FUGITIVE BENZENE EMISSION MEASUREMENT TEST REPORT

TONAWANDA COKE CORPORATION
TONAWANDA, NY

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EXECUTIVE SUMMARY

During the week of May 24 the National Physical Laboratory (NPL) of Middlesex, United Kingdom conducted a fugitive benzene emission measurement program at the Tonawanda Coke Facility on River Road in the Town of Tonawanda, New York. Benzene emissions were determined by measuring the benzene concentrations along a series of downwind sites using the NPL's ultraviolet Differential Absorption Lidar (UV-DIAL) and measurements of wind speed and direction.

During this period NPL conducted 80 UV-DIAL scans of 19 different lines of sight from 7 locations at the Tonawanda Coke Facility. These scans were used to estimate concentration maps of the fugitive benzene plume being emitted from the facility. The wind speed data was used to estimate an emission rate from the concentration data.

The results of this fugitive emission measurement program indicate that the site wide benzene emission rate is estimated at between 6.0 and 12.4 kg/hr. Estimates of the fugitive emissions from the byproduct recovery area are in the range of 5.8 to 7.4 kg/hr. By subtracting the average byproduct emission rate from the site wide emission rate the emission estimate for the coke oven battery is between 1.7 and 3.5 kg/hr.

The overall uncertainty in the flux measurements is at least 35% based on the variability of the flux data collected and the non-ideal measurement locations that were influenced by irregularities in the wind field.

The significance of this study is that the byproduct recovery plant was identified as the primary source of fugitive emissions from the facility. Consequently, the byproduct recovery plant offers Tonawanda Coke the greatest opportunity for reducing fugitive emissions.

Given the range of the reported fluxes, the limited meteorological conditions during the test program, and the relatively short duration of the test program, an annualized emission rates cannot be reliably and certainly derived from the measurements.

1.0 INTRODUCTION

In a Section 114 Letter dated June 6, 2009, the United States Environmental Protection Agency (USEPA) directed Tonawanda Coke Corporation (TCC) to quantify the fugitive emissions of benzene from their facility located at 3875 River Road, Tonawanda, New York. On January 7, 2010, USEPA issued an administrative consent order to TCC pursuant to Section 113(a) of the Clean Air Act. The order directed TCC to submit a fugitive benzene emission test (DIAL Test) protocol as required by and in accordance with the Section 114 letter. Conestoga-Rovers & Associates, Inc. (CRA) prepared and submitted to the USEPA a Fugitive Emission Test Plan in April 2010. This plan was approved by the USEPA in a letter dated May 12, 2010.

This report presents the results of the fugitive emission testing that was conducted over the period from May 25 - 28, 2010.

1.1 OVERVIEW

TCC processes metallurgical grade coal to produce foundry coke. The plant encompasses 188 acres and operates 24-hours per day, 365 days per year. There are two types of emission sources at the TCC facility, point and fugitive sources. Point sources at the facility include the boiler exhaust, battery underfire/waste heat stack, and the coke oven gas (COG) flare. Emissions from the boiler and battery stacks will be quantified under a separate test program. Fugitive emissions at coke plants occur through leaks in the battery (top ports, oven doors, and the collector main) and through leaking equipment at the byproduct recovery plant.

The objectives of this fugitive emission test program were to:

- establish emission factors that TCC can use to report benzene emissions
- identify and quantify any significant sources of benzene at the facility.

Fugitive benzene emission testing was conducted using an open path method with analyses by Ultra-Violet Differential Absorption LIDAR (UV-DIAL). During the test program, 80 scans were made of 19 line of sight (LOS) planes at seven different locations within the TCC property. The UV-DIAL data was used to generate benzene concentration maps across each LOS. The data from the concentration maps was combined with the meteorological data to estimate an emission rate or flux from upwind of each LOS.

In order to verify the UV-DIAL benzene concentration measurements, two additional open path measurement systems were used. Open Path Fourier Transform Infrared Spectroscopy (OP-FTIR) and Ultra-Violet Differential Optical Absorption Spectroscopy (UV-DOAS) measurements were made along similar LOS paths as a quality assurance check of the UV-DIAL.

1.2 TEST PROGRAM ORGANIZATION

This fugitive emission test program was carried out under contracts from Hodgson Russ, LLP as outside counsel to TCC. CRA was responsible for managing the field activities, preparing the test plan and the final report. The National Physical Laboratory (NPL), Middlesex, United Kingdom was responsible for the UV-DIAL measurements and the meteorological data collection. ENVIRON International Corporation (ENVIRON), Chapel Hill, North Carolina was responsible for the OP-FTIR measurements and the USEPA collected the UV-DOAS data.

Table 2.1 is a list of project participants and their contact information. The key project participants are as follows:

Mr. Rick Kennedy is outside environmental counsel to TCC. He was responsible for directing work on the project, managing all communications between the Project Team and USEPA.

Mr. Gordon Reusing, P.E. is the Project Manager for this test program. He was responsible for all communications between the client's representative and the field crew. Mr. Reusing reviewed all project deliverables prior to submission to the client's representative.

Mr. Thomas Ferrara is CRA's Field Team Leader and supervised all field activities. Mr. Ferrara was responsible for preparing the test protocol, coordinating data collection and preparing this test report.

Dr. Melanie Williams is the NPL DIAL Project Manager. She was responsible for ensuring timely delivery of DIAL measurements and reports.

Mr. Rod Robinson, a NPL Senior Scientist and the DIAL Team Leader, led the team on the operation, calibration, and maintenance of the DIAL prior to and during field deployment. He also served as liaison with CRA and the site staff.

Dr. Ram Hashmonay of ENVIRON served as a Technical Consultant to CRA and the test program. Dr. Hashmonay was responsible for technical oversight, data analysis and reporting for DIAL and other open-path technologies.

Mr. Mike Chase of ENVIRON served as a FTIR Team Leader. Mr. Chase was responsible for FTIR data collection, analysis and reporting.

Mr. Cary Secrest of the USEPA was responsible for operating the UV-DOAS and reviewing the draft report.

2.0 RESULTS

During the period from May 25 through 28 NPL conducted 80 UV-DIAL scans of 19 different lines of sight from 7 locations at the Tonawanda Coke facility. These scans were used to construct concentration maps of the fugitive benzene plume being emitted from the facility. In addition to the concentration measurements, NPL collected measurements of wind speed and direction from 4 locations. One set of sensors was mounted on the DIAL trailer, two sets of sensors were mounted, at two elevations, on a fixed mast located at the east end of the facility, and a third portable sonic anemometer was deployed near the scan path. The fixed mast had sensor packages at 3 and 11 meters. These two sensors provided the bulk of the wind data for the flux calculations and also were used to construct a wind speed profile so the wind velocities could be adjusted for elevation. The NPL UV-DIAL report is included as Appendix A.

The DIAL trailer was moved to seven different locations during the measurement period. Locations were selected based on the wind direction relative to the facility. Several of the locations were selected such that fluxes of individual units or portions of the plant could be determined. Figures 2.1 through 2.8 show the lines of sight for each location. Table 2.1 shows the start and stop times for each DIAL measurement location, the scan numbers, and the corresponding process data for each period. Gaps in the scan number sequence are the result of calibration scans; instrument check scans and aborted scans. All scans used to report fluxes have been reported.

Once all the flux measurements were compiled, each scan was reviewed and attributed to a specific source area. This review included evaluating the angle between the average wind direction for the period and the line of sight. The plume maps were reviewed to see if the DIAL was capturing the entire plume. The results of this evaluation are presented in Table 2.2. This table shows that 18 scans represent periods where the fugitive plume from the entire facility was captured, 6 scans represent the background, and 3 scans were able to capture the emissions from the byproduct recovery area (process area). The wind flow patterns around the coal handling building, the battery, and the proximity of the byproduct area made measurements of the flux from the battery alone impossible. The three scans attributed to the byproduct area were collected on May 28 at location 6. During these scans the DIAL was set up to scan down the alley between the battery and the byproduct area with north winds.

The fluxes from each of the scans attributed to each source were averaged and the standard deviation of the mean value was used to quantify the uncertainty in the measurements. Table 2.2 shows estimates of the average site wide fugitive benzene emissions to be 9.2 ± 3.2 kg/hr. For the byproduct recovery area the emission estimate is

6.6 ± 0.8 kg/hr. The background was consistently below the DIAL detection limit for benzene and was reported at 0.3 ± 0.5 kg/hr. By subtracting the byproduct area emission estimates from the site wide estimates the fugitive benzene emissions from battery operations is estimated at 2.6 ± 0.9 kg/hr.

2.1 RELATIVE CONSISTENCY OF REPORTED FLUXES

A review of some scans that could potentially represent full or complete scans of the plume cross-section that represent site fluxes included scans 1, 3, 5, 18, 19, 20, 24, 27, 28, 29, 40, 41, 42, 48, 49, 53, 55, 56, 73, 74, 75, 82, (107+108), and 110. Scans 1, 3, and 5 have not been included in our average for the full site emissions because there is some question as to whether the entire plume was captured. Scans 27, 28, 29, 40, 41, 42, 48, 49, 53, 55, and 56 on May 26,have reported fluxes of approximately 9-16 kg/hr and were fairly close to the north side of the process area while scans(107+108) and 110 of May 28, have reported fluxes that are of similar magnitude (~ 9 kg/hr) but are on the south side of the process area and coke ovens indicating reasonable consistency in the reported fluxes measured under two opposite wind directions. DIAL scans 73, 74, 75, and 82 (scan 82 visualised in Appendix A Figure 2.5b1-b2) of May 27, have reported fluxes in the range (3.2 to 5 kg/hr) also representing good consistency. The approximate factor of 2 to 4 difference in the magnitude of the reported fluxes between these sets of scans is evidence of the inconsistent nature of the emissions with some of the difference attributed to errors in the flux measurements.

A review of the DOAS concentration data corresponding to a number of the DIAL scans on May 26 shows relatively high average concentrations associated with high fluxes and relatively low average concentrations with the lower fluxes supporting the variable nature of the emission source.

2.2 UNCERTAINTIES IN FLUX MEASUREMENTS

A complete description of the DIAL technique is provided by NPL in ANNEX 1 of their report located in Appendix A. This review of the potential uncertainties associated with the flux measurements draws from the information provided in the NPL report and an additional NPL report entitled; *A Determination of the Emissions of Volatile Organic Compounds from Oil Refinery Storage Tanks,* NPL Report DQMA (A) 96, October 1993. A copy of this report is provided in Appendix F. Information that could be used to establish a confidence level or measure of uncertainty in the individual flux measurements was not collected during this test program. The total uncertainty in a

particular flux measurement is made up of the associated uncertainties of each variable defining the flux. The DIAL technique defines a differential flux at a given point along a beam path by calculating the product of measured concentration and the normal wind vector at that point. Each point is assumed to be centered on a differential area defined by the spatial range resolution of the DIAL system, which is based on the theoretical range resolution of 3.75 m. The differential fluxes along each beam path that make up a given scan form an array that is subsequently summed to produce the total flux for that scan. The DIAL processing technique compensates for any sparseness of data in the array due to diverging polar beam paths of a given scan through the use of an interpolation algorithm. Thus, uncertainty in the measured flux for a given scan may be attributed to uncertainties in the measured concentration, application of a specific wind vector and the array processing technique.

The range of variability in the beam path concentrations at this site may be inferred from Figure 2.9 of the NPL report. The first 10 minutes of the record indicates an approximate variability of plus or minus 50% about a mean value. The same magnitude of variability can be seen in both the UV-DIAL and UV-DOAS data indicating that the variability is not an artifact of a specific measurement technique. The plume cross-section is continually moving and changing in shape about some time averaged position in response to the dynamic turbulent wind field and may easily account for some of the variability in the measured concentrations.

A detailed discussion of the potential limitations and uncertainties associated with the DIAL technique is given in the NPL report provided in Appendix F. In this report NPL discusses the effects of the wind field at complex industrial topographies and the associated atmospheric and site turbulence structure on the DIAL measurements. Quantitative uncertainties are not given except through recourse to empirical comparisons.

There is significant potential uncertainty in the wind field around complex industrial sites. As noted in the NPL Report in Appendix F, the DIAL measurement plane is ideally selected to be sufficiently downwind of the site so that irregularities induced in the wind field by these structures are substantially averaged out. Typically one would have to be at least 5 characteristic building dimensions downwind to be outside of the building wake and turbulence that affects the wind field. Most of the DIAL measurement planes in this program did not meet these ideal conditions because of the limited scanning locations available. The magnitude of the uncertainty in the flux measurements due to the wind field uncertainty could not be directly quantified with the data collected in this program, however it should be recognized that the expected wind field fluctuations in both speed and direction in this complex industrial

setting would have a very significant affect on the flux calculations, particularly for the scans that were closest to the buildings.

The technique for empirically determining the precision of the flux measurements is to collect as many downwind scans as possible under a variety of meteorological conditions and different orientations to the source. During this study we were limited by the consistent wind conditions to collecting data under generally light wind speeds. Wind directions varied significantly and data was collected across plumes oriented north, east, and south. The uncertainty in the reported values was estimated by calculating the standard deviation of the average flux measurements. The standard deviation of the average facility wide flux measurements was ±3.2 kg/hr or 35%. The standard deviation of the benzene flux estimates for the process area was measured as ±0.8 kg/hr or 12%, however this is based on only 3 scans.

Overall, the uncertainty in the flux measurements are at least 35% based on the variability of the flux data collected during the program. As noted in the NPL report conclusions, a variability of 10% to 15% can be expected for DIAL measurements conducted under ideal conditions, and under more complex conditions such as this site, the DIAL measurements could vary by 20% to 30%. However, since most of the DIAL scans in this project were not conducted sufficiently downwind so that irregularities in the wind field could be averaged out, the uncertainty is higher.

3.0 MEASUREMENT METHODS

3.1 UV- DIAL

A description of the UV-DIAL measurement technique, the results of the TCC monitoring including sample of the DIAL generated plume maps is provided in NPL's report which can be found in Appendix A.

3.2 OP-FTIR

The OP-FTIR instrument was deployed downwind of the suspected sources along a single optical path at a height of approximately two meters above the surface. The OP-FTIR procedures and test results are provided in the ENVIRON report which is included in Appendix B.

3.3 UV-DOAS

The UV-DOAS instrument was deployed downwind of the suspected sources, along a single optical path at a height of approximately two meters above the surface, to collect path-integrated benzene concentration data. The UV-DOAS beam path was co-located as close as possible to the DIAL beam path. The UV-DOAS instrument was operated by the USEPA. The UV-DOAS results and calibration data are provided in Appendix C.

3.4 PROCESS DATA

In order to document the process operations, CRA had an observer on the coke oven battery throughout the test program. The purpose of the observer was to document any abnormal activities or incidents that might have resulted in fugitive benzene emissions. The production rate of the battery is defined by the number ovens pushed in a 24-hour period. The push sheets were collected by CRA and copies are provided in Appendix D.

In addition to the CRA observer, TCC employs Guardian Environmental Associates Inc. to conduct Reference Method 303 observations. TCC had Guardian conduct additional observations to coincide with the DIAL measurements. Copies of the Method 303 observation data sheets are provided in Appendix E.

4.0 QUALITY ASSURANCE

4.1 QA MEASUREMENTS

The data quality assurance measurements for each of the three methods can be found in the respective reports in Appendices A-C.

4.2 INTERCOMPARISON AND SPECTRAL CONFIRMATION

On May 26 the UV-DOAS and the DIAL were set up along similar lines of sight along the northern boundary of the facility. Path integrated benzene concentration data was collected for approximately one hour. Figure 2.9 in Appendix A (NPL Report) shows the concentration data for each instrument. Figure 4.1 is a scatter plot of both data sets with a best-fit line. This analysis shows that there is reasonable agreement between the two data sets (R^2=0.62) with the DIAL measurements being biased higher than the DOAS data by approximately 31% (m=0.6884). This positive bias can be attributed to the differences in elevations of the two instruments beam paths. The UV-DIAL path was approximately 2 meters higher than the UV-DOAS and is assumed to be closer to the plume centerline resulting in higher measured benzene concentrations.

A similar comparison was attempted for the OP-FTIR versus the DIAL measurements on May 27 and 28. On May 27 the OP-FTIR spectral averaged concentration of 19 ppb benzene is in reasonable agreement with the DIAL results as can be seen in the NPL figure in Appendix A for scan 67 Figure 2.5a1. Similarly, scan 110 on May 28 from the DIAL results, Appendix A Figure2.8a1, agrees reasonably well with the OP-FTIR concentration of 36 ppb.