PM Emissions from Flares:

Chasms among Current Emission Factors, Fundamentai Studies, and Field Observations

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Invited presentation to
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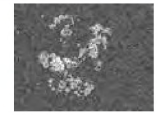
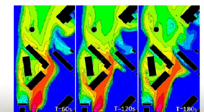
Maui, HI, Sept 30, 2010

Combustion Research
NRC · CNRC
Institute for Chemical Process and Environmental Technology

Current Research Initiatives



- Emissions from flaring
  - Soot (PM) emissions
  - Quantitative Field diagnostics for soot
  - Greenhouse gas emission models / analysis for Alberta
- Economic analysis of flaring and venting mitigation
- Fugitive emissions
  - Optical diagnostic development
  - Innovative detection schemes
- Mine face emissions from oil sands
- Emissions from liquid storage tanks
- Propagation dynamics of premixed and partially premixed flames



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Emissions from flaring & venting

- "Flare efficiency" (Carbon conversion efficiency):

$$\eta = \frac{\textit{Mass} \text{ of Carbon Converted to } CO_2}{\textit{Mass} \text{ of Carbon Originally as Fuel}}$$

- Speciated emissions:
  - Key greenhouse gases
    - CH_4, CO_2
  - Priority pollutants
    - Soot (carbon based PM), SO_2, H_2S, NOx
    - Soot has recently been implicated as a key climate force *(e.g. Ramanathan & Carmichael, 2008; IPCC AR4, 2007)*
  - Minor species
    - Volatile organic compounds (VOCs)

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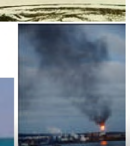
Motivation – Quantifying PM Emissions

- Particulate matter (PM) emission from flares is a global issue
  - Global gas flaring exceeds 135 billion m^3/year
  - No quantitative approaches exist to measure these emissions
  - Current "emission factor" models are flawed
- More than serious heath effects, PM / soot is a key climate forcer (*IPCC, AR4, 2007*)
  - +0.2 ± 0.15 W/m^2 for fossil fuel black carbon
  - –0.05 ± 0.05 W/m^2 for fossil fuel organic carbon
  - One prominent study suggests, climate forcing of BC could be +0.9 W/m^2 (55% that of CO_2) *(Ramanthan & Carmichael, 2008)*



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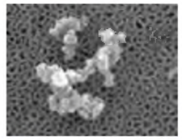
Brief Notes on Soot / PM Emissions

- Formation exceedingly complex; entails:
  - Chemical composition of fuel
  - Turbulent mixing & diffusion of air and fuel species
  - Rate of heat transfer from flame
  - Residence time / temperature history through flame
- No existing practical approaches for quantifying PM in plumes of flares
- Measuring PM in general is a challenge no matter the source



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So what do we *actually* know about soot from *flares*?

- To date, not much…
- One main set of studies on soot from flares via USEPA:
  - McDaniel (1983) – while focusing on efficiency measurements, measured plume concentrations of soot for flares with smoke suppression disabled
  - Pohl et al. (1986) – did not report direct emission rate data but concluded for his test conditions, soot accounts for "less than 0.5% of the combustion inefficiencies"
- Fundamental work on soot emissions from turbulent jet flames
  - Faeth *et al.* (1990s): studies of "overfire soot" for strongly sooting fuels
  - Becker and Liang (1982): measurements of soot from high momentum jet flames from millimeter scale burners
- Extremely limited work on small-scale reacting jets in crossflow
  - Ellzey et al., 1990; University of Alberta, 2002

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So how are soot emissions from flares currently reported?

- In Canada, PM emissions above threshold amount must be reported to the National Pollutant Release Inventory (NPRI)
  - Problematic since PM from flares is not readily measured
- The Canadian Association of Petroleum Producers (CAPP) has developed a guide for reporting
  - Simple emission factors based on volume of gas flared
    - 2.5632 kg soot per 10^3 m^3 fuel flared
  - Assumed constant under all conditions regardless of flare size, fuel composition, wind effects, flowrates, etc.
- Some obvious questions:
  - Where does this number come from? Is this approach reasonable? Is there any other alternative? What liabilities might this create?

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Introduction: Current Emission Factors

Source	Original Factor as reported	Emission Factor (kg PM per 10^3 m^3 fuel)	Gas
CAPP Guide	2.5632 kg PM per 10^3 m^3 fuel	2.5632	"Adjusted" for HHV=45MJ/m^3
USEPA FIRE v.6.25	53 lb PM per 10^6 ft^3 fuel	0.85	Landfill Gas[1]
	17 lb PM per 10^6 ft^3 fuel	0.27	Methane[2]
	0-274 lb PM per 10^6 BTU	0 - 5301	80% Propylene, 20% Propane
USEPA AP-42 Vol. I, sect. 13.5	0-274 μg PM per 10^{-3} m^3 exhaust gas[3]	Not Convertible	80% Propylene, 20% Propane

1. Typ. landfill gas composition: 56% CH_4, 37% CO_2, 1% O_2, and trace amounts of other gases
2. Predominantly enclosed flare measurements at landfill sites
3. The range of 0–274 is based on the "smoking level": non-smoking flares, 0 μg/L; lightly smoking flares, 40 μg/L; average smoking flares, 177 μg/L; and heavily smoking flares, 274 μg/L

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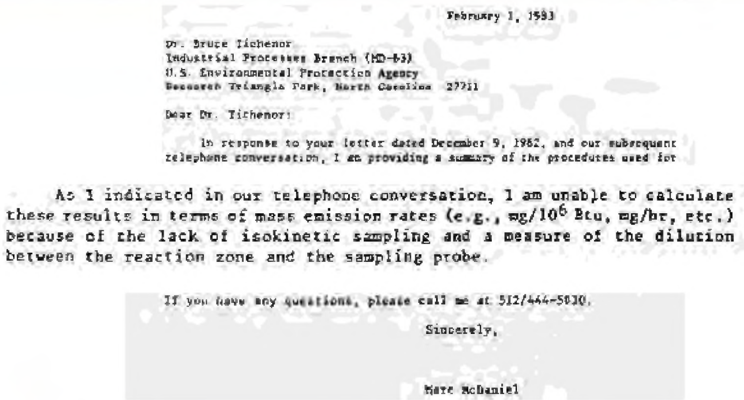
Some Forensics...


February 1, 1983

Dr. Bruce Tichenor
Industrial Processes Branch (MD-63)
U.S. Environmental Protection Agency
Research Triangle Park, North Carolina 27711

Dear Dr. Tichenor:

In response to your letter dated December 9, 1982, and our subsequent telephone conversation, I am providing a summary of the procedures used for

As I indicated in our telephone conversation, I am unable to calculate these results in terms of mass emission rates (e.g., mg/10^6 Btu, mg/hr, etc.) because of the lack of isokinetic sampling and a measure of the dilution between the reaction zone and the sampling probe.

If you have any questions, please call me at 512/444-5030.

Sincerely,

Marc McDaniel

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Current Status of Emission Factors

- Error in US EPA emission factor data base has now been formally reported to the correct authorities who are reviewing the appropriate documents
- Even with correction, there are still only three main sources of emission factor data:
  - Two are based on landfill gas flares (and are likely enclosed flares)
  - One is based on a propylene/propane flare and is reported in original form as a plume concentration only
- Single emission factor approach is also overly simplified
- One motivation of current work is to seek better, measurement-based emission factors for PM in flares

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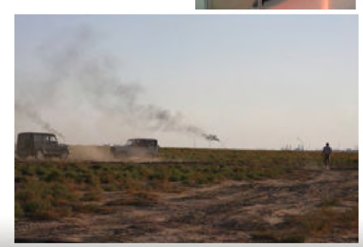
Soot / PM: Current Research Initiatives

Two main areas of focus:

1. Direct measurement of soot emissions from flares in controlled lab setting
   - Sampling protocol development
   - Elemental / Organic Carbon measurements
   - Emission factors development
2. Novel diagnostic to measure soot from flares in the field
   - Desire simple tool to improve upon qualitative "opacity"
   - Related work on measuring optical properties of soot aggregates



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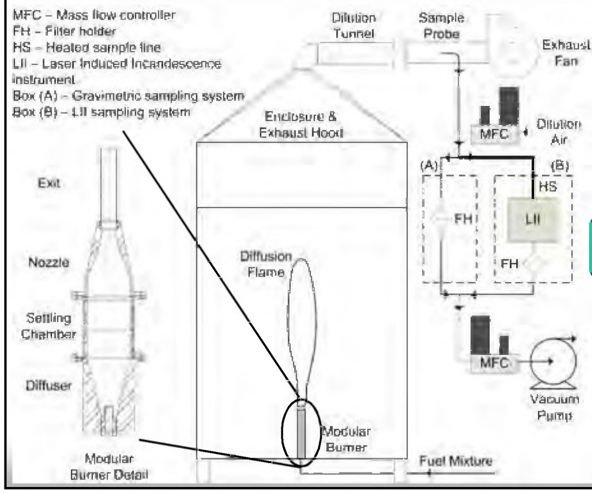
Experimental: Burner & Enclosure



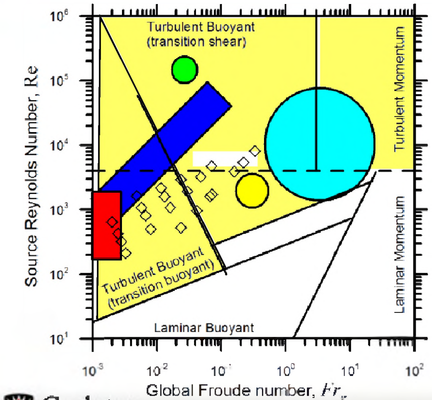
Sampling Protocol Developed based on Diesel sampling

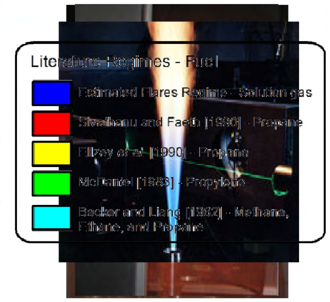
Soot Yield

$$Y_s = \frac{\rho_{soot} f_{v,sample} Q_{DT} T_{sample}}{\dot{m}_{fuel} T_{DT}}$$

Introduction: Flame & Flare "Regimes"

- Regime map of Delichatsios [1993]





$$Fr_g = \frac{u_e f_s^{3/2}}{(g d_e)^{1/2}} \left(\frac{\rho_\infty}{\rho_e} \right)^{1/4}$$

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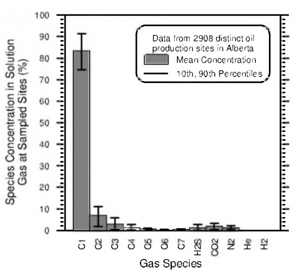
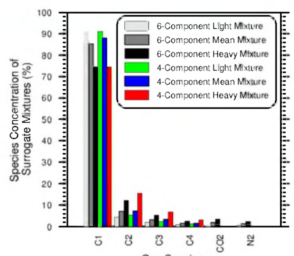
Experimental: Fuel Mixtures

- Based on 2007 PTAC data from 2908 production sites





Fuel Mixture	Smoke-point (mg/s)
Average Raw Mixture	31.2
AVG-6-component	31.8
AVG-4-component	32.8

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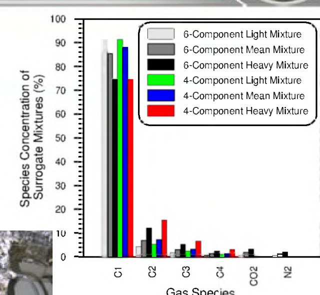
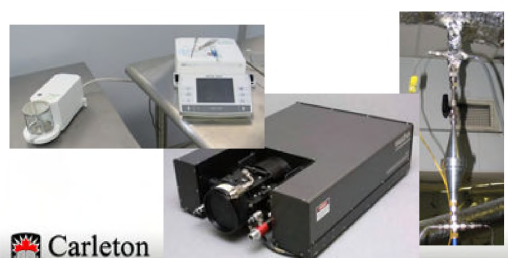
Lab-based Experiments

- Controlled experiments using 3 main techniques:
  - Laser induced incandescence
  - Gravimetric sampling
  - EC/OC (NIOSH 5040)





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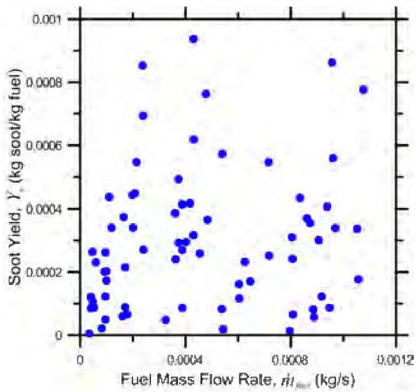
Soot Emissions from Lab Flares

Data from ongoing experiments

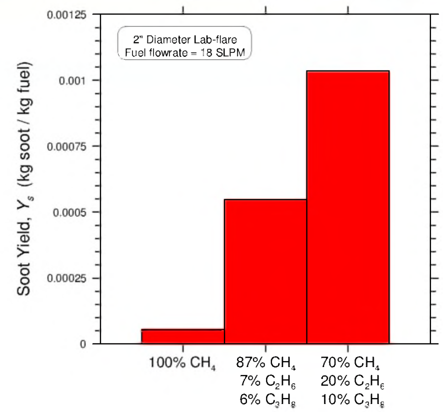
- Not surprisingly, soot emission rate varies with flare conditions
  - Fuel composition
  - Flare diameter
  - Flowrate



Soot Measurement Results

Fuel Composition

- Order of magnitude change in soot yield for very small changes in fuel composition
- Composition is critical for meaningful soot factors

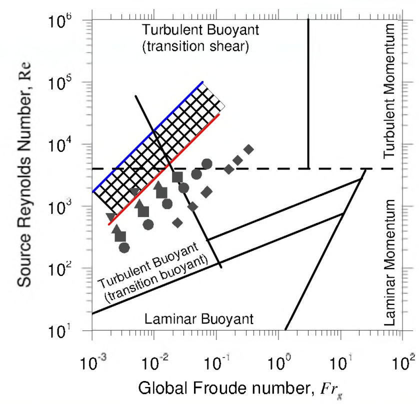


Results: Scaling Aerodynamics

- Several parameters investigated in an attempt to scale aerodynamic affects
  - Flame length Richardson ratio [Becker and Liang, 1982]
  - Global characteristic residence time [Becker and Liang, 1982]
  - Buoyant residence time [Canteenwalla, 2007]
  - Simple residence time [based on Sivathanu and Faeth, 1990]
  - Fire Froude number [Delichatsios, 1993]

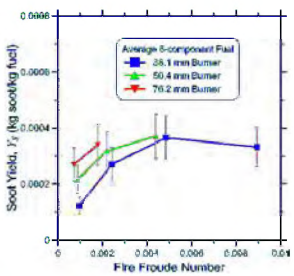
Results: Scaling Aerodynamics

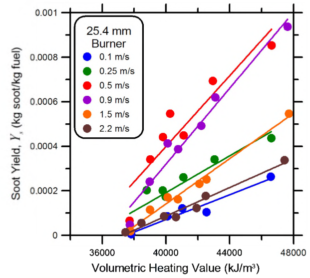
- Suggested by Delichatsios [1993], the "global fire Froude number", Fr_g, appears to show most promise for developing regime based correlations



"Engineering Attempt" EF model

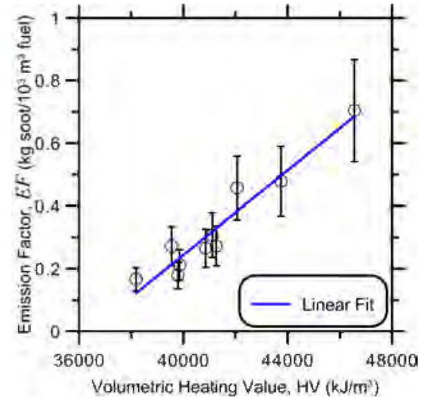
- "Assume" constant soot yield at $Fr_f > 0.003$
- For narrow range of anticipated fuel mixtures, soot yield scales linearly with heating value





"Engineering Attempt" EF model

- In the absence of anything better, can form a simple linear EF model based on heating value
- The chasm remains however, in linking a useful model with fundamental understanding

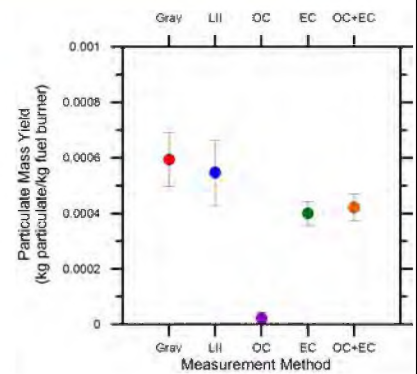


PM from flares: Preliminary Conclusions

- Results of current work are showing (as expected) that soot emission rate is strongly affected by:
  - Fuel composition, flare diameter, and flare gas flowrate
- Current emission factors are oversimplified and are of questionably related origin
- Best engineering guess based on current data suggests CAPP emission factor for flare generated soot is likely a factor of 2 or more too high
  - However, we are a long way from having a defensible model based on fundamental understanding of the soot formation process in a flare

Preliminary Results: EC/OC

- Measurements of elemental carbon content in flare generated soot
- Preliminary results suggest that EC is ~95% of total
  - Good agreement with laser induced incandescence data



Soot / PM: Current Research Initiatives

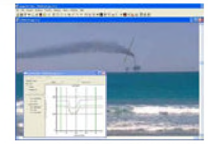
Two main areas of focus:

1. Direct measurement of soot emissions from flares in controlled lab setting
   - Sampling protocol development
   - Elemental / Organic Carbon measurements
   - Emission factors development

2. Novel diagnostic to measure soot from flares in the field
   - Desire simple tool to improve upon qualitative "opacity"
   - Related work on measuring optical properties of soot aggregates

Development of a Novel Soot Plume Field Diagnostic

- Current standards are based on opacity, which is only qualitatively related to soot emission rate
  - E.g. EPA test method 9, a 'human observed' standard
- Is there any way to make quantitative measurements in the field?
  - Would be invaluable not only for corroborating lab-based data but also as a field tool
- Ongoing investigation of a novel concept that is now showing considerable promise…



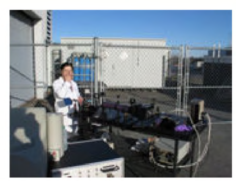
Novel soot diagnostic: principle

- Idea: Can we use sky-light to make a quantitative, open-ended, optical measurement of soot in a plume?
- Mathematical basis:

$$\dot{m}_{soot} = \frac{-u\rho_{soot}\lambda}{6\pi E(m)_{\lambda}(1+\rho_{sa})}\int \ln(\tau_{\lambda}(y))dy$$

- If we can develop a quantitative system to measure transmissivity, we can make field measurements of soot plumes
  - Need optical properties of soot (*paper submitted to Appl. Phys. B*)
  - Need good estimate of plume velocity (see subsequent slides)

New Field Diagnostic for Soot Plumes



- Novel camera based technique under development to directly measure strongly sooting flares under field conditions
- Lab-based development:
  - *Thomson et al., Applied Optics, 2008*
  - *Johnson et al. (1), Environmental Science & Technology, Accepted Sept. 23, 2010.*

- Initial field trial completed to measure emissions from a large sooting flare in Uzbekistan
  - *First ever, quantitative field measurement of soot from a flare*
  - *Johnson et al. (2), currently under review with Env. Sci. Technol. as of Sept. 2010.*

Methodology for Field testing Sky-LOSA

- Sky-LOSA (plume transmissivity) acquisitions
  - 16 bit Peltier-cooled CCD camera with 532nm filter
  - Commercial lens
  - Laptop control via custom written acquisition software

- High-speed images
  - Casio EX-F1 digital camera
  - No collection filter: acquisition in the visible
  - 300 frames per second @ 512 x 384 pixel resolution
- Soot flux calculated as: $\dot{m}_{soot} = \frac{1}{N_f}\frac{1}{N_z}\sum_{j=1}^{N_z}\sum_{i=1}^{N_f} A\int U(y,z)\ln(\tau_\lambda(y,z))dy$

Field testing of new soot diagnostic

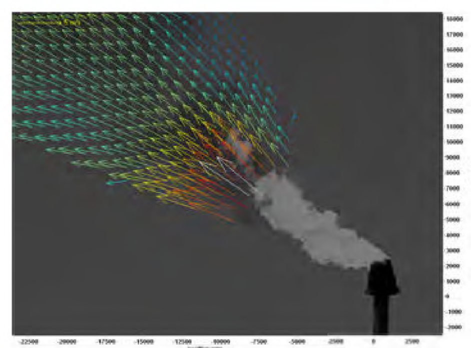
- Field data collected in Uzbekistan, July 2008, as part of separate World Bank funded project to estimate flare volumes in collaboration with Dave Picard (Clearstone Eng.)

First test of new Sky-LOSA diagnostic

- Field measurement of large (1.04 m diameter) flare at a gas plant in Karshi, Uzbekistan
- Flare gas composition and flow rate data not available

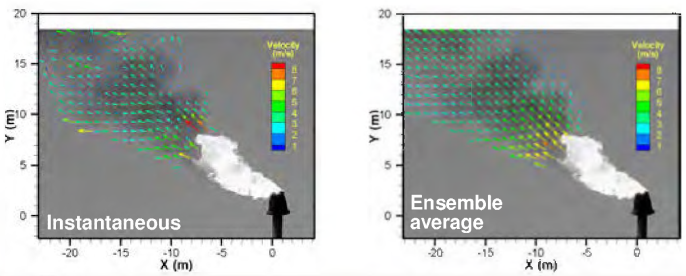
Quantification of Plume Velocity

- "Image correlation velocimetry" to quantitatively measure plume velocity from high-speed video of visible soot plume

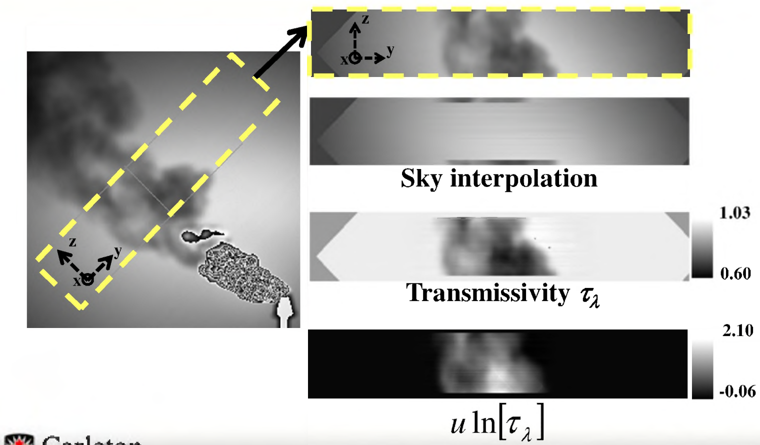


Quantification of Plume Velocity

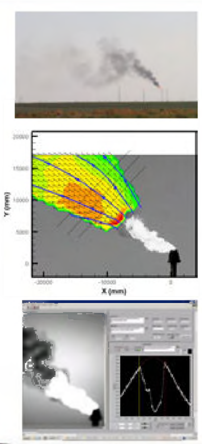
- Velocity measurements presented based on available 61s of high-speed video only (18,300 frames)
  - Uncertainties estimated at 25%, although this could be reduced significantly with better optimization
  - Short time span of velocity measurements also impacts accuracy



Sky-LOSA processing



2. Field Diagnostic for Soot Plumes



- Preliminary results of first tests
  - Results for one (1) flare only and should not be generalized
  - Analysis currently under review in article submitted to Env. Sci. Technol.
- Soot flux quantified at ##-## g/s
  - Uncertainty currently estimated to be +/-33%
  - Approximately equivalent to ##-## buses running continuously *(assuming 299 mg PM2.5/km [Keogh et al., Env. Sci. Poll. Res, 2010])*

Conclusions

- Current emission factors oversimplified and of questionable accuracy
- Best engineering estimate from current data suggests CAPP factor for flare generated soot is likely >2x too high
  - Because of regime transitions, experiments (and possibly models) at a variety of scales will be required to develop a defensible emission factors
  - Some empiricism likely unavoidable -- fundamental understanding of soot formation in turbulent flames remains as the most challenging problem in combustion
- New "Sky-LOSA" field measurement technique continues to show promise and is ready for more field testing

Research Partners

Project Manager: Michael Layer, *Natural Resources Canada*

Natural Resources Canada

CAPP Canadian Association of Petroleum Producers

National Research Council Canada NRC-CNRC

NSERC CRSNG

Environment Canada

Carleton University Canada's Capital University

Current Research Team

- Principle Investigators:
  - Matthew Johnson, *Canada Research Chair in Energy & Combustion Generated Pollutant Emissions, Associate Prof., Carleton University*
  - Kevin Thomson, *Research Officer, NRC-ICPET*
- Post. Doctoral Fellows / Research Engineers:
  - Adam Coderre, *M.A.Sc.* Robin Devillers, *Ph.D.*
  - James McEwen, *M.A.Sc.*
- Graduate Students:
  - Brian Crosland, *Ph.D. candidate* Jan Gorski, *M.A.Sc. candidate*
  - Ian Joynes, *M.A.Sc. Candidate* Clifton Pereira, *M.A.Sc. candidate*
  - Stephen Schoonbaert, *M.A.Sc. cand.* Patrizio Vena, *Ph.D. candidate*
- Recent Alumni:
  - Carol Brereton, *M.A.Sc.* Chen Yang, *M.A.Sc.*
  - Pervez Canteenwalla, *M.A.Sc.*