Scope of Work For

Project 17-032

Spatial Mapping of Ozone Formation near San Antonio

Prepared for

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

By

Ezra Wood
Department of Chemistry, Drexel University

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Version 2

QA Requirements: Audits of Data Quality: 10% Required
Report of QA Findings: Required in Final Report

Approvals

This Scope of Work was approved electronically on 09/29/2016 by Gary McGaughey, The University of Texas at Austin

Gary McGaughey
Project Manager, Texas Air Quality Research Program

Contents

1.0 Abstract

San Antonio will likely violate the new National Ambient Air Quality Standard for ozone (O_3) of 70 ppb. As a result, regulators will need to make science-based decisions on effective mitigation strategies, including emission reduction programs. Such decisions will require knowledge of the amount of ozone that is transported into the city from upwind (usually southeast of San Antonio), the absolute rates of ozone formation in and around San Antonio, the relative importance and interaction of various emission sources (e.g., upwind oil and gas activity and urban emissions from the city itself), and when and where ozone formation is NOx-limited or VOC-limited. In contrast to Houston and Dallas, little is known about ozone formation in San Antonio. The goals of this research project are to address this major shortcoming and elucidate the mechanisms and rates of ozone formation that affect air quality in San Antonio using novel measurements of peroxy radicals aboard a mobile supersite during a 3-week field project during late Spring of 2017. Instantaneous ozone production rates $P(O_3)$ and $P(O_3)$ solely from alkyl peroxy radicals will be quantified aboard the Aerodyne Mobile Laboratory using new but tested measurements of total peroxy radicals and alkyl peroxy radicals. Measurements of total hydroxy and alkyl nitrates will also be used to investigate the role of alkanes and $RONO_2$ formation as a terminator of ozone chemistry.

This research project directly responds to two of the ten research priorities identified in the AQRP Strategic Research Plan FY 16-17: 1. Improving the understanding of ozone and particulate matter formation (in central Texas), and 2. Quantifying the local ozone production that impacts the design value (DV) monitors that exceed the NAAQS in San Antonio. These research priorities will be addressed by the analysis of data generated by the planned air quality field study in central Texas.

2.0 Background

Ozone is the main component of smog and has adverse effects on human health and vegetation. Unlike primary pollutants like carbon monoxide or black carbon, ozone is formed by photochemical reactions involving volatile organic compound (VOCs) and nitrogen oxides (NOx). The photo-oxidation of propane (a component of natural gas) serves as a simple example of this chemistry:

$C_3H_8 + OH + O_2 \rightarrow C_3H_7O_2 + H_2O$	R1
$C_3H_7O_2 + NO \rightarrow C_3H_7O + NO_2$	R2
$C_3H_7O + O_2 \rightarrow CH_3COCH_3 + HO_2$	R3
$HO_2 + NO \rightarrow OH + NO_2$	R4

The NO_2 formed by reactions 2 and 4 will undergo photolysis during the day, thereby forming ozone (O_3):

NO_2 + sunlight $\rightarrow NO + O$	R5
$O + O_2 + M \rightarrow O_3 + M$	R6

Thus the rate at which ozone is formed is effectively equal to the rate at which NO is converted to NO_2 by reaction with peroxy radicals (in this case, $C_3H_7O_2$ and HO_2):

$$P(O_3) = k_{HO2+NO}[HO_2][NO] + k_{RO2+NO}[RO_2][NO] \qquad \text{Eq. 1}$$

"RO_2" represents all organic peroxy radicals (e.g., CH_3O_2, $C_2H_5O_2$, etc.)

Due to the various radical termination steps such as formation of H_2O_2 and HNO_3, the value of $P(O_3)$ does not always simply increase with increased concentrations of VOCs or NOx. Ozone production is said to be "NOx-limited" if, due to low NO concentrations, peroxy radicals react with themselves rather than with NO. Conversely, ozone formation is "VOC-limited" (or "NOx-saturated") if HOx radicals (OH, RO_2, HO_2) are main lost via reactions with NO_X. Knowing in which chemical regime an air mass resides is crucial for designing effective ozone abatement strategies, since reducing NOx emissions can lead to undesirable *increases* in ozone formation rates if the air is in a VOC-limited state. This is the case in southern California, evident by the higher ozone observed on weekends when there is reduced NOx emissions due to lower diesel truck traffic (Pollack et al., 2012).

To address ozone air quality problems, regulators need to know the following:
- Which VOCs act as the "fuel" for ozone formation and from which emission sources?
- What are the absolute ozone production rates in ppb/hr?
- How much ozone is produced locally and how much is transported from upwind?
- Does ozone formation occur under NOx-limited or VOC-limited chemical conditions?

The answers to these questions for the San Antonio area are barely known due to the paucity of studies on the topic. In contrast, a wealth of knowledge has been gained about ozone formation in Houston due to numerous successful air quality studies over the last two decades. To frame the issue, we briefly summarize what has been learned about photochemical ozone formation in Houston and compare to possible differences that might be found in San Antonio.

1. Highly Reactive VOCs (HRVOCs), especially ethylene and propylene, have frequently been identified as the VOCs that lead to the most severe ozone episodes in Houston (Kleinman et al., 2002, Murphy and Allen, 2005, Ryerson et al., 2003). Emissions of HRVOCs from the petrochemical industry are greatly underestimated in emission inventories (Parrish et al., 2009). The exact responsible emission processes are not fully understood, but it is has been identified that chemical flares can operate in conditions in which VOC emissions are underestimated (Torres et al., 2012, Wood et al., 2012). Furthermore, emissions from flares alone appear to have the ability to significantly affect downwind O_3 concentrations (Al-Fadhli et al., 2011). In contrast, little is known about the most important VOCs for ozone formation in San Antonio. Preliminary analysis of GC data from the Karnes City and Floresville Hospital Blvd sites suggest that alkane concentrations are in general much higher than HRVOCs and may play a more important role.

2. O_3 formation can be rapid in Houston, with $P(O_3)$ values exceeding 40 ppb/hr not uncommon during the summer (Kleinman et al., 2002, Mao et al., 2010). $P(O_3)$ values have not been quantified in San Antonio.

3. $P(O_3)$ in Houston is highest in the late morning, occurs under NOx-saturated (VOC-limited) conditions, and transitions to being NOx-limited in the afternoon. This suggests that reducing VOC emissions is likely to be more effective than reducing NOx emissions in reducing ozone (Mao et al., 2010, Ren, 2015). San Antonio's chemical regime is not well understood.

4. P(HOx) in Houston is dominated by HONO photolysis, the reaction of H_2O with $O(^1D)$ (from O_3 photolysis), and photolysis of oxygenated VOCs like formaldehyde and acetaldehyde (Mao et al., 2010, Ren et al., 2013). When $P(O_3)$ is VOC-limited, the dominant HOx removal mechanism is the reaction of OH with NO_2, forming HNO_3. Formation of organic nitrates is not an important removal process for HOx or NOx. The dominant formation mechanisms for HONO are poorly understood, though NOx is the parent compound. The role of HONO in San Antonio is unknown. Given the possible greater importance of alkanes in San Antonio ozone chemistry, it is conceivable that organic nitrate formation is a significant sink of both NOx and HOx. An example of atmospheric chemistry dominated by C5 and greater alkanes from oil evaporation was observed downwind of the Deepwater Horizon oil spill of 2010 (Neuman et al., 2012). Although alkyl nitrate formation rates were not directly quantified, this study concluded that C5-C9 alkyl nitrate formation accounted for ~75% of NOx oxidation. This resulted in a very short atmospheric lifetime of only 0.8 hrs for NOx – comparable to that if the reaction of NO_2 with 10^7 molecules/cm^3 OH were the only loss process. $RONO_2$ formation was undoubtedly also a large removal process for HOx.

These possible differences listed above, are in need of observation-based investigation.

A preliminary examination of TCEQ monitoring site data during a high ozone episode in San Antonio suggests that there are important contributions to P(O_3) both upwind of and inside San Antonio:
On May 2, 2015 winds were from the Southeast at 5 to 7 miles/hr. Ozone concentrations at the Calaveras Lake site (C59), which is southeast of the urban core of San Antonio and thus upwind of San Antonio that day, were in the mid-60's ppb from 10:00 until 18:00, with daytime nitrogen oxide concentrations between 1 and 2 ppb. Northwest of San Antonio at site C23, which is approximately 10 miles and 1.5 hours downwind of Calaveras Lake, ozone concentrations exceeded 73 ppb from 10:00 until 17:00, reaching a 1-hour maximum of 84 ppb at 12:00, with daytime nitrogen oxide concentrations between 2 and 4 ppb. Auto-GC measurements at the Floresville Hospital Blvd monitoring site (C1038, 30 miles southeast of San Antonio) indicated elevated concentrations of ethane, propane, and butanes, but with sub-ppb concentrations of ethylene and propylene.

The most relevant conclusions to draw from these observations are the following:

1. Elevated light (C2-C5) alkanes, commonly emitted by natural gas extraction and processing activities, had a combined OH reactivity (k_{OH+VOC}[VOC]) greater than that from the alkenes of comparable size.

2. Ozone concentrations reaching the southeastern-most parts of San Antonio were already elevated (60+ ppb) by 10:00, with low nitrogen oxide concentrations.

3. Additional ozone formed in the air after passing over the center of San Antonio, suggesting an important role for emissions from San Antonio itself – possibly nitrogen oxides but impossible to deduce without further information (VOCs are not measured at sites C59 and C23).

4. A very rough ozone production rate of 10 to 15 ppb/hr in between the upwind (C59) and downwind (C23) sites can be estimated based on the increase in ozone observed – not a particularly high value compared to those observed in Houston, but enough to produce an exceedance of the 8-hr NAAQS of 70 ppb at site C23. Higher P(O_3) values would be required if wind speeds aloft were significantly higher than the surface readings.

3.0 Objectives

The overall objectives of this project are to elucidate the cause of high ozone concentrations in San Antonio and to inform regulatory decisions regarding mitigation procedures using analysis of data from an air quality study in and around San Antonio during May and June of 2017. More detailed objectives are to answer the following science questions:

1. What are the rates of instantaneous ozone production ($P(O_3)$) upwind, within the urban core, and downwind of San Antonio? How much ozone is produced locally and how much is transported? During what times of day and where (upwind/downwind) is $P(O_3)$ NOx-limited vs. VOC-limited?
These questions will be addressed with mobile measurements of total peroxy radicals and NO, which will be used with equation 1 ($P(O_3) = k_{HO2+NO}[HO_2][NO] + k_{RO2+NO}[RO_2][NO]$) to calculate the rates of ozone formation ($P(O_3)$). The NOx-limited or VOC-limited nature will be investigated by the relationship between $P(O_3)$ and [NO] and will complement separate analyses using radical budgets and indicator species by collaborators at Aerodyne.

2. What is role of alkanes in O_3 formation? Alkanes comprise the majority of emissions from oil and gas activities but not urban or biogenic emissions. *The role of alkanes in San Antonio ozone formation will be probed with both local and integrated markers:*
A. $P(O_3)$ resulting just from alkyl peroxy radicals (formed by the first generation oxidation of alkanes) will be determined using novel measurements of alkyl peroxy radicals.
B. Measurements of total hydroxy and alkyl nitrates will be used to infer the overall alkyl nitrate branching ratio. Higher values imply an important role of $RONO_2$ as a radical termination step and would likely implicate large (C5-C10) alkanes due to their known high $RONO_2$ branching ratios.

4.0 Task Descriptions

Task 4.1: Recruit post-doc (September 2016 – January 2016)
A postdoctoral associate will be recruited, hired and trained in operation of the ECHAMP radical sensor. If a postdoctoral associate cannot be hired by January or February 2017, a “co-op” student from Drexel will be recruited. Drexel undergraduates complete one to three 6-month co-ops as a requirement for graduation, and work full-time at a non-profit, in industry, or in a research laboratory. This task will be conducted primarily by the PI with assistance provided by the postdoc at Drexel University. The primary milestone from this task is the successful recruitment of either a post-doc or co-op student.

Task 4.2: Laboratory preparation (February 2017 – April 2017)
The ECHAMP peroxy radical sensor will be prepared for integration into the Aerodyne mobile laboratory. The instrument design and operation will be modified as needed due to space constraints. The measurement of alkyl RO_2 measurements will be refined based on first results from laboratory tests last spring and initial field tests from July 2016. The impact of ambient NO

and NO_2 concentrations on the thermal dissociation organic nitrate measurements will be quantified in the laboratory. Finally, the analytical instrumentation will be integrated into the Aerodyne Mobile laboratory. This task will be conducted by the PI and postdoc at Drexel University with assistance from an undergraduate researcher. The outcome of this task will be successful integration of field-ready instruments into the Aerodyne mobile laboratory.

Task 4.3: Field deployment (May 2017 – June 2017)
The three-week field project is planned for May and June in the greater San Antonio area. Measurements of peroxy radicals and NO will be made on board the Aerodyne mobile laboratory at the Gulf Coast (e.g., Corpus Christi), northwest of San Antonio (usually a downwind high O_3 site), and in between (i.e, upwind of San Antonio and in the urban core region). This task will be conducted by the PI and postdoc at Drexel University in close collaboration with researchers from Aerodyne Research, Inc. The outcome of this task will be the raw data collected during the field deployment.

Task 4.4: Follow-up laboratory work (June 2017 – August 2017)
Following the completion of the field measurements, additional laboratory work will be conducted if necessary as part of the data quality assurance procedure. This would likely involve additional instrumental calibrations and diagnostic tests to ensure the accuracy of the data collected. This task will be conducted by the PI and postdoc at Drexel University. The outcome of this task is a fuller understanding of the performance of the instruments during the field deployment.

Task 4.5: Data work-up and analysis (August 2017)
The raw data collected during the field deployment will be processed to produce the final time series data set as well as calculated ozone production rates as a function of location. Brief analysis of the roles of upwind emission sources and intra-urban sources on the rate of ozone formation will be analyzed. This task will be conducted by the PI and postdoc at Drexel University. The deliverable resulting from this task will be the quality-assured dataset and the project final report which summarizes the preliminary analysis performed.

Task 4.6. Project Reporting and Presentation (September 2016 – August 2017)
As specified in Section 7.0 "Deliverables" of this Scope of Work, AQRP requires the regular and timely submission of monthly technical, monthly financial status and quarterly reports as well as an abstract at project initiation and, near the end of the project, submission of the draft final and final reports. Additionally, the PI or postdoc from the Drexel team will attend and present at the AQRP data workshop. For each reporting deliverable, the lead PI will electronically submit each report to both the AQRP and TCEQ liaisons and will follow the State of Texas accessibility requirements as set forth by the Texas State Department of Information Resources. The report templates and accessibility guidelines found on the AQRP website at http://aqrp.ceer.utexas.edu/ will be followed. Draft copies of any planned presentations (such as at technical conferences) or manuscripts to be submitted for publication resulting from this project will be provided to both the AQRP and TCEQ liaisons per the Publication/Publicity

Guidelines included in Attachment G of the subaward. Finally, our team will prepare and submit our final project data and associated metadata to the AQRP archive.

Deliverables: Abstract, monthly technical reports, monthly financial status reports, quarterly reports, draft final report, final report, attendance and presentation at AQRP data workshop, submissions of presentations and manuscripts, project data and associated metadata

Schedule: The schedule for Task 4.6 "Deliverables" is shown in Section 7.

5.0 Project Participants and Responsibilities

Name	Title/Affiliation	Responsibilities
Ezra Wood	PI, Assoc. Professor, Drexel University Dept. of Chemistry	The PI will oversee, manage, and be directly involved in all tasks.
(to be recruited)	Postdoctoral fellow or co-op student, Drexel University Dept of Chemistry or Dept. of Civil, Architectural and Environmental Engineering	The postdoc or co-op student will conduct most of the day-to-day operations leading up to the field deployment, will conduct most of the field measurements, and contribute to the data analysis.
(to be recruited)	Undergraduate researcher, Drexel University Dept. of Chemistry	The undergraduate will assist in the preparation for the field deployment.

6.0 Timeline

The tasks described in section 4 will be executed following the following timeline:

- *Task 4.1: Recruit post-doc (September 2016 – January 2016)*
- *Task 4.2: Laboratory preparation (February 2017 – April 2017)*
- *Task 4.3: Field deployment (May 2017 – June 2017)*
- *Task 4.4: Follow-up laboratory work (June 2017 – August 2017)*
- *Task 4.5: Data work-up and analysis (August 2017)*
- *Task 4.6. Project Reporting and Presentation (September 2016 – August 2017)*

7.0 Deliverables

AQRP requires certain reports to 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.

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

Abstract Due Date: Wednesday, August 31, 2016

Quarterly Reports: Each Quarterly Report will provide a summary of the project status for each reporting period. It will be submitted to the Project Manager as a Microsoft Word 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.

Quarterly Report Due Dates:

Report	Period Covered	Due Date
Aug2016 Quarterly Report	June, July, August 2016	Wednesday, August 31, 2016
Nov2016 Quarterly Report	September, October, November 2016	Wednesday, November 30, 2016
Feb2017 Quarterly Report	December 2016, January & February 2017	Tuesday, February 28, 2017
May2017 Quarterly Report	March, April, May 2017	Friday, May 31, 2017
Aug2017 Quarterly Report	June, July, August 2017	Thursday, August 31, 2017
Nov2017 Quarterly Report	September, October, November 2017	Thursday, November 30, 2017

Monthly Technical Reports (MTRs): Technical Reports will be submitted monthly to the Project Manager and TCEQ Liaison in Microsoft Word format using the AQRP FY16-17 MTR Template found on the AQRP website.

MTR Due Dates:

Report	Period Covered	Due Date
Aug2016 MTR	Project Start - August 31, 2016	Thursday, September 8, 2016
Sep2016 MTR	September 1 - 30, 2016	Monday, October 10, 2016
Oct2016 MTR	October 1 - 31, 2016	Tuesday, November 8, 2016
Nov2016 MTR	November 1 - 30 2016	Thursday, December 8, 2016
Dec2016 MTR	December 1 - 31, 2016	Monday, January 9, 2017
Jan2017 MTR	January 1 - 31, 2017	Wednesday, February 8, 2017
Feb2017 MTR	February 1 - 28, 2017	Wednesday, March 8, 2017
Mar2017 MTR	March 1 - 31, 2017	Monday, April 10, 2017
Apr2017 MTR	April 1 - 28, 2017	Monday, May 8, 2017
May2017 MTR	May 1 - 31, 2017	Thursday, June 8, 2017
Jun2017 MTR	June 1 - 30, 2017	Monday, July 10, 2017
Jul2017 MTR	July 1 - 31, 2017	Tuesday, August 8, 2017

Financial Status Reports (FSRs): Financial Status Reports will be submitted monthly to the AQRP Grant Manager (Maria Stanzione) by each institution on the project using the AQRP FY16-17 FSR Template found on the AQRP website.

FSR Due Dates:

Report	Period Covered	Due Date
Aug2016 FSR	Project Start - August 31	Thursday, September 15, 2016
Sep2016 FSR	September 1 - 30, 2016	Monday, October 17, 2016
Oct2016 FSR	October 1 - 31, 2016	Tuesday, November 15, 2016
Nov2016 FSR	November 1 - 30 2016	Thursday, December 15, 2016
Dec2016 FSR	December 1 - 31, 2016	Tuesday, January 17, 2017
Jan2017 FSR	January 1 - 31, 2017	Wednesday, February 15, 2017
Feb2017 FSR	February 1 - 28, 2017	Wednesday, March 15, 2017
Mar2017 FSR	March 1 - 31, 2017	Monday, April 17, 2017
Apr2017 FSR	April 1 - 28, 2017	Monday, May 15, 2017
May2017 FSR	May 1 - 31, 2017	Thursday, June 15, 2017
Jun2017 FSR	June 1 - 30, 2017	Monday, July 17, 2017
Jul2017 FSR	July 1 - 31, 2017	Tuesday, August 15, 2017
Aug2017 FSR	August 1 - 31, 2017	Friday, September 15, 2017
FINAL FSR	Final FSR	Monday, October 16, 2017

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. It will also include a report of the QA findings.

Draft Final Report Due Date: Tuesday, August 1, 2017

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.

Final Report Due Date: Thursday, August 31, 2017

Project Data: All project data including but not limited to QA/QC measurement data, metadata, databases, modeling inputs and outputs, etc., will be submitted to the AQRP Project Manager within 30 days of project completion (September 29, 2017). The data will be submitted in a format that will allow AQRP or TCEQ or other outside parties to utilize the information. It will also include a report of the QA findings.

AQRP Workshop: A representative from the project will present at the AQRP Workshop in the first half of August 2017.

Presentations and Publications/Posters: All data and other information developed under this project which is included in **published papers, symposia, presentations, press releases, websites and/or other publications** shall be submitted to the AQRP Project Manager and the TCEQ Liaison per the Publication/Publicity Guidelines included in Attachment G of the Subaward.

8.0 References

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