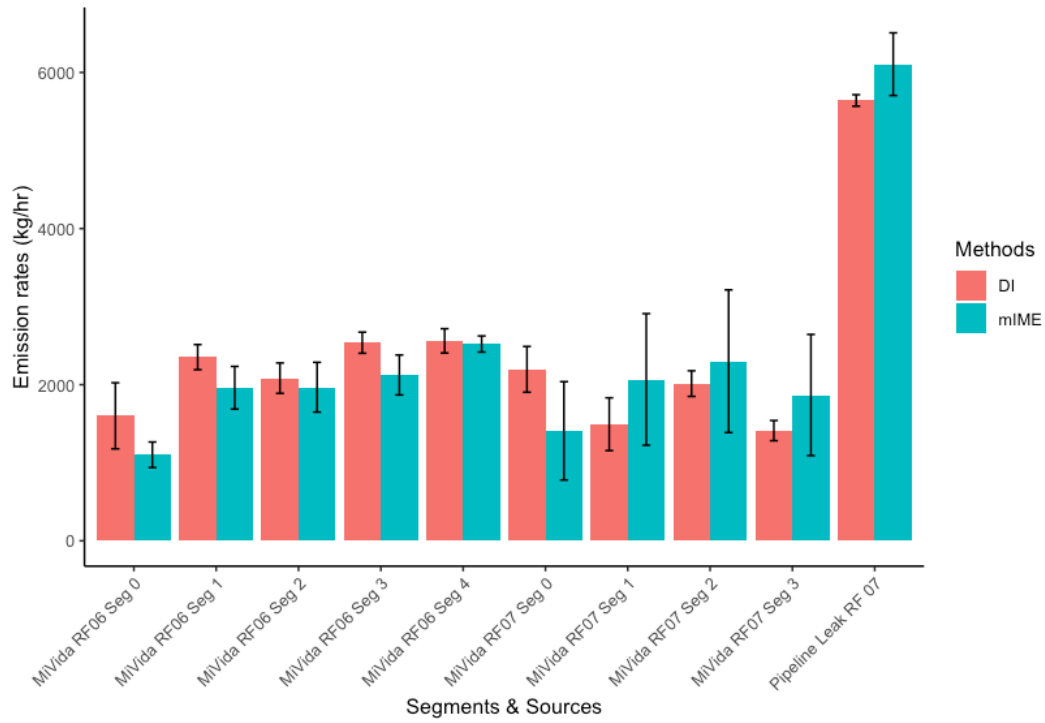
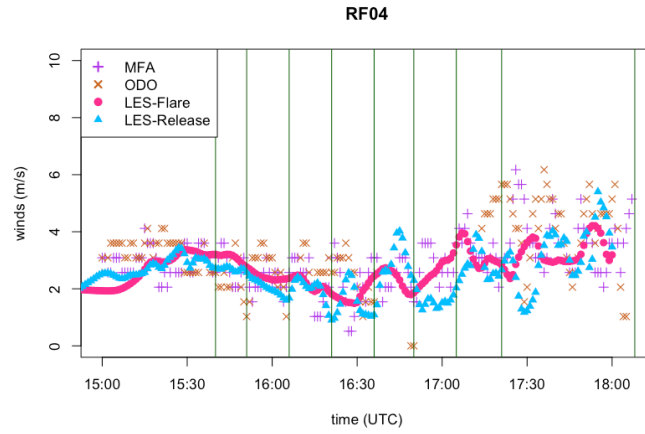


Figure A.12: Barplot of the miscellaneous sources quantified in the Delaware Basin.

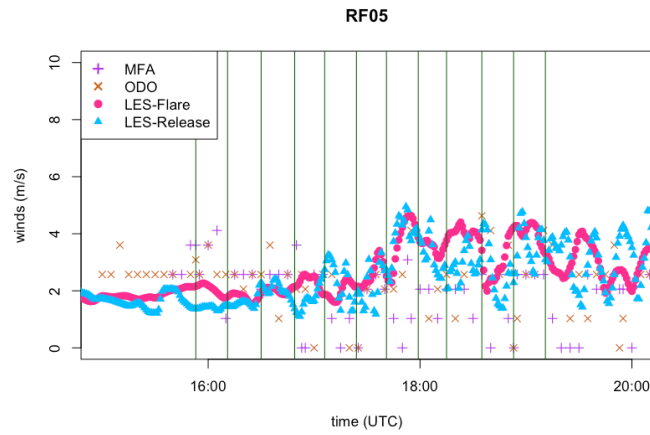


## A.5 ANALYSIS METHODS

### A.5.1 WIND ANALYSIS



(a) RF04 30 July 2021

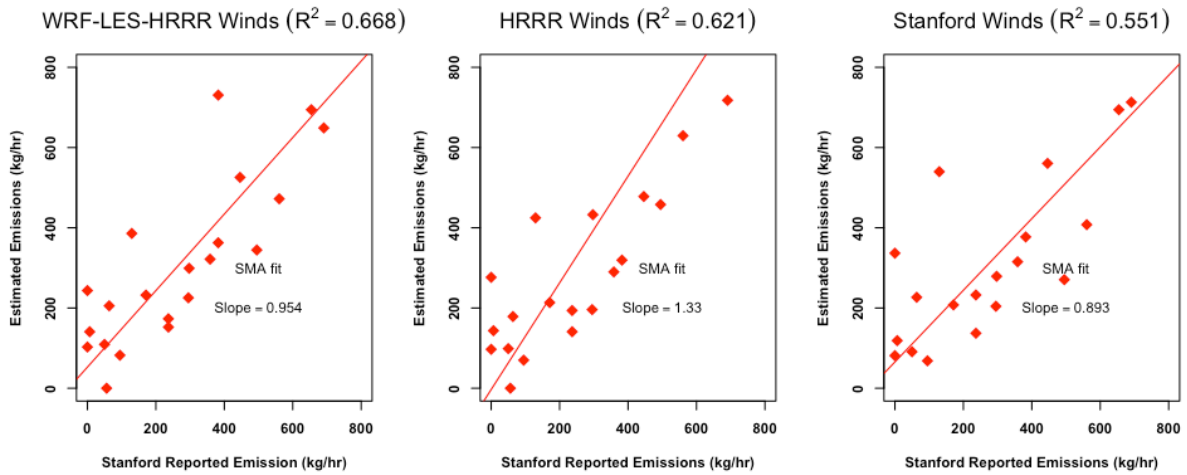


(b) RF05 3 August 2021

**Figure A.13:** ASOS surface wind comparison from the single-blind volume-controlled release experiments during (a) RF04 on 30 July 2021 and (b) RF05 on 3 August 2021. The purple crosses represent ASOS winds from the Midland International Air & Space Port (MAF), which is 15 km from the single-blind volume-controlled release site. The orange x's represent ASOS winds from the Odessa-schlemeyer Field Airport (ODO), which is 17 km from the single-blind volume-controlled release site. The magenta circles represent surface winds from the LES at the flare. The blue triangles represent surface winds from the LES at the single-blind volume-controlled release site. The dark green vertical lines represent times when MAIR flew over the targets. All LES winds during the controlled release experiments and unlit flare observation agree with the ASOS winds (two-sided t-test p-values are greater than 0.05).

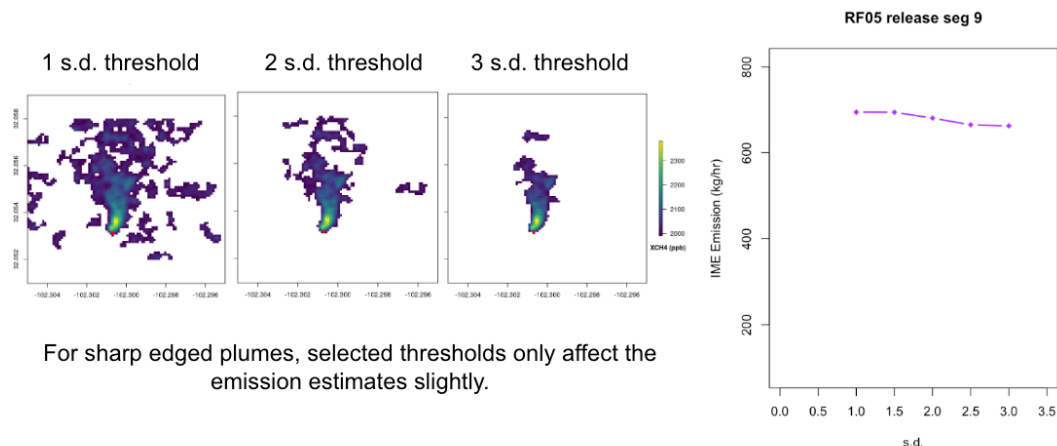
### A.5.2 WIND PRODUCT ANALYSIS

To assess our LES winds compared to mesoscale HRRR winds and winds observed at the release site, we plot the estimated emission rates against the known emission rates from the Stanford team using different wind products. As shown in Figure A.14, effective wind from the LES provides the highest  $R^2$  value and slope closest to 1. Note that the choice of driven wind product is not limited to HRRR data; any wind products that can drive WRF can run WRF-LES. Although HRRR is the highest temporal and spatial resolution wind product for the Contiguous United States (CONUS), further study of other wind products outside CONUS is needed to determine the most appropriate wind products.



**Figure A.14:** Estimated emission rates with the mIME using different wind products vs. the known emission rates from the Stanford team. From left to right: estimated emission rates based on effective winds from WRF-LES-HRRR, HRRR, and measured winds. The paired t-test between the estimated emissions and Stanford reported emissions suggests that we cannot reject the null hypothesis that a true difference in means is equal to 0 within 95% confidence interval for the estimated emissions based on WRF-LES-HRRR and Stanford observed winds with p-values of 0.166 and 0.309 respectively. However, the test suggests the alternative hypothesis of a true difference in means between the emissions derived from HRRR winds and the reported emissions with a p-value of 0.0408.

### A.5.3 THRESHOLD ANALYSIS



**Figure A.15:** The thresholds do not affect the IME values of the plume of interest.

### A.5.4 DECISION TREE

To understand the relationship between the actual emission rates and the emission estimates, we attempted to use several regression models for our data. York regression was the most appropriate one because York includes correlated errors from both axes. OLS was included in this study since OLS is often used in this type of experiment. See Sherwin et al. <sup>43</sup>, SI Section S4, for further discussion of tradeoffs associated with York and OLS fits in single-blind tests of methane remote sensing.

Before the actual single-blind volume-controlled release values were revealed, we developed a decision tree workflow that assigns a confidence flag to each case (Section A.5.4). The original version of our decision tree was developed purely based on our analysis of our measurement system and algorithms. It was then adjusted slightly after unblinding the controlled release metered emissions. When the signal-to-noise ratio (SNR) for the plume, relative to the background, is below 1 or significant interference is observed, we classify the case as either no detection (Endpoint 0) or poor quality (Endpoint 1). When the SNR is between 1 and 2.5, the estimates are near the detection limit, pro-

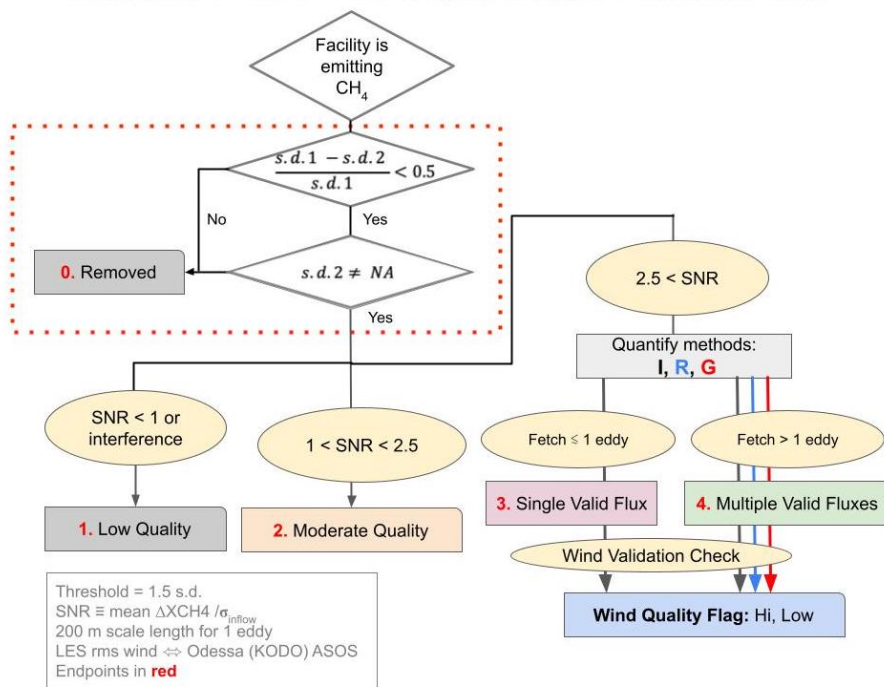
viding a moderate-quality estimate (Endpoint 2). Only the mIME method can be used, and the confidence will likely be low. When the SNR is above 2.5, we checked if the plume size is greater than one eddy. If the plumes are smaller than one eddy, only the mIME method can be used (Endpoint 3). If the plumes are larger than one eddy, all three methods developed were applied (Endpoint 4). For the last step, the WRF-HRRR-LES were compared to the nearby ASOS winds and later assigned quality flags based on agreement to the observed winds. If the winds disagree (two-sided t-test p-value is less than 0.05), the wind quality is low. When winds agree (two-sided t-test p-value is greater than 0.05), the wind quality is high. All LES winds during the controlled release experiments and unlit flare observation agree within tolerance with the ASOS winds.

As expected, the DI and the Ratio approaches work well with larger sources such as the unlit flare, MiVida Gas Processing Plant, or the pipeline leak plume. However, the ratio approach is limited by the resolution mismatch with the sensor and the LES domain size, making this approach less favorable. So, we decided to proceed with the mIME and the DI methods.

After the metered single-blind volume-controlled release values were revealed, the decision tree was revised to include additional steps that help remove false positive cases and help us determine the detection limits of MethaneAIR.

By filtering out cases that (i) have the absolute differences between mIME emission estimates using a threshold of 1 and 2 standard deviations divided by emission estimate using a threshold of 1 standard deviation greater than 0.5 (i.e., 2 s.d. case has a much different plume shape) or (ii) no emission estimates at the threshold of 2 standard deviations, we designate as “below detection limit” 6 cases from the single-blind volume-controlled release experiment. One was a false negative case, where methane enhancements were not detected. The other four were emission estimates around the detection limit of 200 kg/hr. The part of the decision tree added after unblinding, intended to reduce false positives in the contaminated environment of this test, is boxed in red in Figure A.16.

## MethaneAIR/SAT Plume Quantification Decision Tree



**Figure A.16:** Updated decision tree classifying quality levels of emission estimates. Most of the steps were developed purely based on our analysis of the data and signal processing. The steps in the red dashed box were added after the unblinding process. First, we filter out all the estimates that (i) have the absolute differences between mIME emission estimates using thresholds of 1 and 2 standard deviations divided by emission estimate using a threshold of 1 standard deviation to be greater than 0.5 or (ii) no emission estimates at the threshold of 2 standard deviations. After that, for each estimate with SNR below 1 or significant interference was observed, we classify the case as either no detection (Endpoint 0) or poor quality (Endpoint 1). The estimates are moderate quality when the SNR is between 1 and 2.5 (Endpoint 2). Only the mIME method can be used. When the SNR is above 2.5, we checked if the plume size is greater than one eddy. If the plumes are smaller than one eddy, only the mIME method can be used (Endpoint 3). All three different methods developed were applied if the plumes are larger than one eddy (Endpoint 4). For the last step, the WRF-HRRR-LES were compared to the nearby ASOS winds and later assigned quality flags based on agreement to the observed winds. If the winds disagree, the wind quality is low. When winds agree, the wind quality is high.



#### A.5.5 ADDITIONAL ATTEMPTS TO DETERMINE CONTAMINATION NEAR THE METHANE PLUMES

Although our decision tree (Section A.5.4) removed six plumes (below detection) from the total 21 plumes based on the signal-to-noise ratio and the size of the plumes, we wanted to investigate the limitations of our sensors and methods further. We decided to perform two more analyses with the hypothesis that these two will help us better understand the limitations.

The first analysis was to perform a k-s test between  $\text{XCH}_4$  in the inflow region of each scene and a clear scene defined as latitude 32.10 - 32.11 and longitude -102.32 to -102.31. When the p-values of the k-s test are less than 0.05, we can reject the null hypothesis that the  $\text{XCH}_4$  observed in the inflow and clear scene came from the same distribution. A positive outcome from the K-S test suggests that there may be extraneous methane contamination in the inflow.

The second analysis was to filter the clumps with 60 pixels and 100 pixels. The new choices of thresholds are arbitrarily rounded to the multiples of the original 20-pixel threshold and fractions of a large eddy (approximately 100 m). The original 20-pixel threshold or  $10 \text{ m} \times 10 \text{ m} \times 20 \text{ pixels}$  is equivalent to a  $45 \text{ m} \times 45 \text{ m}$  plume, which is approximately half of the size of an eddy. The 60-pixel threshold is equivalent to a  $77 \text{ m} \times 77 \text{ m}$  plume, which is approximately two-thirds of the size of an eddy. The 100-pixel threshold is equivalent to a  $100 \text{ m} \times 100 \text{ m}$  plume, which is approximately the size of an eddy.

**Table A.7:** Summary table of removed (not detected) RF04 plumes based on three methods: decision tree (Section A.5.4), 60 pixel threshold, and K-S test.

| Segment | Decision Tree | 60 pixels | K-S test |
|---------|---------------|-----------|----------|
| 1       |               |           | x        |
| 2       |               |           | x        |
| 3       | x             | x         |          |
| 4       | x             | x         |          |
| 5       |               | x         | x        |
| 6       |               |           |          |
| 7       | x             | x         |          |
| 8       |               |           |          |
| 9       | x             | x         |          |

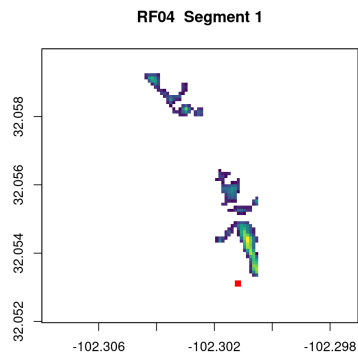
**Table A.8:** Summary table of removed RF05 plumes based on three methods: decision tree (Section A.5.4), 60 pixel threshold, and K-S test.

| Segment | Decision Tree | 60 pixels | K-S test |
|---------|---------------|-----------|----------|
| 1       | x             | x         | x        |
| 3       |               |           | x        |
| 5       | x             | x         | x        |
| 7       | x             | x         |          |
| 9       |               | x         | x        |
| 11      |               |           |          |
| 13      | x             | x         | x        |
| 15      |               |           |          |
| 17      | x             | x         |          |
| 21      |               |           |          |
| 23      |               |           |          |

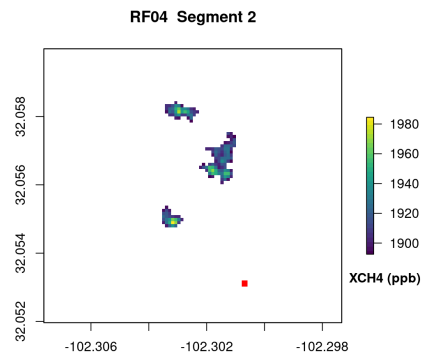
## A.6 CROPPED BLINDED-VOLUME CONTROLLED RELEASE PLUMES

The following figures are cropped single-blind volume-controlled release plumes from RF04 and RF05. The plumes were identified based on the known location of the single-blind volume-controlled release site (grey triangles). To reduce the noise of the raw  $1 \times 1$  Level 3 data, the data were

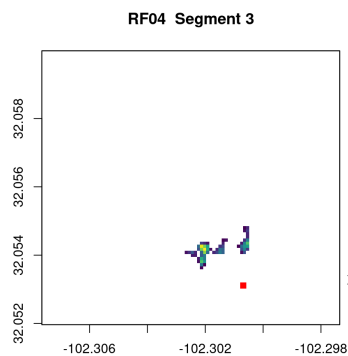
later filtered by a Gaussian filter (7-pixel x 7-pixel kernel with the full width at a maximum of 2 pixels). We then identified the inflow region and removed the pixels below the 1.5 standard deviation of the inflow threshold. After that, we removed the small clumps with fewer than 20 pixels from the scene of interest.



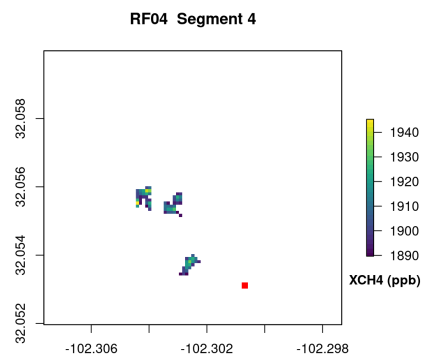
**Figure A.17:** Filtered single-blind volume-controlled release RF04 Segment 1.



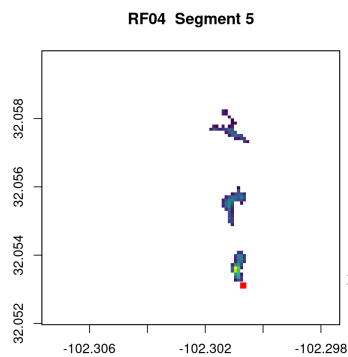
**Figure A.18:** Filtered single-blind volume-controlled release RF04 Segment 2.



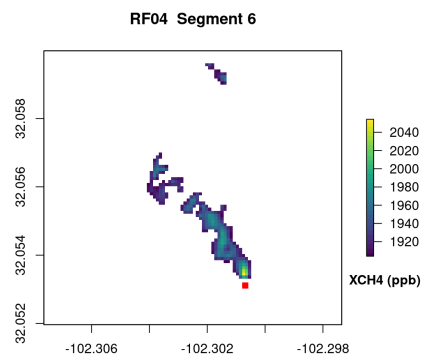
**Figure A.19:** Filtered single-blind volume-controlled release RF04 Segment 3.



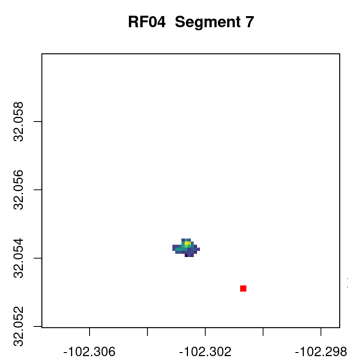
**Figure A.20:** Filtered single-blind volume-controlled release RF04 Segment 4.



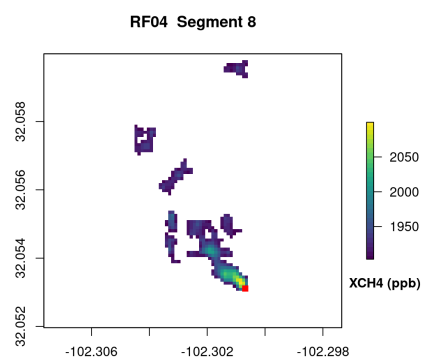
**Figure A.21:** Filtered single-blind volume-controlled release RF04 Segment 5.



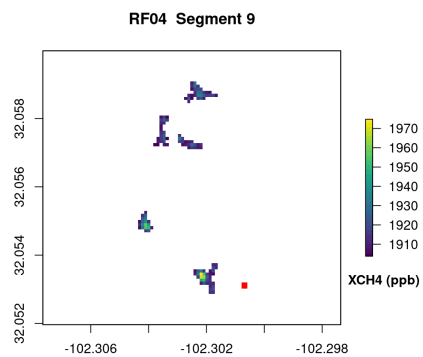
**Figure A.22:** Filtered single-blind volume-controlled release RF04 Segment 6.



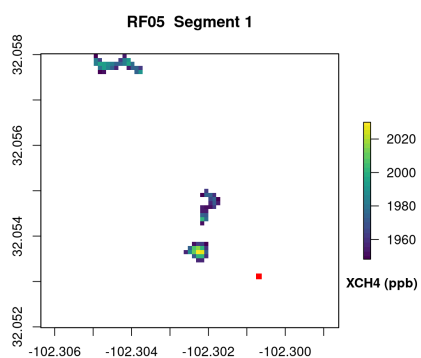
**Figure A.23:** Filtered single-blind volume-controlled release RF04 Segment 7.



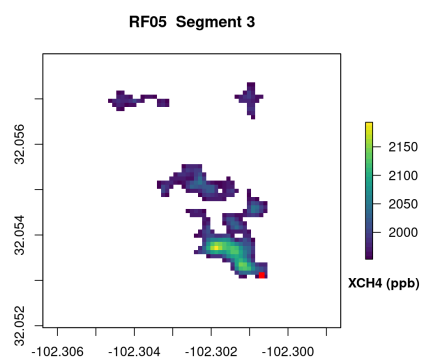
**Figure A.24:** Filtered single-blind volume-controlled release RF04 Segment 8.



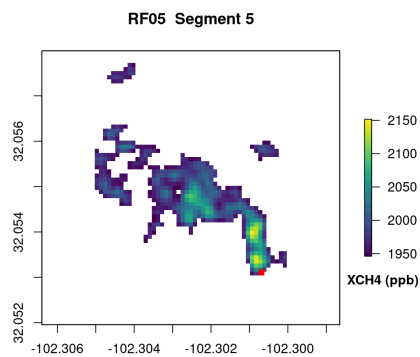
**Figure A.25:** Filtered single-blind volume-controlled release  
RF04 Segment 9.



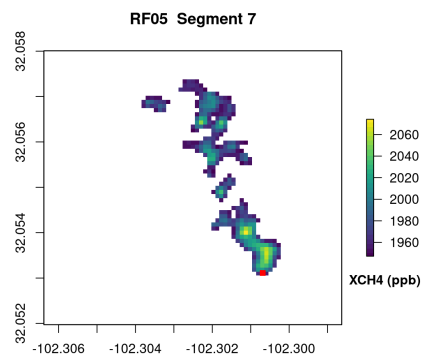
**Figure A.26:** Filtered single-blind volume-controlled release  
RF05 Segment 1.



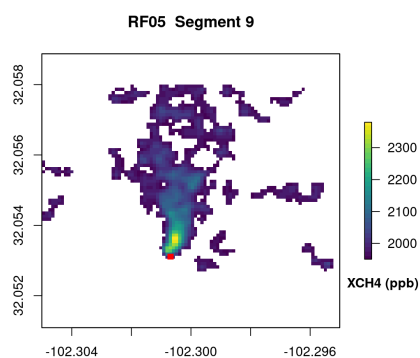
**Figure A.27:** Filtered single-blind volume-controlled release  
RF05 Segment 3.



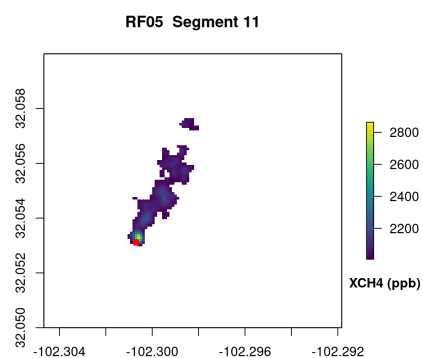
**Figure A.28:** Filtered single-blind volume-controlled release RF05 Segment 5.



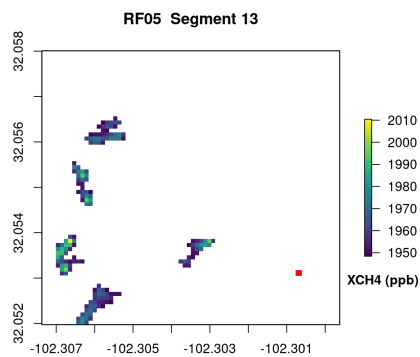
**Figure A.29:** Filtered single-blind volume-controlled release RF05 Segment 7.



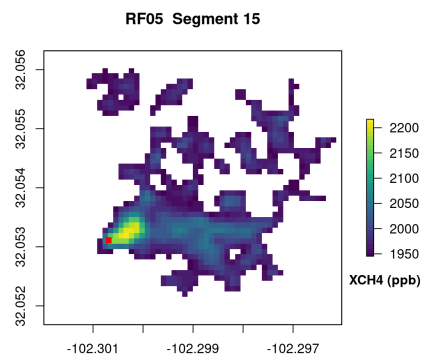
**Figure A.30:** Filtered single-blind volume-controlled release RF05 Segment 9.



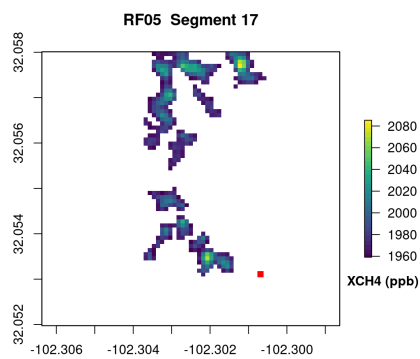
**Figure A.31:** Filtered single-blind volume-controlled release RF05 Segment 11.



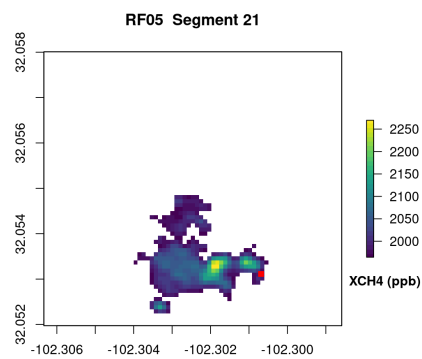
**Figure A.32:** Filtered single-blind volume-controlled release  
RF05 Segment 13.



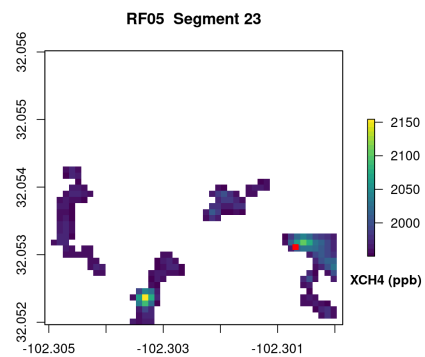
**Figure A.33:** Filtered single-blind volume-controlled release  
RF05 Segment 15.



**Figure A.34:** Filtered single-blind volume-controlled release  
RF05 Segment 17.

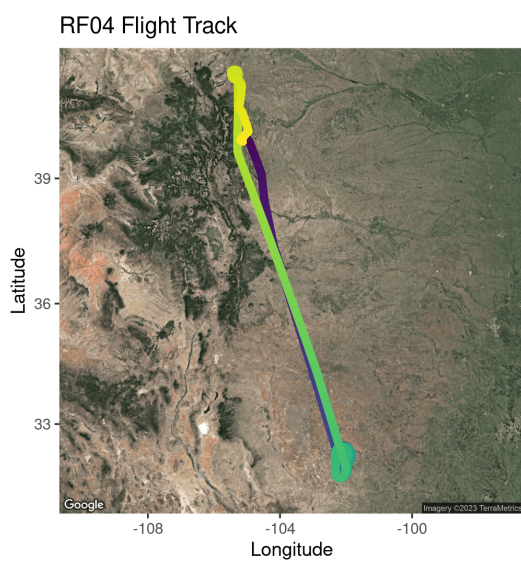


**Figure A.35:** Filtered single-blind volume-controlled release  
RF05 Segment 21.

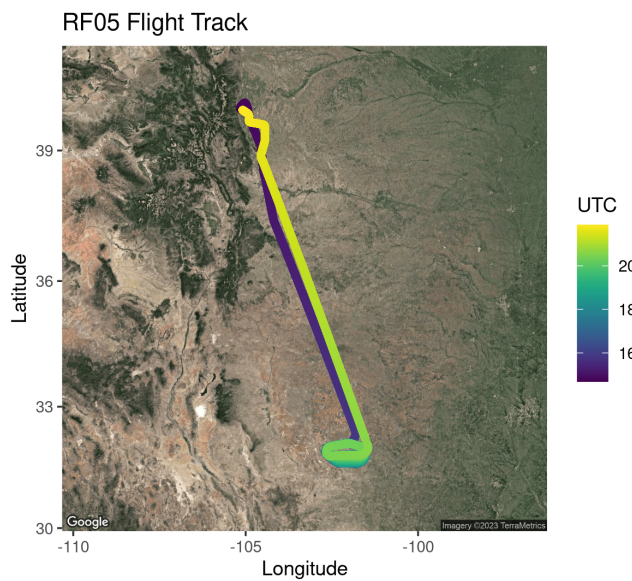


**Figure A.36:** Filtered single-blind volume-controlled release  
RF05 Segment 23.

## A.7 METHANEAIR FLIGHT TRACKS



**Figure A.37:** MethaneAIR RF04 flight track



**Figure A.38:** MethaneAIR RF05 flight track



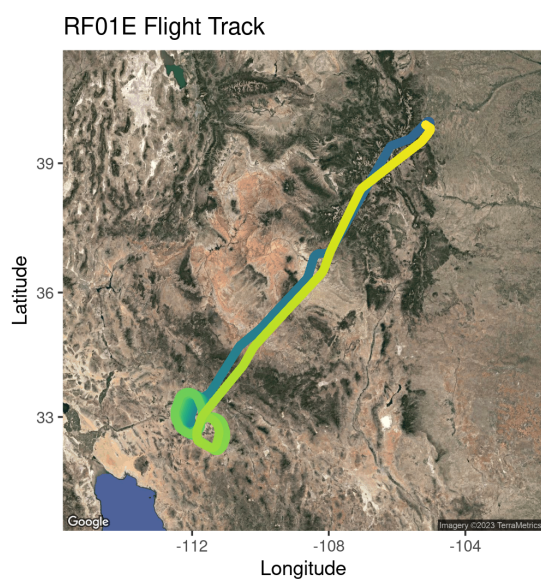


Figure A.39: MethaneAIR RF01E flight tracks

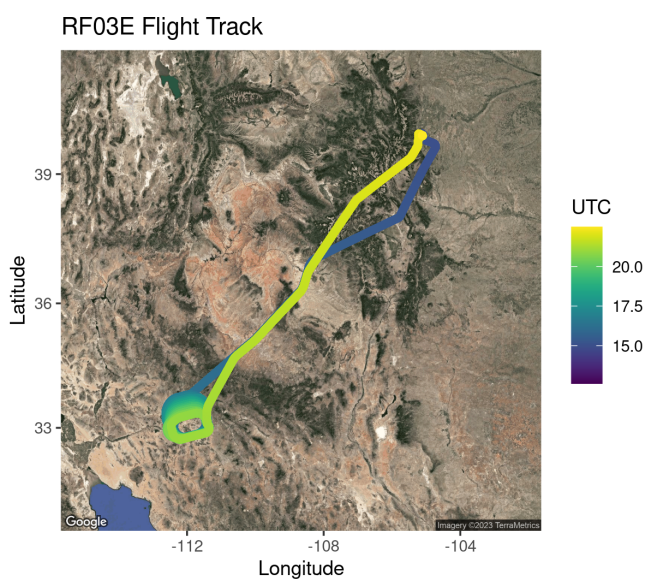


Figure A.40: MethaneAIR RF03E flight tracks

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