Casting Emission Reduction Program

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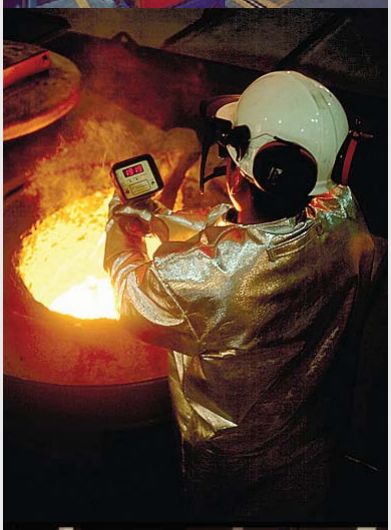

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Method Comparison Between a Cyclone-Impinger and a Dilution Tunnel for the Characterization of Particulate Matter Emissions from a Ferrous Metal Foundry

(Article submitted to the Journal of Air & Waste Management)

1412-319 NA

April 2006

(Revvised for public distribution)

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Method Comparison Between a Cyclone-Impinger and a Dilution Tunnel for the Characterization of Particulate Matter Emissions from a Ferrous Metal Foundry

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This report has been reviewed for completeness and accuracy and approved for release by the following:

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EXECUTIVE SUMMARY

An evaluation and comparison study of two stack sampling methodologies for the collection of particulate matter (PM) emissions from ferrous iron metal casting operations was completed. An EPA Method 201A/Method 202 sampling train (combining cyclone samplers for filterable particulate with an impinger train to capture condensable particulate) was compared to a dilution tunnel sampling method that concurrently collects both filterable and condensable PM. Testing involved the simultaneous collection of continuous stack samples using both sampling methods in a parallel sampling configuration over a seventy-five minute period that included metal pouring, cooling, and shakeout processes. Conventional dry sand molds were made of a 4-on gear pattern (a single mold containing four gear cavities) using Michigan lake sand containing a phenolic urethane No-Bake® binder. PM emission samples from both methods were collected on filters and analyzed gravimetrically. In addition, PM less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) collected from the dilution tunnel sampling method underwent chemical and physical characterization. Results indicate that for all casting processes, the $PM_{2.5}$ mass ratio of the cyclone/impinger method is 2 to 4 times that of the dilution tunnel method. For the pouring and cooling processes alone, the mass ratio increases to over 7 times. Condensable particulate contributed 32-38% to the total particulate mass, with the organic containing portion contributing 96-98%, the balance consisting of inorganic particulate. Of the remaining mass, 52 to 59% consisted of particles greater than PM_{10}, while 4–6% of the mass was in the intermediate range between $PM_{2.5}$ and PM_{10}, and 3–5% was less than $PM_{2.5}$. The total $PM_{2.5}$ fraction was found to be composed of organic carbon (35 - 60%), elemental carbon (20-30%), inorganic materials (10 - 20%), and other elements (4-10%).

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1.0 INTRODUCTION

1.1. CERP Background and Objectives

The Casting Emission Reduction Program (CERP) is a co-operative initiative between the Department of Defense (US Army) and the United States Council for Automotive Research (USCAR). The signers of the CERP Cooperative Research and Development Agreement (CRADA) include: the Environmental Research Consortium (ERC), a partnership of DaimlerChrysler Corporation, Ford Motor Company, and General Motors Corporation; the U.S. Army Research, Development, and Engineering Command (RDECOM-ARDEC); the American Foundry Society (AFS); and the Casting Industry Suppliers Association (CISA). The US Environmental Protection Agency (US EPA) and the California Air Resources Board (CARB) also have been participants in the CERP program and rely on CERP published reports for regulatory compliance data. All published reports are available on the CERP web site at www.cerp-us.org.

CERP's primary purpose is to evaluate materials, equipment, and processes, quality and energy usage in the production of metal castings. Technikon's facility was designed to evaluate alternate materials and production processes designed to achieve significant air emission reductions. The facility's principal testing arena is designed to measure airborne emissions from individually poured molds. This testing arena facilitates the repeatable collection and evaluation of airborne emissions and associated process data.

1.2. Study Implications

Government environmental regulations act as a driving force

in the domestic metal casting industry by stimulating efforts to reduce emissions and costs associated with regulatory compliance. The results of this side-by-side comparison study suggest that the common EPA approved methods used for stack sampling of particulate matter from stationary sources overestimate emitted particulate mass. Traditional hot filter/impinger methods suffer from significant high bias of condensable PM mass which can dominate PM results measured using these methods. This could lead to inaccurate targeting of sources for emission control measures. Significant improvements in cost and compliance may therefore be achieved through the use of dilution tunnel methods for particulate sampling by increasing the accuracy of PM emission measurements.

1.3. Report Organization

This section of the report is designed to document methodology, process or applications of casting industry research. Section 2 of this report includes a summary of the methodologies used for data collection and analysis, procedures for calculations, QA/QC procedures, and data management and reduction methods. Specific data collected during this study are summarized in Section 3 of this report, with detailed data included in the appendices of this report. Section 4 of this report contains a discussion of the results.

The raw data for this test series are archived at the Technikon facility.

2.0 EXPERIMENTAL METHODS

2.1 Test Facility

The comparative stack emissions testing reported here was conducted at Technikon's Research Foundry under the Casting Emissions Reduction Program (CERP), a coalition of government and private industry. The Research Foundry develops new measurement processes for metal casting manufacturing and emissions control. The reports detailing these processes can be found on the CERP website (http://www.cerp-us.org).

Most of the HAP and PM emissions from metal casting operations arise from the organic binders that are used in sand molds and cores. Emissions originate from evaporation of low boiling point solvent binder components or from thermal decomposition, volatilization, and recombination processes when hot metal is poured into a mold. Organic fumes are also released during mold shakeout; the process by which the mold is physically broken apart to release the cooled casting.

2.2 Description of Testing Program

Molds containing four cavities of an irregular gear shape were made with Wexford W450 Lakesand. The binder used was an HA International Techniset® (Westmont, Illinois) phenolic urethane No-Bake® mold binder at 1.8% total binder (based on sand) and was composed of number 6000 Part I resin (55%), 6433 Part II co-reactant (45%), and 17-727 Part III activator at 7% of Part I. This binder is designed for iron applications. The amount of metal melted was determined from the poured weight of the casting and the number of molds to be poured. These parameters resulted in an approximate cast weight for each of the gear molds of approximately 122 lbs.

Replicate test pours were executed on eight mold packages, each package consisting of a single mold containing four gear cavities. Mold weight was approximately 325 lbs of sand. The mold package was placed on a test stand that was enclosed in an emission hood that meets EPA Method 204 requirements for a Total Temporary Enclosure. The initial sand temperature and system process air temperature in the hood enclosure were maintained at 80-90 °F and at least 110 °F, respectively. Molds were poured with Class 30 gray cast iron at 2,600 –2,700 °F through an opening in the top of the enclosure. At the conclusion of the pouring time, which lasted for 12 to 25 seconds, the opening was covered and remained so for the duration of the test. A complete mold cycle consisted of a forty-five minute period that included the metal pouring and cooling processes, a fifteen-minute shakeout of the mold, and an additional fifteen minute cooling period following shakeout. The total mold cycle and sampling time was seventy-five minutes.

2.3 Sampling Procedures

Simultaneous collection of continuous stack emissions was accomplished using a modified Method 201A/202 sampling train (particle cyclone separator followed by an inline glass fiber filter and impinger train) and a dilution tunnel sampling system in a parallel sampling configuration. Method details for the impinger sampling set-up and test procedure may be found elsewhere.[10-13] For these comparison tests, a standard button-hook type sampling probe was used to isokinetically remove samples from the center of the 6 inch diameter horizontal stack. At the probe outlet, an electrically and thermally insulated "Y" connector split the flow into the inlets of both sampling systems. Initially, a PM_{10} cyclone (Graseby Andersen, Smyrna, GA) was installed prior to the impinger train to better correlate resultant particulate masses between the two methods. A modified Method 201A/202 that

additionally utilized a $PM_{2.5}$ cyclone was employed for several of the runs (See Figure 1). This sampling configuration is similar to that of EPA Conditional Test Method 40. A schematic of the foundry and sampling components is shown in Figure 2.

Figure 1. The Impinger Sampling System

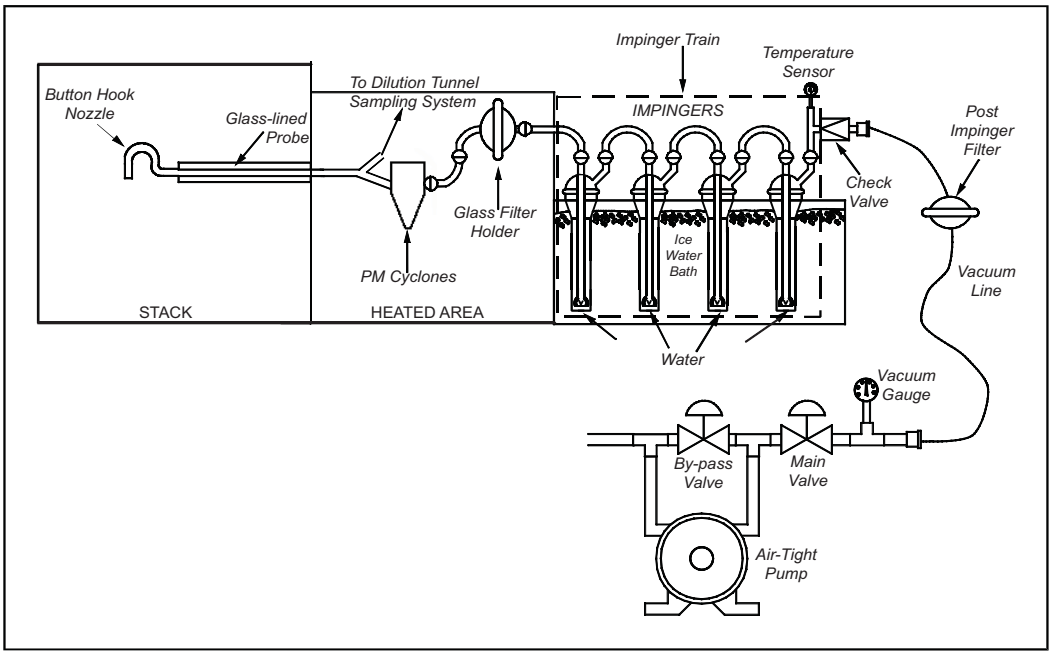
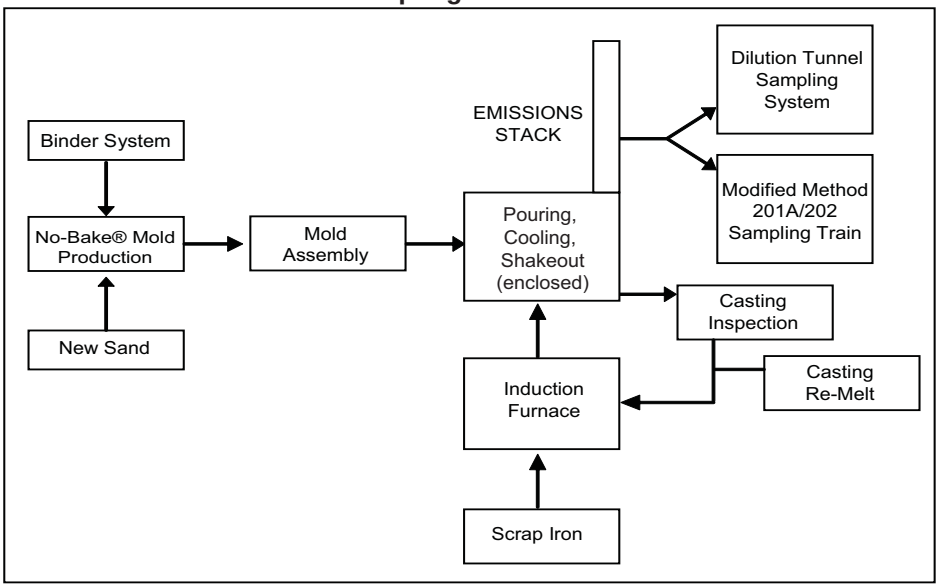


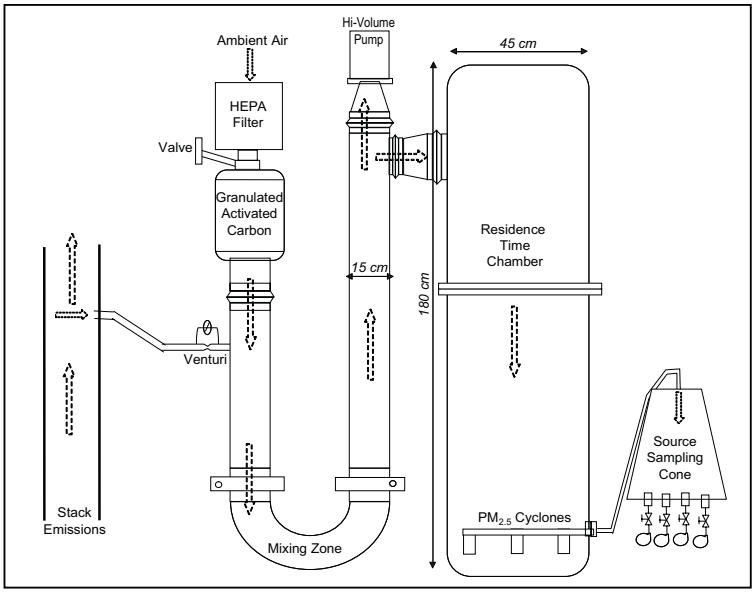
Figure 2. Schematic Diagram of the Research Foundry Process and Sampling Locations



The modified Method 201A/202 impinger method for collection of condensable particulate matter was conducted by Technikon, LLC personnel, while the dilution tunnel sampling was performed by personnel from Desert Research Institute (DRI) using the DRI Source Dilution Sampling System, which is similar to that of EPA Conditional Test Method 39. [14] Particulate emission samples for both the Method 201A/202 sampling train and the dilution tunnel sampling system were collected during the entire mold cycle.

The DRI Source Dilution Sampling System, shown schematically in Figure 3, draws 20-25 liters per minute (lpm) of sample through a venturi flow meter. Concurrently, ambient air at a flow rate sufficient to obtain a predetermined target dilution ratio passes through a high-efficiency particle arresting (HEPA) filter to remove PM, followed by a granulated activated carbon (GAC) bed to remove gaseous species and volatilized PM. The clean, cool ambient air is mixed with the sample gas. At the end of the mixing zone, 113 lpm of the diluted sample is drawn into an aging chamber where nucleation and particle growth may occur. At the outlet of the aging chamber, particles larger than $PM_{2.5}$ are removed by $PM_{2.5}$ cyclones. Equilibrium is achieved with 10 to 60 seconds of aging. A full operational description of the system may be found elsewhere.[15] This system meets the requirements of the proposed ASTM Method D22.03/wk8124 for determining PM from stationary sources.[16]

Figure 3. DRI Source Dilution Sampling System



PM emission samples from both methods were collected on filters and analyzed gravimetrically. Filterable mass was determined

as the difference between pre- and post-sampling filter weight. For the modified Method 201A/202 sampling method, QMA type quartz fiber filters (Whatman, Hillsboro, OR) were used to collect the filterable particulate after separation by the cyclone. For the DRI Source Dilution Sampling System, the mass of $PM_{2.5}$ was measured on Teflon-membrane filters (2.0μm pore size, Gelman Life Sciences, Ann Arbor, MI) with an MT 5 microbalance (Mettler-Toledo, Columbus, OH).

Condensable PM (CPM) was collected by Method 202 in the impinger portion of a Method 5 type sampling train for the modified Method 201A/202 sampling. The solution from the impingers was extracted with methylene chloride to separate the organic and aqueous fractions. After separation, both the organic and aqueous portions were taken to dryness and the residues weighed. All extraction procedures followed the protocols outlined in EPA Method 202.[10]

To ensure the collection of sufficient condensable particulate matter for gravimetric analysis in the impingers, several test runs were sampled for the duration of two mold cycles, whereas one mold cycle supplied sufficient sample for the dilution tunnel sampling system.

Measured metal casting and dry sand process parameters included the weights of the casting and mold, weight loss on ignition (LOI) values for the mold sand prior to the test, and metallurgical data. Monitored stack parameters included temperature, pressure, volumetric flow rate and moisture content. All parameters were maintained within prescribed ranges in order to ensure the reproducibility of the sampling runs and are shown in Table 1.

Table 1 Summary of Casting Process Parameters

Test Parameter	Sample Number			
	2A[1]	Average of 3A and 3B[2]	3C[2]	Average of 4A and 4B
Sand Dispensing Rate, lbs/15 sec	30	30	30	30
Calculated Standard % Binder	1.78	1.78	1.78	1.78
Calculated % Binder (based on sand)	1.81	1.81	1.81	1.81
Mold Weight, lbs	328.5	327.8	332.0	329.4
Calculated Total Binder Weight, lbs	5.85	5.84	5.91	5.86
1800F loss on ignition, %	1.81	1.59	1.73	1.75
Pouring Temperature, °F	2,721	2,635	2,623	2,637
Pouring Time, sec	68	41	29	44
Cast Weight (all metal inside mold), lbs	122.0	105.5	95.6	113.9
Process Air Temperature in Hood, °F	124	121	134	123
Ambient Temperature, °F	64	66	66	66
Mold Age when Poured, hr	43	55	48	39
Test Length, min	36	75	75	71

[1] Power outage after 36 minutes

[2] Samples 3B and 3C had only three cavities filled

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3.0 STUDY RESULTS

3.1 Impinger Method

The results of the PM emissions tests provided the mass of filterable and/or condensable particulate from the binder/sand/metal sample. Filterable PM from the modified Method 201A/202 was determined gravimetrically using an analytical balance (Mettler-Toledo AE163, Columbus, Ohio). CPM was determined by weighing the residues of the organic and aqueous extracted fractions from the impinger train. The sum of both fractions represents the total CPM for the impinger train method.

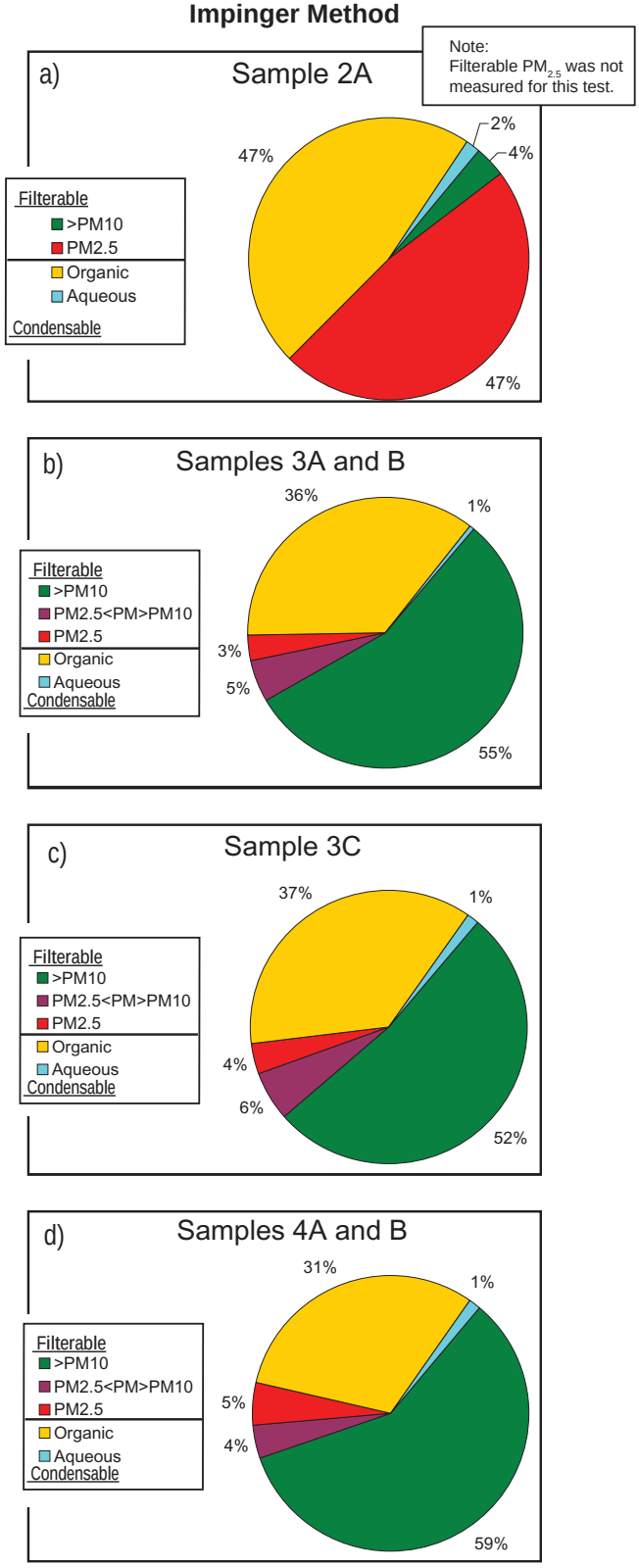
The mass emission rate was calculated using laboratory analytical results, the measured source data, and the weight of each casting. The concentration of the emitted PM was calculated by multiplying the PM mass in the sample with the ratio of total stack gas volume to sampling volume. The total stack gas volume was calculated from the measured stack gas velocity and duct diameter, and corrected to dry standard conditions using the measured stack pressures, temperatures, gas molecular weight and moisture content. The total mass of particulate was then divided by the weight of the metal poured to provide emissions data in pounds of particulate matter per ton of metal (lb/ton). Table 2 includes calculated lb/ton emission factors for both filterable and condensable particulate from the various collected size fractions, as well as the organic and inorganic compo-

Table 2 Average PM Emission Summary Results, Modified Method 201A/202 - Lb/Tn Metal

	Particulate Size Fraction	Sample Number			
		2A	3A & 3 B	3C	4A & 4B
Filterable Particulate	$>PM_{10}$	0.092	6.5804	4.256	6.178
	$<PM_{10}$	1.238	ND[1]	ND	ND
	$PM_{2.5}<PM<PM_{10}$	ND	0.595	0.455	0.408
	$<PM_{2.5}$	ND	0.366	0.304	0.535
	Post Impinger Filter	0.097	0.003	<0.001	0.006
	Total Filterable	1.426	7.544	5.015	7.128
Condensable Particulate	Organic	1.219	4.225	2.973	3.270
	Aqueous	0.040	0.070	0.102	0.141
	Total Condensables	1.259	4.295	3.075	3.411
Filterable and Condensable Particulate	Total PM	2.684	11.838	8.090	10.538
	Total $<PM_{10}$	2.593	5.258	3.834	4.360
	Total $<PM_{2.5}$	1.355	4.663	3.379	3.9518

[1]ND= Not Determined

Figure 4. Relative Percentage of Average Total PM Emissions from the Impinger Method



nents extracted from the condensable fraction collected from the impinger train.

Concentrations for the samples which combined two mold cycles (3A, 3B and 4A, 4B) were averaged using combined sample times and cast weights. These two tests as well as Sample 3C utilized both a PM_{10} and a $PM_{2.5}$ cyclone. For this reason, the PM_{10} fraction from the first cyclone was not collected, but underwent additional separation in the second cyclone. Sample number 2A included data only from the pouring and partial cooling of the mold due to a power failure from an area-wide blackout caused by bad weather. However, this shortened pour presented a unique opportunity to measure the PM resulting from these two processes alone, without the contribution from the shakeout process. In addition, sampling for this test used only a PM_{10} cyclone, whereas the other tests used both PM_{10} and $PM_{2.5}$ cyclones.

Figures 4, 5 and 6 show mass concentration data in lb/ton metal, normalized to 100 percent. In comparing total particulates emitted during the three tests sampled under similar conditions using the same $PM_{10}/PM_{2.5}$ cyclone sampler and impinger train (samples 3A and B, 3C, 4A and B), it is readily apparent that excellent reproducibility was obtained (Figure 4). For each of these runs, 52-59% of the total mass consisted of filterable coarse particles larger than PM_{10}, 4–6% were intermediate filterable particles larger

than $PM_{2.5}$ and smaller than PM_{10}, and 3–5% were filterable particles smaller than $PM_{2.5}$. Condensable particulate (combined organic and aqueous fractions) contributed 32-38% to the total particulate mass, with the organic containing portion contributing 96-98%, the balance consisting of inorganic particulate.

If PM_{10} rather than total PM is considered (Figure 5), the proportionate contributions increase to 9-12% and 7-12% for intermediate PM and $PM_{2.5}$, respectively, while that for condensable PM increases to 79-82% of the total particulate mass. As shown in Figure 6, the emission factors for both the filterable and condensable fractions of $PM_{2.5}$ are accordant with those for the larger size fractions.

Although Sample 2A test conditions differed from the other tests in the shortened testing period that was composed of pouring and partial cooling before the run was terminated, as well as in the use of a single PM_{10} cyclone, the condensable $PM_{2.5}$ fraction contribution remained consistent with the other samples at 97% organic and 3% inorganic. The comparative mass fraction of $PM_{2.5}$ is less than the other size fractions regardless of which particulate size range is considered, although as the size fraction under examination decreases the proportionate amount of $PM_{2.5}$ increases.

During shakeout, the mold physically breaks down releasing particulates, heat and fumes

Figure 5. Relative percentage of average PM_{10} Emissions from the Impinger Method

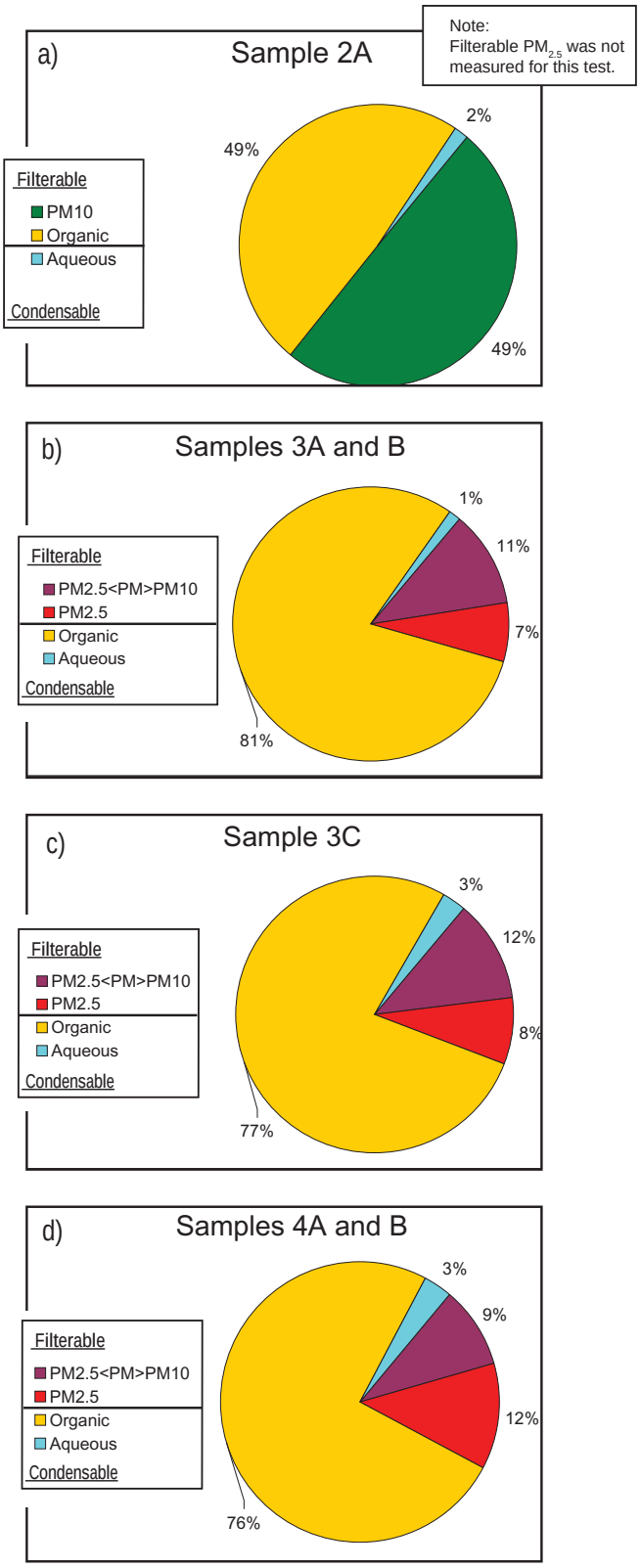
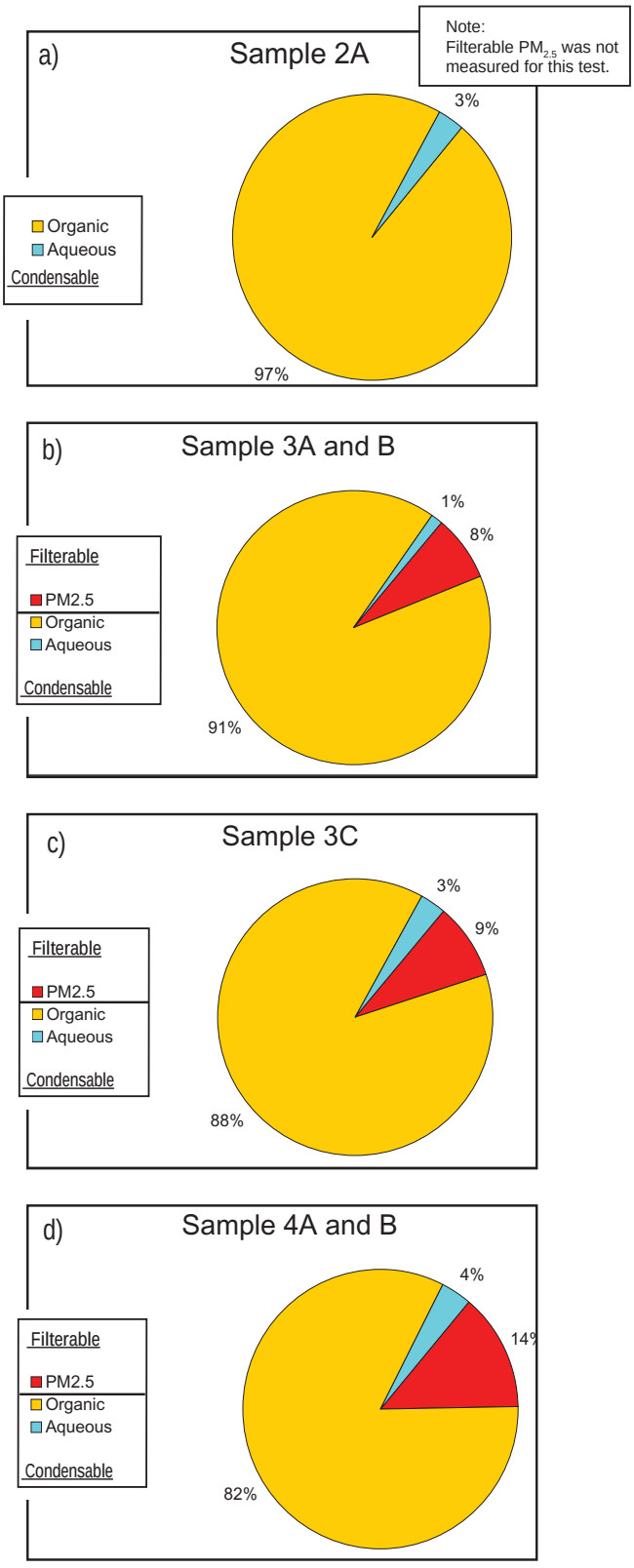


Figure 6. Relative Percentage of Average $PM_{2.5}$ Emissions from the Impinger Method



causing an increase in larger particulate due to suspension, and finer particulate due to nucleation and growth. The duration of the individual processes in the mold cycle including the pouring, cooling and shakeout and increase in temperature at shakeout is illustrated in Figure 7.

3.2 Dilution Tunnel Method

PM collected and measured from the dilution tunnel method represents both filterable and condensable particulate and is presented in Figure 8 as $PM_{2.5}$ mass concentration in lbs/ton.

$PM_{2.5}$ emission factors determined by the dilution tunnel method on a lb/ton metal basis range from 0.84 to 1.8 as is shown in Figure 8 (samples 3A, 3C, 4A, and 4B). Without the contribution from the shakeout process, the pouring and cooling processes (which are evidenced by Sample 2A) resulted in an approximate ten-fold decrease in the mass of $PM_{2.5}$.

In addition to gravimetric analysis, $PM_{2.5}$ from the dilution sampling system was analyzed for 40 elements using energy dispersive x-ray fluorescence (ED-XRF), and organic and inorganic carbon was determined by a DRI Model 2001 thermal/optical carbon analyzer. Particle size and number distributions were measured by an Electric Low Pressure Impactor (ELPI) that collected data

Figure 7. Mold Process Time Intervals and Stack Temperature Profile

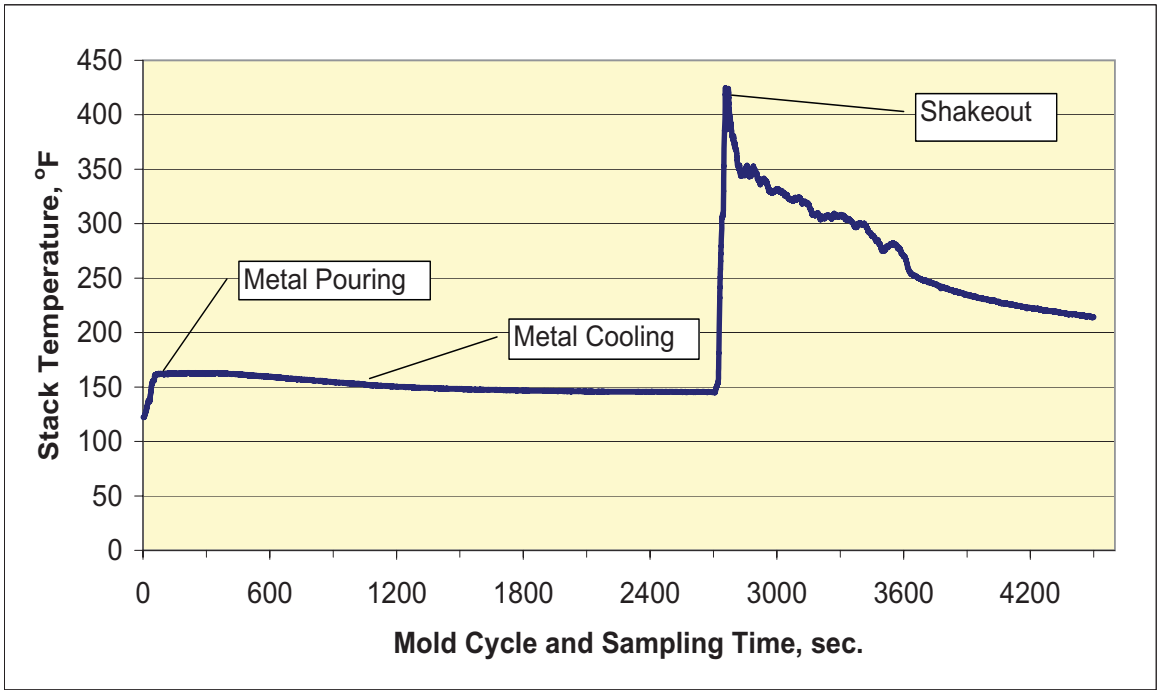
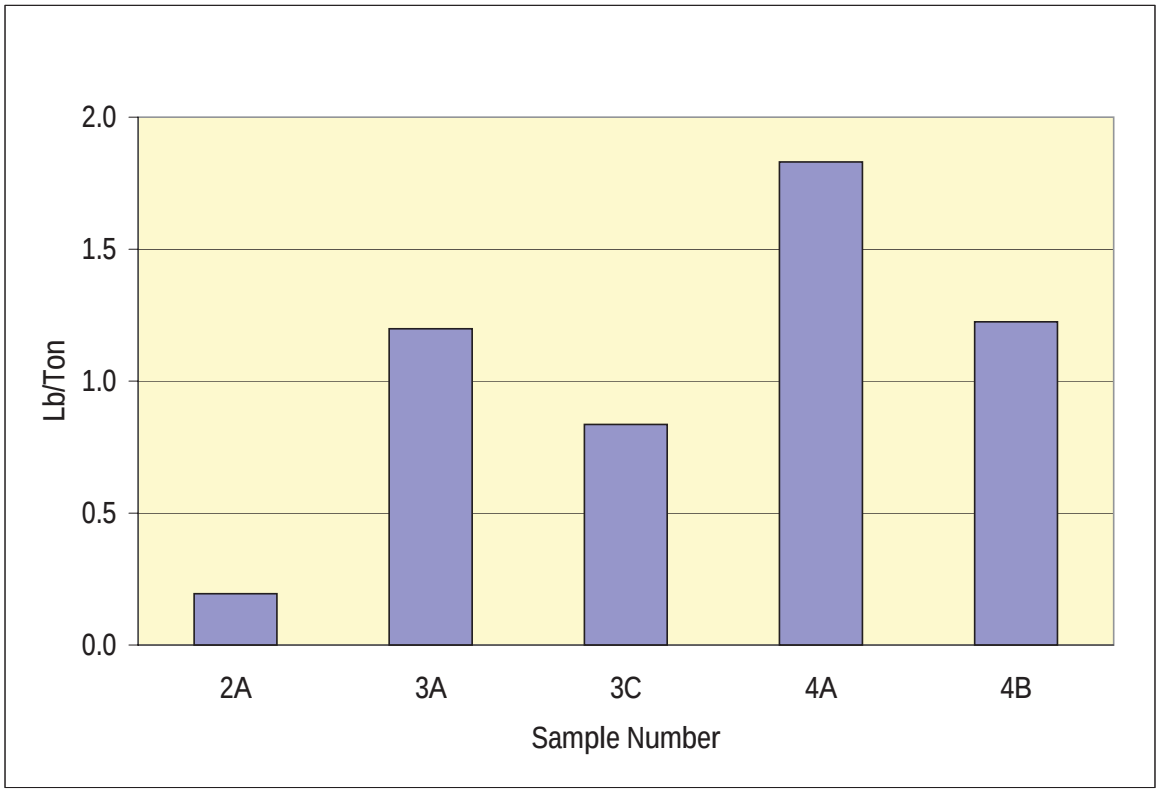


Figure 8. Average $PM_{2.5}$ Mass Emission Factors in Lb/Tn, Dilution Tunnel Method



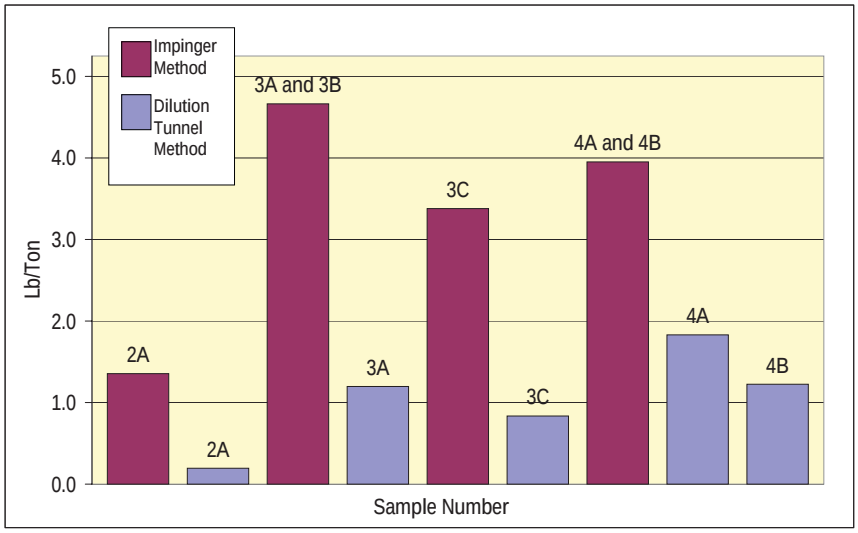
continuously during testing. Ion chromatography was used for the analysis of gaseous ammonia and chloride, nitrate, sulfate, and ammonia ions. Results from these analyses are reported elsewhere.[16]

3.3 Method Comparison

Because the Dilution Tunnel Sampling Method uses a $PM_{2.5}$ cyclone, further discussion will focus on results related to $PM_{2.5}$. Unlike the impinger method for PM sampling, there is no distinction in the dilution tunnel method between condensable or filterable particulate as both are collected on the filters after the cyclones. For that reason, dilution sampling results do not quantify filterable and condensable PM separately. Comparisons to this method are therefore based on the summation of the filterable and condensable $PM_{2.5}$ obtained from the filter and impinger of the modified Method 201/202A method.

In comparing the total $PM_{2.5}$ mass ratio results for all the samples run under identical conditions (which excludes Sample 2A), the impinger method shows an over-estimation factor of 2 to 4 times that obtained by the dilution tunnel sampling method, as shown in Figure 9. For the metal pouring and cooling processes which were sampled separately in Test 2A, the mass ratio of the $PM_{2.5}$ filter plus

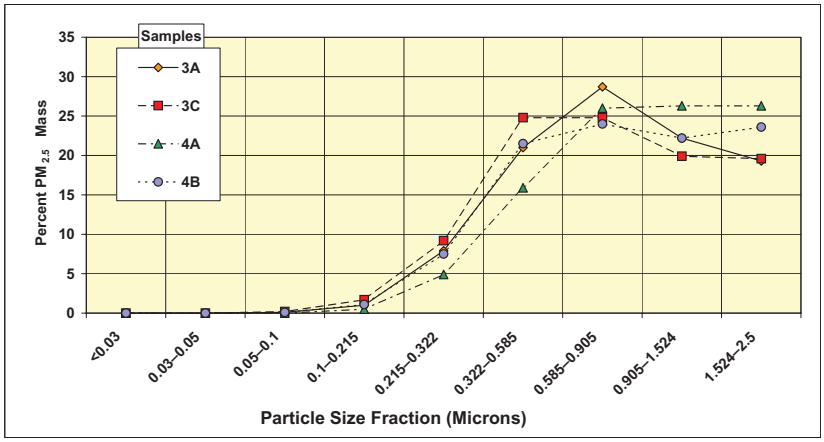
Figure 9. Method Comparison of $PM_{2.5}$ Emissions, Lb/Tn Metal



the impinger residue increased to seven times that of the dilution sampler emission factor. Different particulate formation mechanisms between the pouring, cooling, and shakeout processes account for variations in particle number and mass distributions.[15]

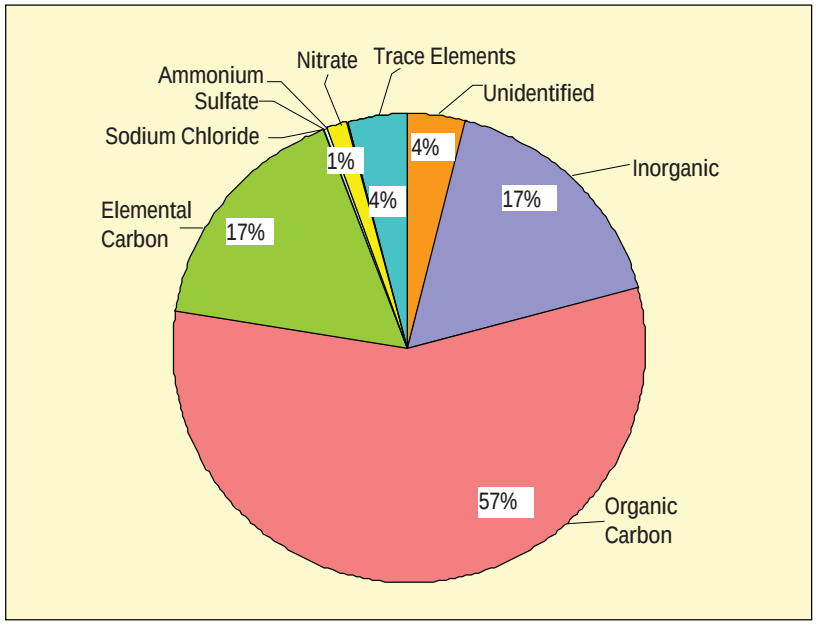
Results from the ELPI indicate the $PM_{2.5}$ mass for these samples is composed mainly of particles larger than 0.215 µm, which is illustrated graphically in Figure 10.

Figure 10. Percent $PM_{2.5}$ Mass Distribution by Size Fraction as Determined by ELPI



The resultant chemical analysis and elemental speciation conducted on $PM_{2.5}$ collected from the dilution tunnel and represented by Sample 3A is shown in Figure 11 as a pie chart. As illustrated by this figure, most of the mass for the sample was found to be composed of organic carbon (35 - 60%), elemental carbon (20 - 30%), inorganic materials (10 - 20%), and other elements (4 - 10%). Unidentified materials range from 0 - 20% of the total mass. This unidentified fraction is the difference between the gravimetric mass and the reconstructed mass determined through chemical speciation.

Figure 11. Chemical Analysis of $PM_{2.5}$ from Sample 3A Obtained by Dilution Tunnel Sampling



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4.0 CONCLUSIONS

An evaluation and comparison of two stack sampling methodologies for the collection of particulate matter from stationary sources was undertaken using emissions from gray iron metal casting operations, including metal pouring, cooling and shakeout processes. The data indicate that the traditional methods for PM collection using a cooled impinger sampling train overestimate the amount of emitted particulate relative to cooling of the emissions through dilution. Condensable particulate mass determined by this method consisted of greater than 95% organic compounds. Metal pouring and cooling processes contribute less to particulate formation than does the shakeout process. More extensive comparative testing needs to be conducted at a production foundry to further understand the relationship between process event and particulate formation and release.

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APPENDIX A ACRONYMS AND ABBREVIATIONS

AFS	American Foundry Society
ARDEC	(US) Army Armament Research, Development and Engineering Center
ASAM	Association for the Standardization of Automation and Measurement Systems
BO	Based on ( ).
BOS	Based on Sand.
CAAA	Clean Air Act Amendments of 1990
CARB	California Air Resources Board
CEMS	Continuous Emissions Monitoring Systems
CERP	Casting Emission Reduction Program
CISA	Casting Industry Suppliers Association
CO	Carbon Monoxide
COR	Contracting Officer's Representative
CRADA	Cooperative Research and Development Agreement
DOD	Department of Defense
DOE	Department of Energy
EEF	Established Emission Factors
EPA	Environmental Protection Agency
ERC	Environmental Research Consortium
FID	Flame Ionization Detector
GC	Gas Chromatograph
GS	Greensand
HAP	Hazardous Air Pollutant defined by the 1990 Clean Air Act Amendment
HC as Hexane	The quantity of undifferentiated hydrocarbons determined by Wisconsin Cast Metals Association – maximum potential to emit method, revised 07/26/01.
I	Invalidated Data
IVI	Interchangeable Virtual Instruments
Lb/Lb	Pound per pound of binder used
Lb/Tn	Pound per ton of metal poured
LOI	Loss on ignition

MACT	Maximum Achievable Control Technology
MMS	Mixing, Making, Storage
MSDS	Material Safety Data Sheets
NA	Not Applicable; Not Available
NCMS/UAB	National Center of Manufacturing Science/University of Alabama at Birmingham
ND	Non-Detect
NESHAPs	National Emission Standards for Hazardous Air Pollutants
NIST	National Institute of Standards and Technology
NT	Not Tested - Lab testing was not done
OCMA	Ohio Cast Metals Association
ODS	Open Data Systems
PCS	Pouring, Cooling, Shakeout
PM	Particulate Matter
POM	Polycyclic Organic Matter (POM) including Naphthalene and other compounds that contain more than one benzene ring and have a boiling point greater than or equal to 100 degrees Celsius.
PPE	Personal Protective Equipment
PTE	Potential to Emit
PUCB	Phenolic Urethane Cold Box
PUNB	Phenolic Urethane No Bake
QA/QC	Quality Assurance/Quality Control
RRF	Relative Response Factor
SCPI	Standard Commands for Programmable Instruments
SERDP	Strategic Environmental Research & Development Program
SIVL	System Integration Validation Lab
TEA	Triethylamine
TGOC	Total Gaseous Organic Concentration
TGOC as Propane	Quantity of undifferentiated hydrocarbons including methane determined by EPA Method 25A.
THC	Total Hydrocarbon Concentration
TTE	Temporary Total Enclosure
US EPA	United States Environmental Protection Agency
USCAR	United States Council for Automotive Research
VOC	Volatile Organic Compound
WBS	Work Breakdown Structure