Scope of Work For

Project 14-002
Analysis of Airborne Formaldehyde Data Over Houston Texas Acquired During the 2013 DISCOVER-AQ and SEAC4RS Campaigns

Prepared for

Air Quality Research Program (AQRP)
The University of Texas at Austin

by

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1. Background

Houston, Texas is one of the largest cities in the United States and certainly the largest city in Texas. The Houston-Galveston-Brazoria Metropolitan Area (HGBMA), an area with over five million inhabitants, often exhibits high ozone concentrations during stagnant wind conditions. The 2000 Texas Air Quality Study (TexAQS I), which investigated such events, revealed that highly reactive volatile organic compounds (HRVOC's, ethene, propene, 1,3-butadiene, and butenes) from very large petrochemical industrial facilities, particularly in and around the Houston Ship Channel, react with elevated NO_x ($NO + NO_2$) co-emitted from these facilities resulting in ozone (O_3) levels exceeding federal standards. A major finding of this study was that industrial emissions of these HRVOC's were underreported by these facilities by over an order of magnitude [Daum et al., 2003; Ryerson et al., 2003; and Wert et al., 2003a]. This important finding led to numerous follow-up studies and analyses for the HGBMA, including: the 2nd Texas Air Quality Study (TexAQS II) in 2005 and 2006 [Parrish et al., 2009]; the TexAQS II Radical and Aerosol Measurement Project (TRAMP) [Lefer et al., 2010]; the 2009 Study of Houston Atmospheric Radical Precursors (SHARP) [Olaguer et al., 2009]; and measurements and modeling analysis by Rappenglück et al. [2010], Johansson et al. [2013], and Parrish et al. [2012], to name a few of the many studies.

In all cases, significantly elevated levels (>30 parts-per-billion, ppbv) of formaldehyde (CH_2O) have been measured downwind of these petrochemical facilities arising from very rapid oxidation of the two most abundant HRVOC's in the HGBMA, ethene and propene, with OH. Formaldehyde analysis of Parrish et al. [2012], which is based upon CH_2O measurements by the PI's (Fried) group acquired during the 2000 TexAQS I study [Wert et al., 2003a], provides very convincing evidence of this enhanced CH_2O photochemical production downwind of petrochemical facilities located along the Houston Ship Channel. Parrish et al. [2012] also show the enhanced production of O_3 that subsequently results when CH_2O rapidly decomposes in the atmosphere by photolysis and reactions with OH.

In addition to CH_2O production from HRVOC's, this gas is also produced from incomplete fossil fuel combustion (automotive sources and industrial flares, as examples), biomass burning, and oxidation of isoprene and various terpenes as well as slower oxidation of alkanes and aromatic compounds (Fried et al., 1997 and 2003a,b and references therein). In the remote atmosphere, methane (CH_4) oxidation becomes the dominant source of this gas. Nighttime oxidation of emitted VOC's by O_3 and/or the nitrate radical (NO_3) may also be important sources of this gas that could contribute to early morning radical formation. Olauger et al. [2009] also postulate the presence of undercounted primary emission sources.

A comprehensive understanding of O_3 in the HGBMA therefore requires a comprehensive understanding of CH_2O emissions, photochemical production rates, and transport processes. Despite the extensive efforts and advances from past studies, there are still major gaps in our understanding of CH_2O in the HGBMA. As discussed by Parrish et al. [2012], quantifying the relative contributions of primary versus secondary sources of CH_2O is crucial for developing effective ozone control strategies. Unfortunately, the various studies above arrive at different conclusions regarding the relative contributions of primary (P) versus secondary (S) sources of CH_2O over the entire Houston-Galveston-Brazoria Metropolitan Area. For example, the analysis of Parrish et al. [2012] report that 92 ± 4% of the total CH_2O source over the HGBMA is from secondary production formed by the oxidation of the HRVOC's emitted by the petrochemical facilities and that the primary emissions from these facilities and motor vehicles only accounts for

~ 5% of the total. By contrast, Johansson et al. [2013] employing optical remote sensing measurements close to petrochemical refineries in the Houston Ship Channel, Texas City, and Mont Belvieu areas coupled with a Lagrangian plume model determined a primary contribution of 90% in these cases. These are only two of the many apparent contradictory studies. Despite such divergent conclusions, both sources of CH_2O may actually be important in different regimes. Close to large petrochemical complexes direct emissions of CH_2O from flaring and other operations can indeed dominate, while further downwind secondary sources can become more important, particularly during summer months where photochemical activity is high. Because of the importance of this issue, it is highly desirable to revisit the issue of CH_2O source apportionment employing new data acquired in 2013, the most up-to-date emission inventories, as well as new analysis approaches.

Updating the emission inventories and temporal trends for CH_2O and its HRVOC precursors is another area requiring attention. The study by Washenfelder et al. [2010] based upon airborne measurements of various constituents (including our CH_2O measurements) carried out during the TexAQS I and II campaigns examined temporal trends for the 2000 to 2006-time period. Based upon trends in the ratios of ethene to NO_x and propene to NO_x observed for isolated petrochemical sources (6 facilities: Sweeny, Freeport, Chocolate Bayou, Texas City, Mont Belvieu, and the Houston Ship Channel), this study reported a 30% ± 30% decrease in these ratios over this 6-year period with significant day-to-day and within plume variability (-50% to +100%). The median CH_2O concentration, based upon our measurements, decreased by ~ 40% for this same sampling region over this 6 year period. It is important to note that this 40% drop in CH_2O is consistent with a ~ 30 to 40% drop in CH_2O from 24-hour averaged DNPH (2,4-dinitrophenylhydrazine) cartridge measurements over this same 6 year period. The Texas Commission on Environmental Quality (TCEQ) acquired such measurements every 6^{th} day at the Clinton, Deer Park and Channelview sites, which are very close to the Houston Ship Channel. Despite this consistency, Washenfelder et al. [2010] point out that measured ratios for ethene to NO_x and propene to NO_x exceed emission inventories for the above 6 facilities by factors of 1.4–20 and 1–24, respectively, using the updated 2006-point source emission inventories provided by TCEQ. De Gouw et al. [2009], furthermore, report ethene emissions a factor of 6 higher than the 2006 TCEQ point source emission inventory for the Mont Belvieu industrial complex. Accurate predictions of future O_3 levels in the greater Houston area employing chemical transport models require that the above emission inventory discrepancies get resolved.

In addition, although there is consistency between the ground-based DNPH CH_2O temporal trends and those inferred by aircraft measurements between the 2000 and 2006 TexAQS studies, it is highly desirable to further extend these temporal comparisons out to 2013. A major benefit of this process is that the highly accurate in situ CH_2O measurements acquired on the P3 aircraft during spirals and missed approaches close to the DNPH sampling sites can be used to further validate the cartridge results under a variety of conditions. This is important since past studies by Herrington and Hays [2012] and by Gilpin et al. [1997] have shown that DNPH cartridge determinations of CH_2O can contain systematic biases even when KI O_3 traps are employed.

2.0 Statement of Work

2.1 Objectives of the Present Study

The overall objective of the present study is to address the 3 major issues discussed above. To accomplish this, the proposing team will analyze ambient CH_2O data they acquired on the P3 and DC-8 aircraft over the greater Houston area in the summer of 2013 during the DISCOVER-AQ

(Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality) and SEAC^{4}RS (Studies of Emissions and Atmospheric Composition, Clouds and Climate Coupling by Regional Surveys) studies. The Fried group deployed two highly sensitive, selective, accurate, and fast airborne spectrometers for CH_2O on NASA's P3 and DC-8 aircraft platforms during these campaigns. The analysis will rely on the Community Multiscale Air Quality (CMAQ) model with Process Analysis, in very high-resolution mode (1 km resolution), driven by the WRF (Weather Research and Forecasting) meteorological model. The following 7 specific tasks will be carried out.

2.2 Specific Implementation Tasks

1. Prepare WRF and CMAQ input files and run the models using nested domains down to a horizontal resolution of 1 km using 2012 TCEQ emission inventory. Drs. Loughner & Pickering will be responsible for this task.
2. Identify time periods when sampling large petrochemical refinery plumes as well as other clearly identifiable sources (e.x., ship plumes, etc.) close to their source and downwind and compare the high resolution WRF-CMAQ model with measurements at these times to arrive at updated emission rates for CH_2O. CMAQ will be re-run with the updated emissions. CMAQ output will be analyzed along back trajectories calculated from WRF model output to assess upstream influence. The whole team, which includes Dr. Fried's group at the University of Colorado (CU) and Drs. Loughner & Pickering, will be responsible for this task.
3. Working with Sept. 25, 2013 data, a day with very favorable meteorological conditions and large emissions from the Baytown Exxon/Mobil petrochemical facility, the high resolution WRF-CMAQ model with updated emissions from (2) will be analyzed along a forward trajectory calculated from the WRF output south to Smith Point to help in validating the model meteorology and the chemistry. During the 2nd DISCOVER-AQ circuit (around noon local time) on this day the wind direction and speed changed (see Fig. 1). During the 1st circuit, the winds were out of the southwest with a well-defined capped inversion layer, allowing the P3 to capture a significant portion of the Exxon-Mobil petrochemical plume. During the 2nd circuit the wind direction shifted to the southeast and the speed diminished by a factor of ~ 4 to around 1.6 m/s. As a result the boundary layer over the ship channel all the way down to Smith Point experienced the significant direct Exxon-Mobil emissions as well as the photochemically produced products down at Smith Point. This situation will provide an excellent opportunity to compare modeled concentrations with ground-based and airborne observations at Smith Point. The entire team will be involved in this activity.
4. Examine the CMAQ model output run with the Process Analysis Mode to quantify the relative importance of the three major CH_2O sources (primary emissions, secondary photochemical production, and regional transport), first on the Sept 25 plume and then on the HGBMA. Drs. Loughner & Pickering will be responsible for this task.
5. Tabulate optimal time periods for select comparisons of airborne CH_2O measurements with ground and mobile CH_2O measurements, focusing on overflights close to DNPH cartridge sampling sites at Clinton, Deer Park and Channelview. Dr. Fried and his team will be responsible for this task. Compare integrated DNPH measurements with *24-hour synthesized integrated airborne measurements* based upon the temporal dependence calculated from the CMAQ model and the P3 aircraft measurements acquired at different

times throughout the day. The entire team will be responsible for this task.

6. Investigate where appropriate airborne CH_2O measurements as well as other measurements from the P3 and DC-8 flights to assess production of CH_2O from flares and isoprene using isoprene, MACR, and MVK measurements and CMAQ model output to derive a biogenic contribution to CH_2O in the HGBMA. The entire team will be responsible for this task.
7. Submission of monthly technical reports by the 8th day of each month with an accompanying financial report submitted by the 12th day of each month throughout the project duration. A final technical report will be submitted by June 30, 2015, preceded by a draft final report on May 20, 2015. Other reports (e.g., Executive Summary, Quartiles) will be submitted as requested by AQRP.

Section 2.3: Deliverables

1. Develop a project Work Plan, which includes a background introduction of this project, a statement of work (including goals, tasks, key personnel, deliverables, and schedule), and a budget with justification.
2. Develop a Quality Assurance Project Plan (QAPP).
3. Several reports will be submitted on a timely basis and at regular intervals. A description of the specific reports to be submitted and their due dates are outlined below. One report per project will be submitted (collaborators will not submit separate reports), with the exception of the Financial Status Reports (FSRs). The lead PI will submit the reports, unless that responsibility is otherwise delegated with the approval of the Project Manager. All reports will be written in third person and will follow the State of Texas accessibility requirements as set forth by the Texas State Department of Information Resources. Report templates and accessibility guidelines found on the AQRP website at http://aqrp.ceer.utexas.edu/ will be followed.

Executive Summary

At the beginning of the project, an Executive Summary will be submitted to the Project Manager for use on the AQRP website. The Executive Summary will provide a brief description of the planned project activities, and will be written for a non-technical audience.

Due Date: Friday, May 30, 2014

Quarterly Reports
The Quarterly Report will provide a summary of the project status for each reporting period. It will be submitted to the Project Manager as a Word doc file. It will not exceed 2 pages and will be text only. No cover page is required. This document will be inserted into an AQRP compiled report to the TCEQ.

Due Dates:

Report	Period Covered	Due Date
Quarterly Report #1	June, July, August 2014	Friday, August 29, 2014
Quarterly Report #2	September, October, November 2014	Monday, December 1, 2014
Quarterly Report #3	December 2014, January & February 2015	Friday, February 27, 2015
Quarterly Report #4	March, April, May 2015	Friday, May 29, 2015
Quarterly Report #5	June, 2015	June 30, 2015

Technical Reports and Data Deliverables
Technical Reports will be submitted monthly to the Project Manager and TCEQ Liaison as a Word doc using the AQRP FY14-15 MTR Template found on the AQRP website.

Due Dates:

Report	Period Covered	Due Date
Technical Report #1	June 1 - 30, 2014	Tuesday, July 8, 2014
Technical Report #2	July 1 - 31, 2014	Friday, August 8, 2014
Technical Report #3	August 1 - 31, 2014	Monday, September 8, 2014
Technical Report #4	September 1 - 30, 2014	Wednesday, October 8, 2014
Technical Report #5	October 1 - 31, 2014	Monday, November 10, 2014
Technical Report #6	November 1 - 30 2014	Monday, December 8, 2014
Technical Report #7	December 1 - 31, 2014	Thursday, January 8, 2015
Technical Report #8	January 1 - 31, 2015	Monday, February 9, 2015
Technical Report #9	February 1 - 28, 2015	Monday, March 9, 2015
Technical Report #10	March 1 - 31, 2015	Wednesday, April 8, 2015
Technical Report #11	April 1 - 28, 2015	Friday, May 8, 2015
Technical Report #12	May 1 - 31, 2015	Monday, June 8, 2015
Project Data (observations, model inputs and outputs, and analysis products)	June 1, 2015 – June 30, 2015	July 30, 2015

Financial Status Reports
Financial Status Reports will be submitted monthly to the AQRP Grant Manager (Maria Stanzione) by each institution on the project using the AQRP FY14-15 FSR Template found on the AQRP website.

Due Dates:

Report	Period Covered	Due Date
FSR #1	June 1 - 30, 2014	Tuesday, July 15, 2014
FSR #2	July 1 - 31, 2014	Friday, August 15, 2014
FSR #3	August 1 - 31, 2014	Monday, September 15, 2014
FSR #4	September 1 - 30, 2014	Wednesday, October 15, 2014
FSR #5	October 1 - 31, 2014	Monday, November 17, 2014
FSR #6	November 1 - 30 2014	Monday, December 15, 2014
FSR #7	December 1 - 31, 2014	Thursday, January 15, 2015
FSR #8	January 1 - 31, 2015	Monday, February 16, 2015
FSR #9	February 1 - 28, 2015	Monday, March 16, 2015
FSR #10	March 1 - 31, 2015	Wednesday, April 15, 2015
FSR #11	April 1 - 28, 2015	Friday, May 15, 2015
FSR #12	May 1 - 31, 2015	Monday, June 15, 2015
FSR #13	June 1 - 30, 2015, Final FSR	Wednesday, July 15, 2015

Draft Final Report
A Draft Final Report will be submitted to the Project Manager and the TCEQ Liaison. It will include an Executive Summary. It will be written in third person and will follow the State of Texas accessibility requirements as set forth by the Texas State Department of Information Resources.

Due Date: Monday, May 18, 2015

Final Report
A Final Report incorporating comments from the AQRP and TCEQ review of the Draft Final Report will be submitted to the Project Manager and the TCEQ Liaison. It will be written in third person and will follow the State of Texas accessibility requirements as set forth by the Texas State Department of Information Resources.

Due Date: Tuesday, June 30, 2015

Project Data
The final report and presentation will cover the results of tasks:

a. 2.2.2 updated emission inventories for both CH_2O and its HRVOC precursors

b. 2.2.3 employ opportunistic days like Sept. 25 and others to help in validating WRF-CMAQ model meteorology and the chemistry.

c. 2.2.4 quantify the relative importance of the three-major CH_2O sources (primary emissions, secondary photochemical production, and regional transport) for select opportunistic days.

d. 2.2.6 derive *24-hour synthesized integrated airborne measurements* and provide comparisons with integrated DNPH measurements. This will help to validate the DNPH temporal trends.

e. 2.27 derive the biogenic contribution to CH_2O production over the HGBMA

All high-resolution WRF-CMAQ model runs, with updated emissions inventories and other inputs as well as outputs, will be submitted along with appropriate CH_2O measurement comparisons. In addition, all comparisons of the *24-hour synthesized integrated airborne measurements* with 24-hour integrated ground-based DNPH cartridge measurements will be submitted. These results will be submitted electronically, the exact format for which will be determined by AQRP.

2.4 Project Timeline

The following table describes the project timeline.

	2014							2015					
Task	June	J	A	S	O	N	Dec	Jan	F	M	A	M	June
Contract, Work Plan, QPP													
Task 1 - Prepare WRF & CMAQ													
Task 2 - Run Models, Update Emissions													
Task 3 - Sept. 25 Analysis													
Task 4 - Source Apportionment													
Task 5- Aircraft/DNPH Comparisons													
Task 6 - Assess Other CH2O Sources													
Task 7 - Monthly Technical and Financial Reports													
Draft Final Report, AQRP Review													
AQRP/TCEQ Presentation													
Final Report													

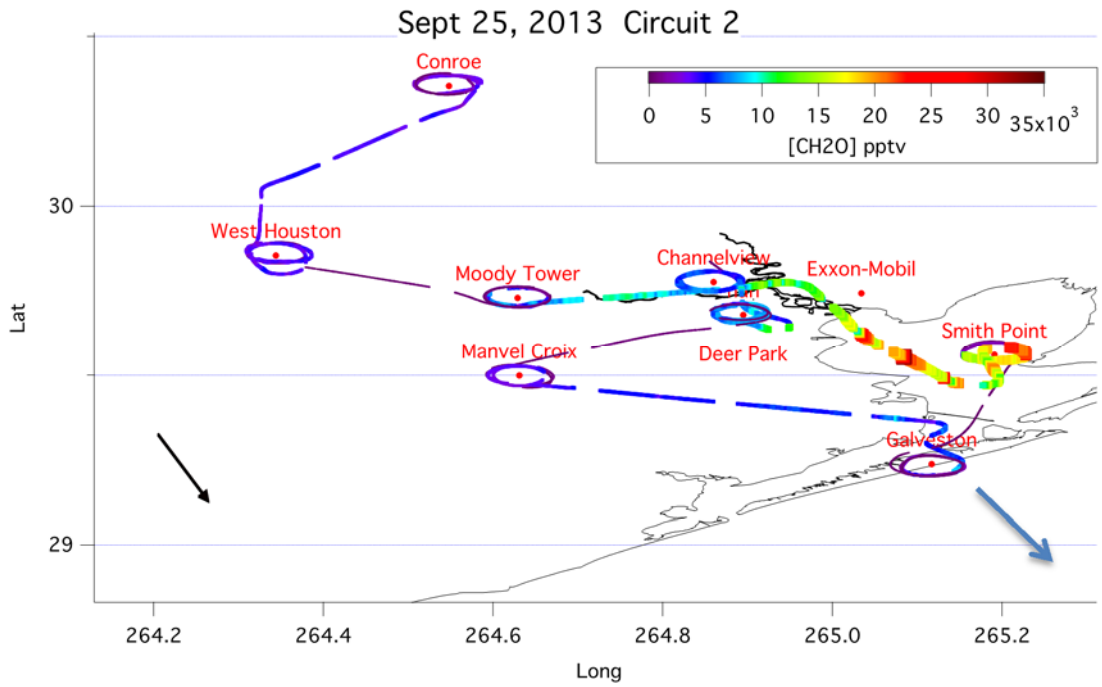

Figure 1: CH_2O concentrations measured on the NASA P3 during the Sept. 25, 2013 DISCOVER-AQ 2nd transit over the HGBMA. The wind direction during the start of this circuit is shown by the arrow at the lower right.

3.0 References

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