ADVANCES IN UNDERSTANDING THE HEALTH HAZARDS OF VEHICLE EMISSIONS

Topics:

- Toxicity of emissions from alternate diesel fuels
- Toxic components of vehicle emissions
- Potential Benefits of clean diesel technologies
- Comparative effect of engine emissions and other complex source emissions

Joe Mauderly (et many al.)

NATIONAL ENVIRONMENTAL RESPIRATORY CENTER

Because You Never Breathe Only One Pollutant !

Lovelace Respiratory Research Institute
Albuquerque, NM

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THERE IS SOLID EVIDENCE THAT AIR POLLUTANTS DERIVED IN PART FROM MOBILE SOURCE EMISSIONS EXERT A PUBLIC HEALTH BURDEN

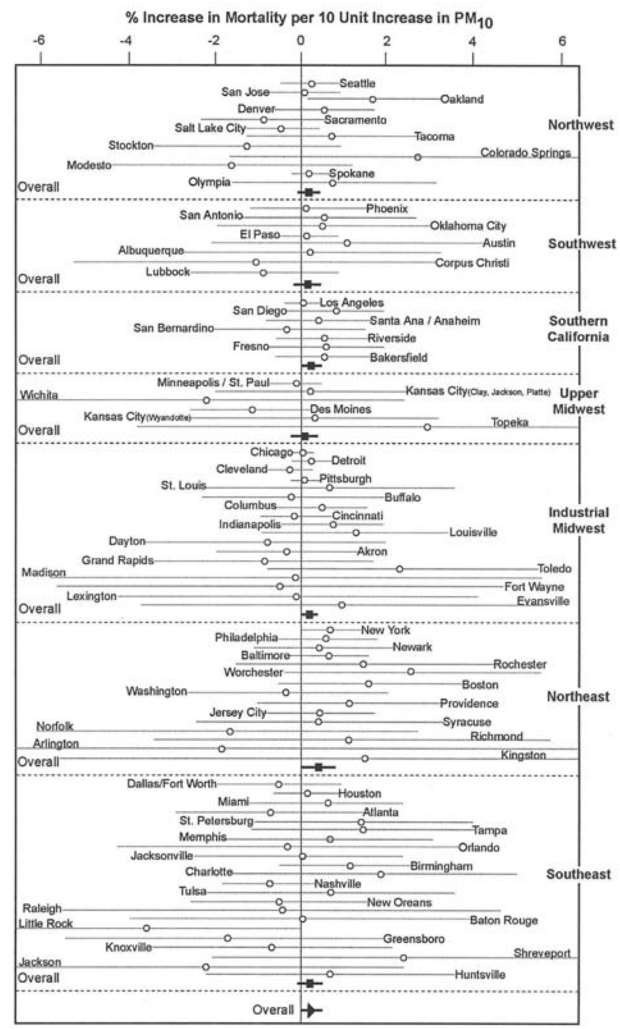
Example: particulate matter (PM) is associated with increased daily mortality and morbidity

Studies examining different PM components have shown the significance of mobile sources

Laden et al. 2000 and Schwartz et al., 2003, mortality in 6 cities study

Mar et al. 2000, 2003, cardiovascular mortality in Phoenix

Tsai et al. 2000, mortality in New Jersey



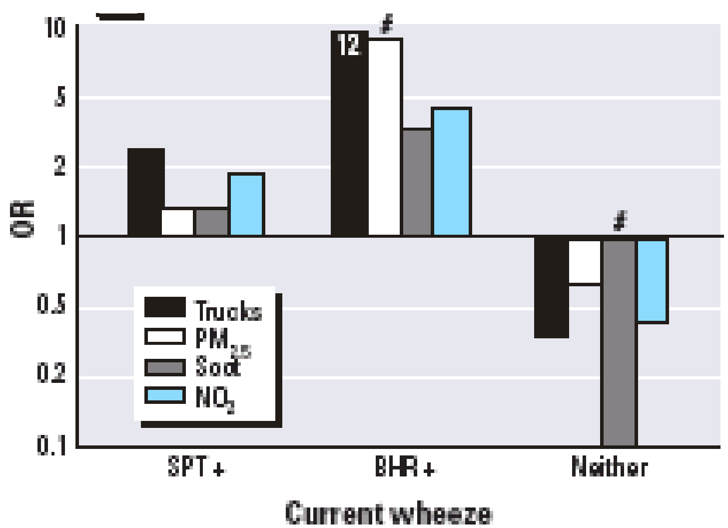
NMMAPS Sudy, Dominici et al., 2002

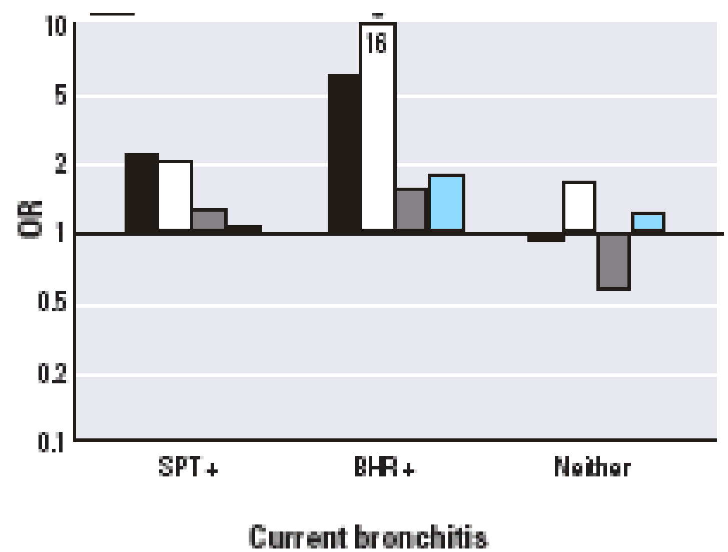
EVIDENCE IS STILL GROWING FOR RELATIONSHIPS BETWEEN RESPIRATORY SYMPTOMS IN CHILDREN AND PROXIMITY TO HEAVY TRAFFIC

Example: Janssen et al., *Environ. Health Perspect.* 111: 1512-1518, 2003

- **Children in Netherlands attending school within 400 m of highways**
- **Traffic counts and air pollution measurements at schools**
- **Allergies by skin prick tests (SPT)**
- **Bronchial hyperreactivity (BHR) by hypertonic aerosol challenge**
- **Health questionnaires**

Results: The odd ratios for wheeze, bronchitis, and other symptoms <u>in allergic children</u> were related to PM, truck traffic, and NO_2





EMISSIONS FROM ALTERNATE DIESEL FUELS

EPA test requirements for new fuels and additives

- **Clean Air Act, Title II, Section 211b, 40 CFR 79(E), Tier 2 toxicity**
- **Expose rats 6 hr/day, 5 days/wk for 13 wks (+ 28 day recovery group)**
  - **Emissions from "typical" engine on variable-load cycle**
  - **3 levels + clean air control (highest level to cause effects)**
- **Health measurements:**
  - **Body weight, food consumption, clinical signs**
  - **Histopathology**
  - **Hematology & Serum chemistry**
  - **Fertility & reproductive toxicity**
  - **Ophthalmology**
  - **Neurotoxicity**
  - **Chromosome injury (micronucleus & sister chromatid exchange)**
  - **Bacterial mutagenicity (Ames Salmonella)**

Comparisons are handicapped by the lack of a Tier 2 study of conventional petroleum diesel

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THREE TIER 2 STUDIES DONE TO DATE

"Biodiesel"

100% soy methyl ester (Soygold® Soydiesel, AG Environmental Products)

Diesel-Water Emulsions

1. CARB petroleum diesel + water (Purinox® Summer blend, Lubrizol)
2. CARB petroleum diesel + water + methanol (Purinox® Winter blend, Lubrizol)

Exposures (PM = µg/m³, NOx = ppm)

	Biodiesel		Purinox Summer		Purinox Winter	
	PM	NOx	PM	NOx	PM	NOx
maximum >	500	51	401	20	500	37
	200	26	203	12	250	22
	40	5	103	7	125	14
controls >	2	1	6	<1	6	<1

[Finch et al., *Inhal. Toxicol*. 14: 101, 2002]
[Reed et al., *Toxicologist* 78:1385, 2004]
[Reed et a., *Inhal. Toxicol*. submitted, 2004]

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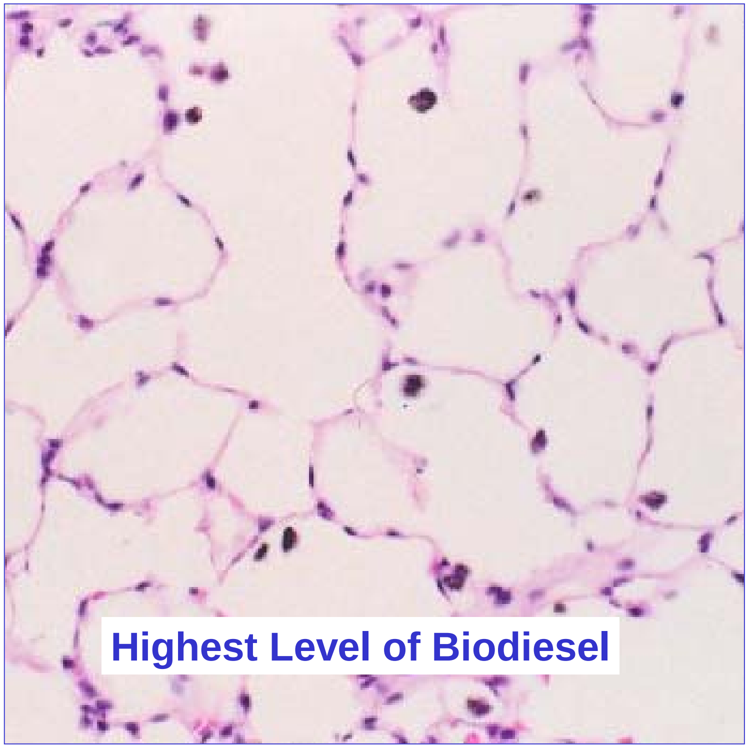
SIMILAR RESULTS FOR ALL THREE FUELS

Significant dose-related effects on:

- Macrophages with PM in lung (reduced during recovery)
- Decreased serum cholesterol
- Misc. other serum effects, but no corresponding histopathology

No effects at any level on:

- Mortality, clinical signs, or body weight
- Fertility or reproductive toxicity
- Ophthalmology
- Neurotoxicity
- Chromosome injury



The middle exposure level was considered a "no significant adverse effects" level for all 3 fuels

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TOXIC COMPONENTS OF VEHICLE EMISSIONS

Which physical-chemical components drive the health effects ?

Many studies demonstrated effects of "whole" emissions

Several have shown that:

- Individual components can have effects
- PM is not the sole culprit

None have methodically compared different engine & fuel types

We can't just focus on one component

Example: 2000 model Cummins 5.9L turbo diesel operated on heavy-duty certification cycle and burning national average fuel

At a dilution yielding a PM mass concentration of 1,000 µg/m^3:

NOx (48 ppm) = 62,400 µg/m^3

Vapor-phase semi-volatile organic compounds = 2,330 µg/m^3

SO_2 (0.35 ppm) = 950 µg/m^3

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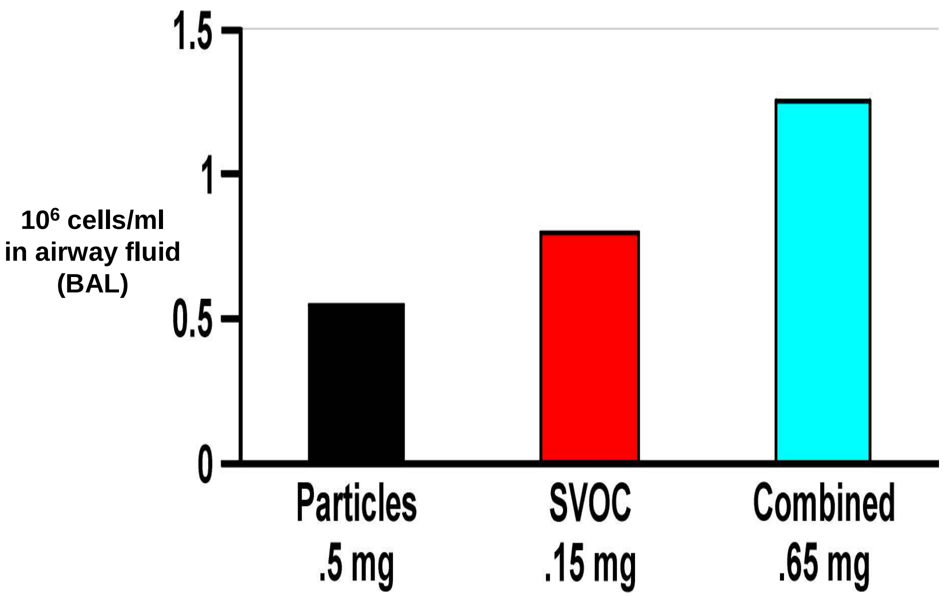
VAPOR-PHASE SEMI-VOLATILE ORGANIC COMPOUNDS ARE AN MPORTANT CLASS OF EMISSIONS

Example: Semi-volatile organic compounds in Traffic tunnel samples

1. PM and vapor-phase SVOC collected from traffic tunnel and instilled into rat lungs
2. Measured Inflammatory cells in airway fluid 24 hr later



SVOC caused most of the effect

(4x more toxic per unit of mass than PM)

[Seagrave et al., *Toxicologist* 60:192, 2001]

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COMPOSITION vs. TOXICITY OF ENGINE EMISSION SAMPLES

1. In-use vehicles tested as received
2. PM and vapor-phase SVOCs collected on urban driving cycle
3. Detailed chemical analysis
4. Re-combined the 2 fractions in original ratio

   Instilled into lungs of rats, evaluated at 24 hours

   Measured mutagenicity in bacteria (Ames assay)

Samples	
Gasoline (5)	G
Gasoline 30°	G_{30}
White smoker gas.	WG
Black smoker gas.	BG
Diesel (3)	D
Diesel 30°	D_{30}
High-emitter diesel	HD

Lung Response Variables

Lung Lavage

- Cell counts
- LDH
- Protein
- Cytokines

Lung Histopathology

[Seagrave et al. *Toxicol. Sci.* 70: 212-226, 2002]

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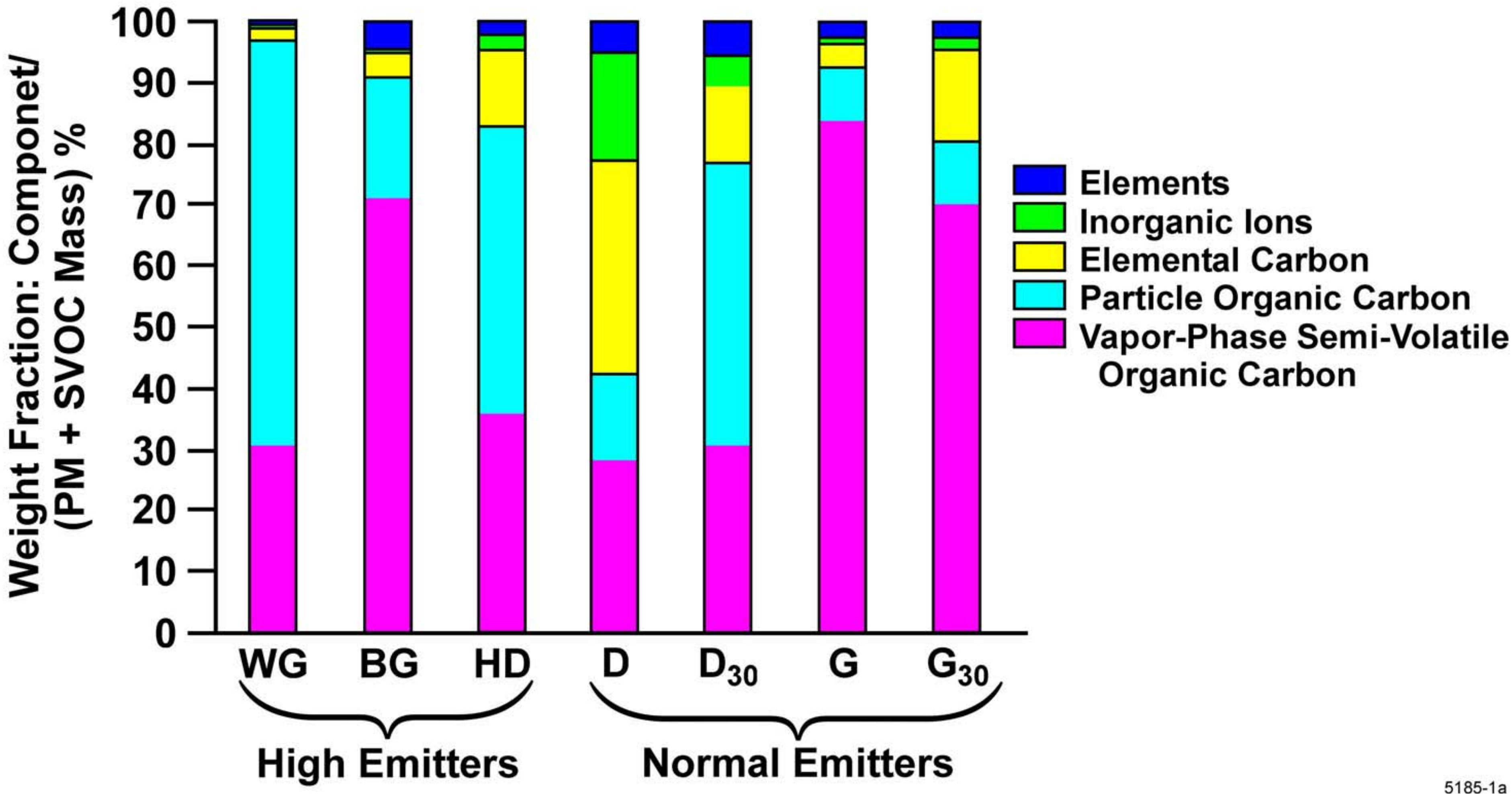
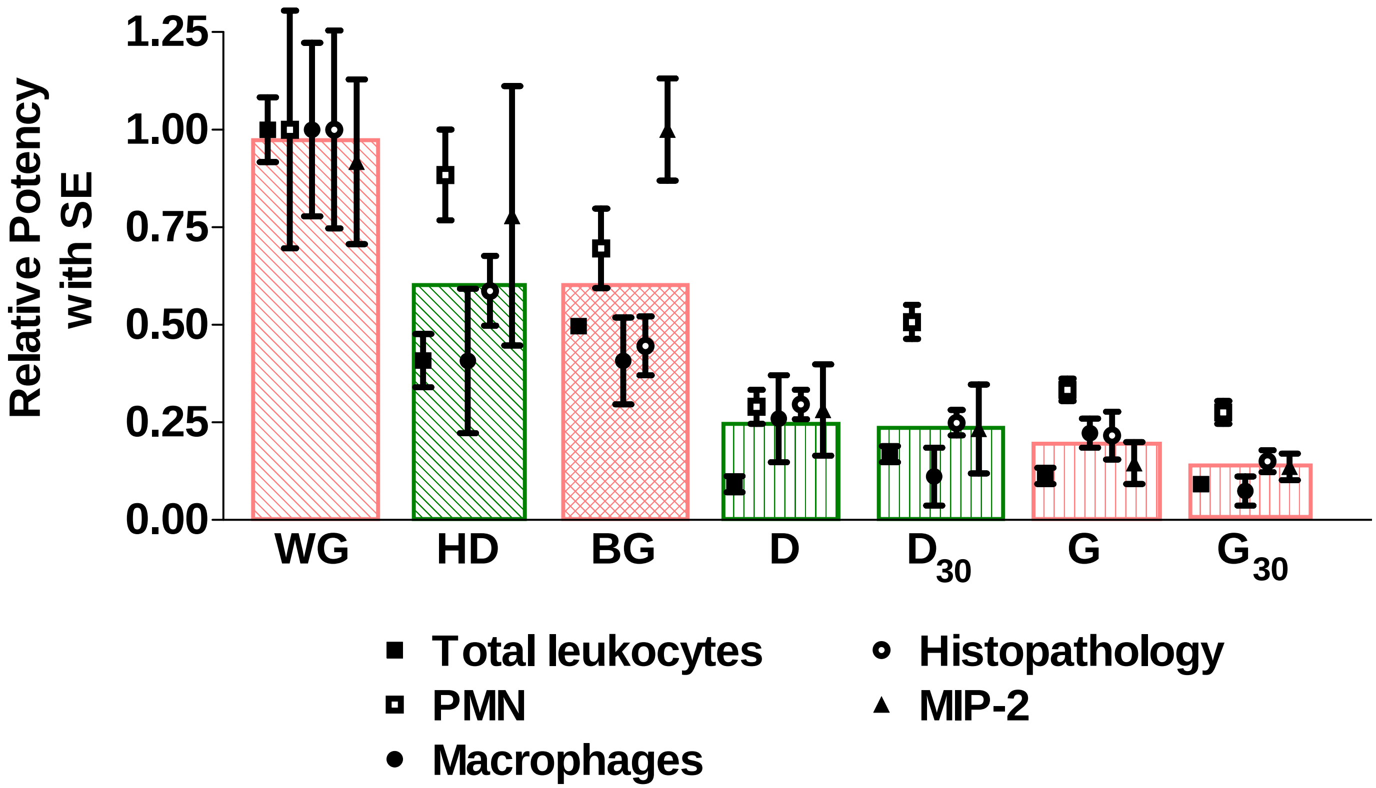
SAMPLE COMPOSITION BY MASS FRACTION



[Zielinska et al., *J. Air Waste Man. Assoc.* 54: 1138-1150, 2004]

THE SAMPLES HAD A 5-FOLD RANGE OF LUNG TOXICITY PER UNIT OF MASS

Example: Lung inflammation at equal doses

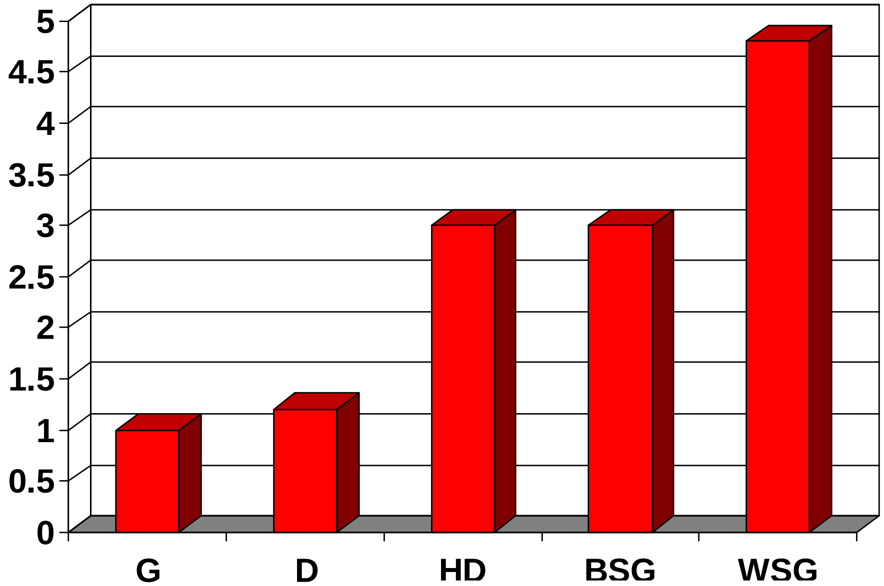


1. **High-emitters produced more toxic emissions**
2. **Normal-emitter diesel and gasoline had similar toxicity**

[Seagrave et al. *Toxicol. Sci.* 70: 212-226, 2002]

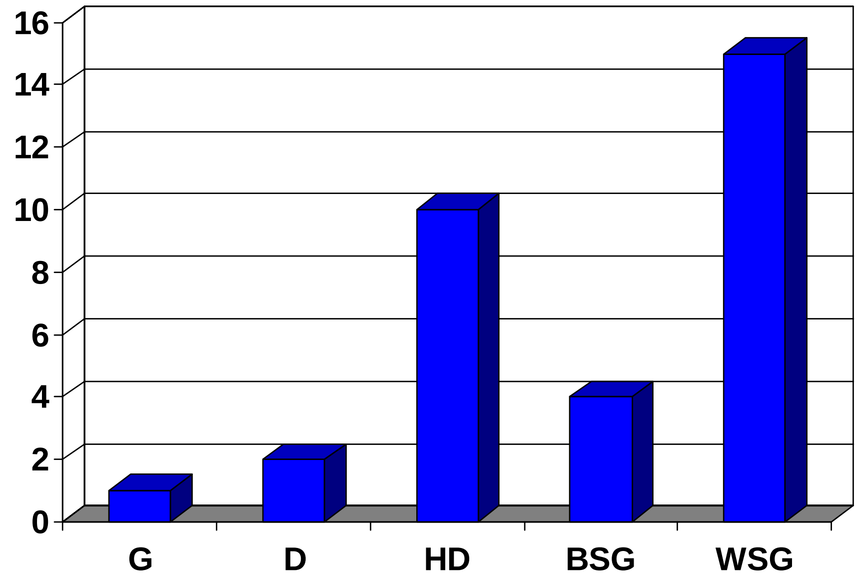
RESULTS REINFORCE IMPORTANCE OF HIGH-EMITTERS

RELATIVE INFLAMMATORY EFFECT PER UNIT OF MASS

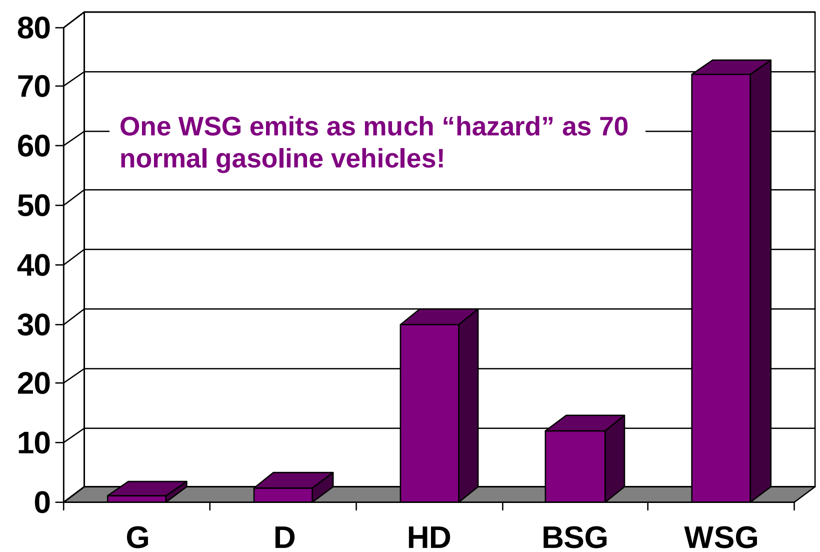


"Normal gasoline" set to 1.0

RELATIVE EMISSION RATES



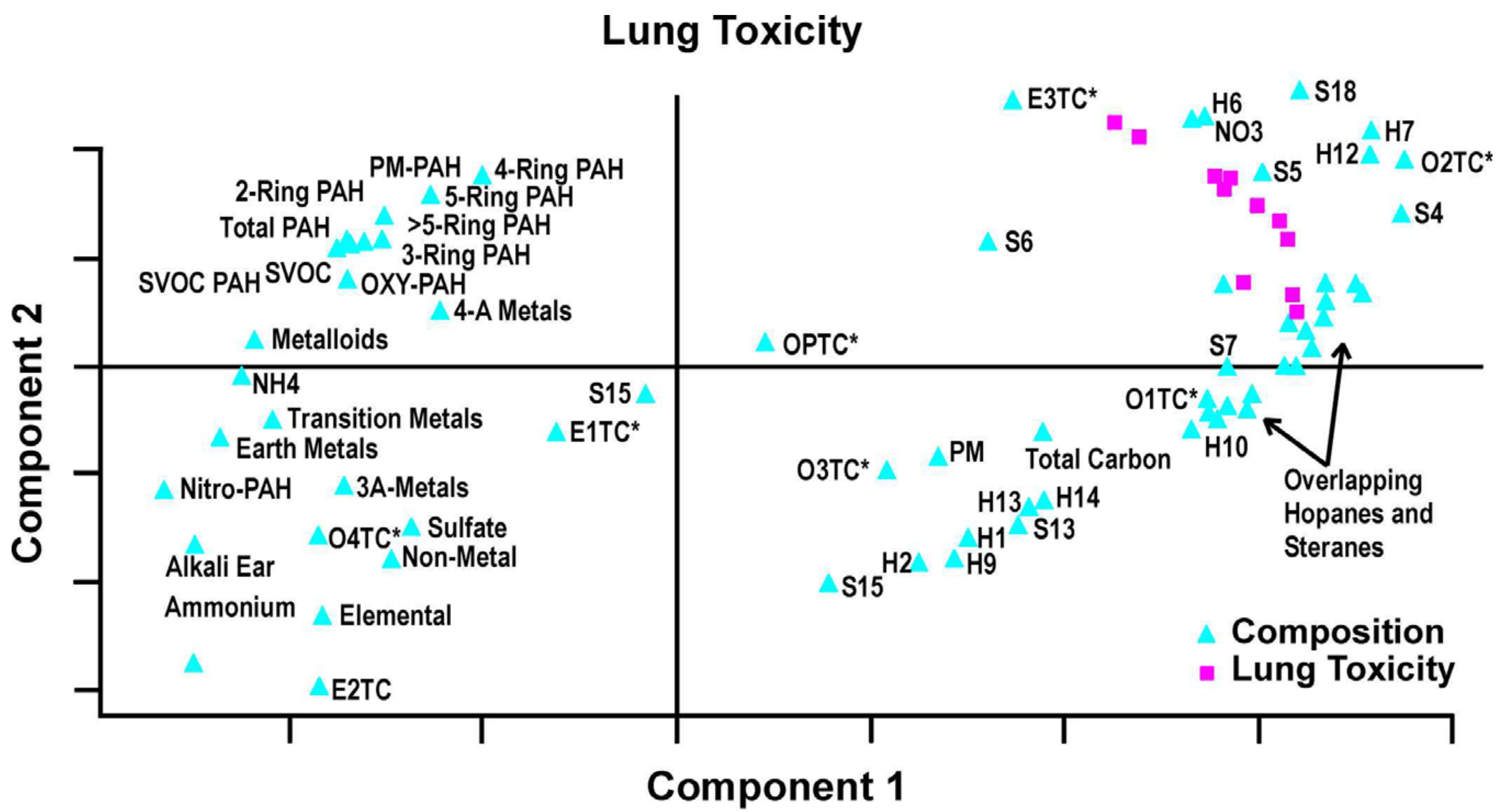
"RELATIVE INFLAMMATORY HAZARD" PER UNIT OF VEHICLE TRAVEL



What was it about emissions from high-emitters that made them so toxic ?

PCA/PLS REVEALED WHICH PHYSICAL-CHEMICAL SAMPLE COMPONENTS CO-VARIED WITH TOXICITY

PCA = Principal Component Analysis
PLS = Partial Least Squares Regression



[McDonald et al., *Env. Health Perspect. 112: 1527-1538, 2004*]

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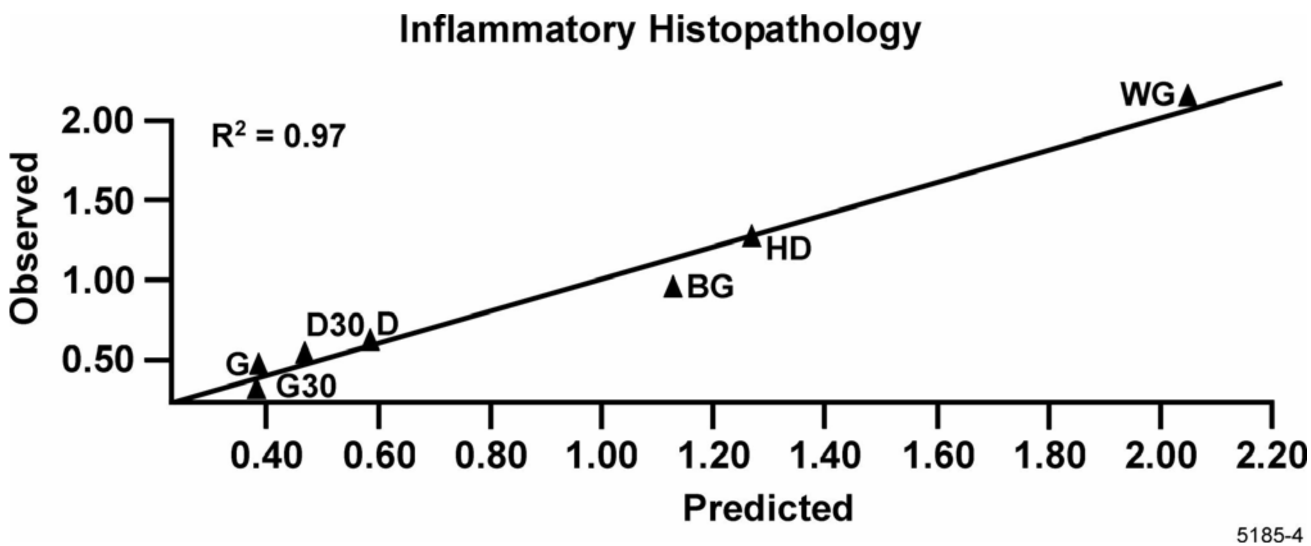
PCA/PLS PRODUCED GOOD MODELS FOR PREDICTING DIFFERENCES IN TOXICITY FROM COMPOSITION



PM-associated organic carbon, particularly <u>hopanes and stearanes</u> (markers of crankcase oil), co-varied most closely with toxicity in this set of samples

The finding that bacterial mutagenicity was related to certain, but not total, nitro-aromatic compounds validated the approach against years of chemical fractionation

[McDonald et al., *Env. Health Perspect. 112: 1527-1538, 2004*]

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DO DIESEL EMISSION CONTROL TECHNOLOGIES YIELD HEALTH BENEFITS ?

Pilot study produced first look at impact on lung health hazards:

1. **Single-cylinder 490cc Yanmar YDG5500 5500-watt generator**

   Steady-state at 100% load

2. **Compared 2 emission cases at <u>same load and dilution</u>:**

   1) **Uncontrolled – certification fuel, no after-treatment (“DEE”)**
   2) **Low-sulfur fuel and catalyzed PM trap (“DEE+ER”)**

3. **Exposed mice 6 hr/day x 7 days**

4. **Instilled Respiratory Syncytial Virus and examined at 4 days:**

   Lung retention of RSV

   Lung histopathology

   Chemical mediators of inflammation in lung lavage

   Heme-oxygenase (an index of oxidative stress)

[McDonald et al., *Environ. Health Perspect*. 112: 1307-1312, 2004]

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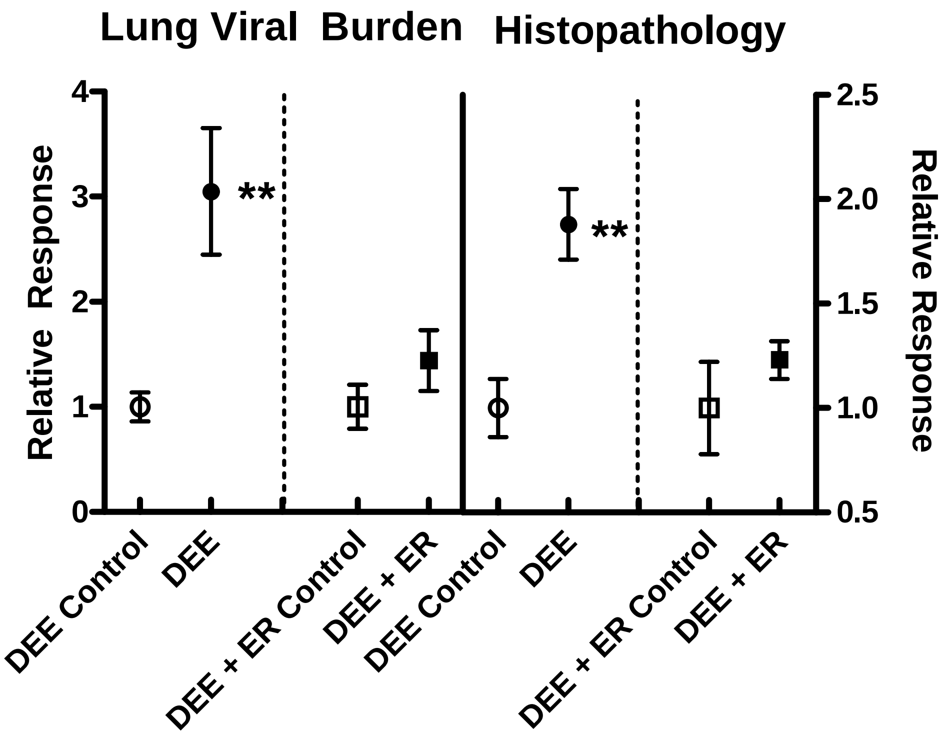
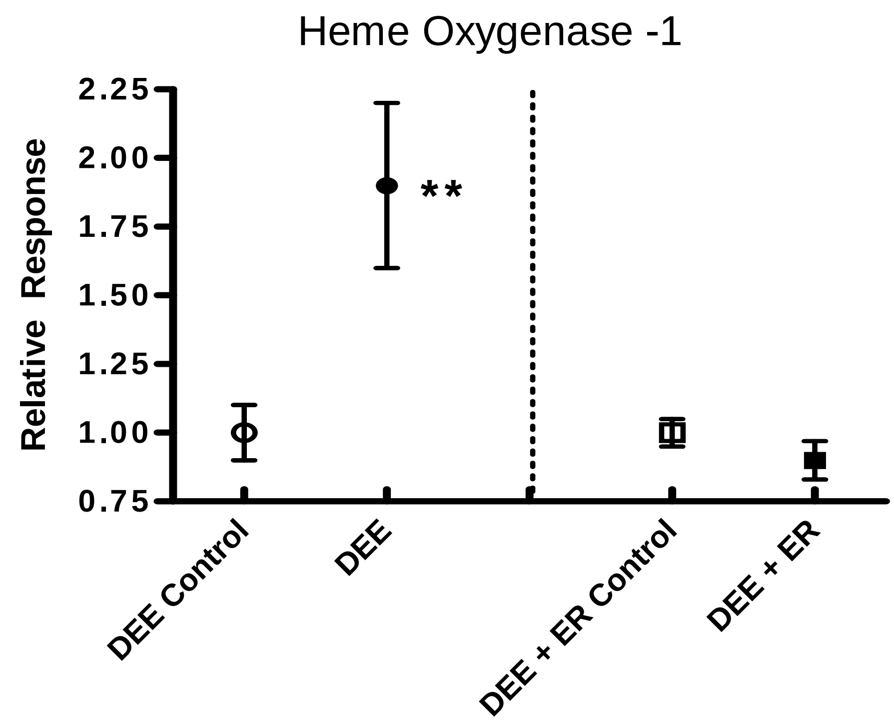
THE RESULTS DEMONSTRATED A STRIKING REDUCTION OF HEALTH EFFECTS





- **Markedly reduced emissions (e.g., PM near background)**
- **Eliminated significant health effects !**
- **Suggests great benefits from both future OEM and retrofits**

[McDonald et al., *Environ. Health Perspect*. 112: 1307-1312, 2004]

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HOW DO THE HAZARDS FROM VEHICLE EMISSIONS COMPARE TO THOSE FROM OTHER SOURCES OF AIR POLLUTION ?

The "Multi-pollutant" Dilemma

Which air contaminants are causally related to which health effects, and which are the most toxic ?

Which sources?

Which combinations?

When can we lump pollutants for regulatory purposes ?

What plausible changes in air quality would yield the greatest, most cost-effective, health benefit ?

We tend to regulate ⇒ debate ⇒ study pollutants and sources one-at-a-time

This strategy has done considerable good, but has not provided a good ability to deal with mixtures of pollutants

NATIONAL ENVIRONMENTAL RESPIRATORY CENTER

NERC: A joint government-industry program to determine the contributions of different air contaminants and their sources to the respiratory and cardiovascular health impacts of complex air pollution mixtures

Strategy: Create and analyze a composition-dose-response data matrix developed by identical laboratory studies of different source-based complex atmospheres

Involve stakeholders in planning and support of research

Vest approval authority in independent advisory group

Because You Never Breathe Only One Pollutant !

[www.nercenter.org]

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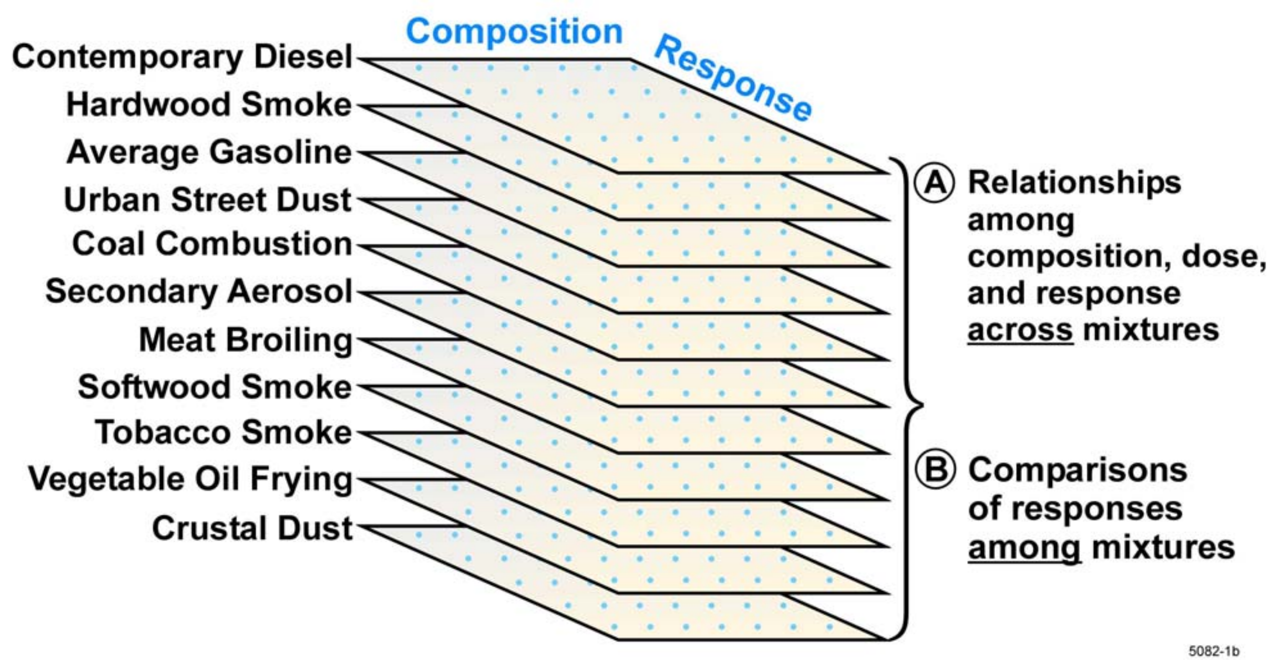
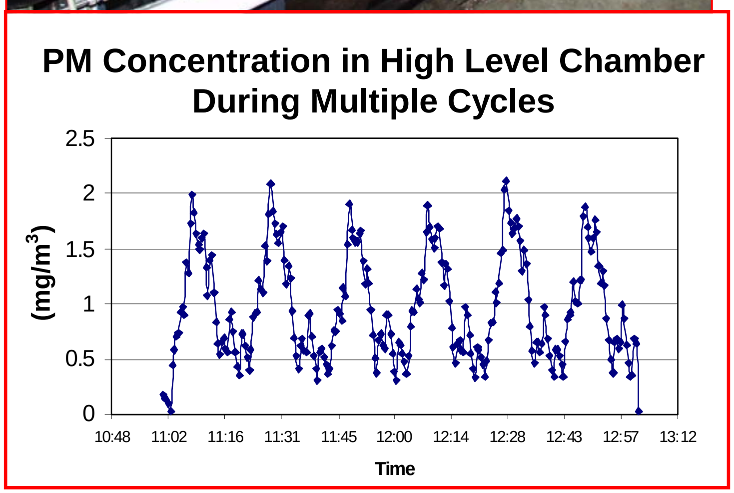
BUILDING A DATABASE FOR MULTIVARIATE ANALYSES OF COMPOSITION- RESPONSE RELATIONSHIPS



- Dose-response inhalation studies
- Expose 7 d/wk up to 6 mo
- Characterize exposure at highest practical level of detail (>500 analytes)
- Measure health outcomes in 5 general categories (>200 parameters)

[www.nercenter.org]

"CONTEMPORARY" DIESEL EMISSIONS
(Now a "Baseline" for "Clean Diesel")



<u>2000</u> Cummins 5.9L ISB 6 cyl. Turbo

D-2 Cert. Fuel (370 ppm S, 29% aromatics)

Shell Rotella-T® 15W-40 crankcase oil

Stock exhaust system with muffler

Repeated heavy-duty certification cycle

Cold start excluded

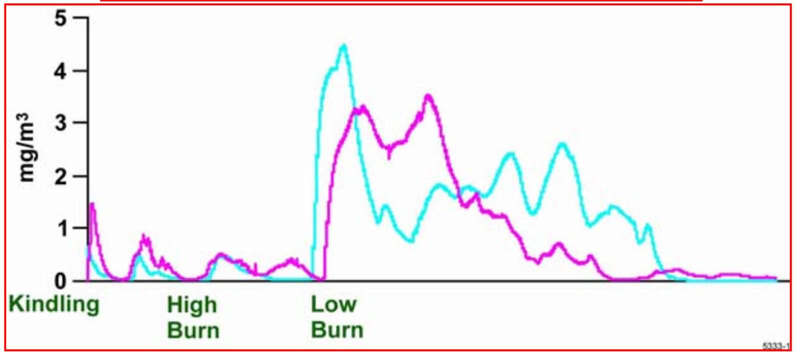
Emissions diluted with carbon- and HEPA-filtered air

Expose at 1000, 300, 100, 30, 0 µg PM/m^3
(dilutions ≈ 1:10 to 1:300)

Exposures are completed and most results are published

[Reed et al., *Inhal. Toxicol*. 16:177, 2004]

HARDWOOD SMOKE



Uncertified heating stove (Pineridge, 2 ft^2)

Scaled room air conditioned to absorb heat load

Oak from Missouri at 20% moisture

15 ft (4.6 m) stack from floor of stove

Constant draft conditions at top of stack

3-phase burn cycle

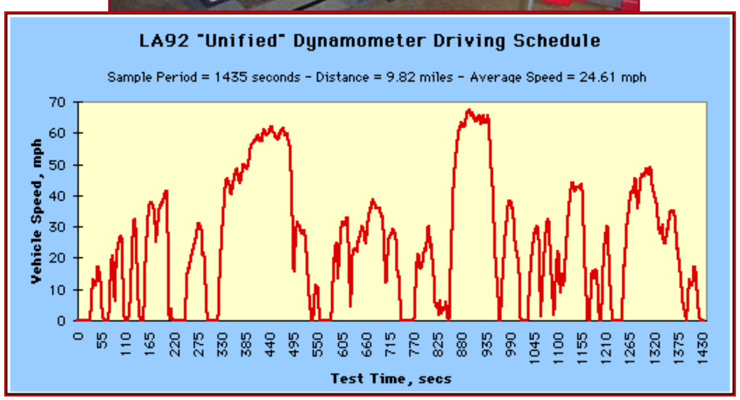
- Kindle with same wood and blank newsprint
- High burn, open air intake
- Low burn, nearly closed air intake

Extracted smoke 0.3 m from top of stack and diluted with conditioned air

Exposed at same PM mass concentration as diesel - 1000, 300, 100, 30, 0 μg/m^3
(Dilutions ≈ 1:300 to 1:9000)

Exposure are completed and data are being analyzed

GASOLINE EMISSION EXPOSURES



1996 4.3 L General Motors V-6 engines

- 3 in-use Chevrolet S-10 pickup trucks
- Mid-range mileage (40-70k miles)
- Normal emissions

(California) Unified Driving Cycle

- 3-phase cycle mapped from chassis dynamometer and modified for continuous use on engine stand
- Use 2 engines for 2 cold starts/day

Gasoline blended to 2002 U.S. national average regular unleaded

- No added oxygenates
- Reid vapor pressure =10.3 psia
- 275 ppm sulfur, 30% aromatics

Pennzoil® multi-grade oil

AC Delco Duraguard® filters

Exposure concentrations not yet determined

- Likely same dilutions as diesel

Exposures began on October 18

[www.nercenter.org]

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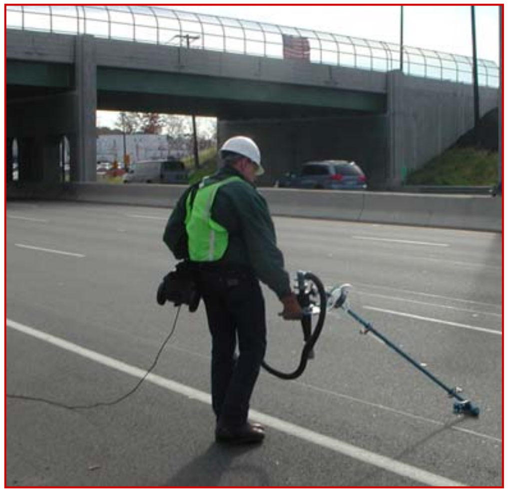
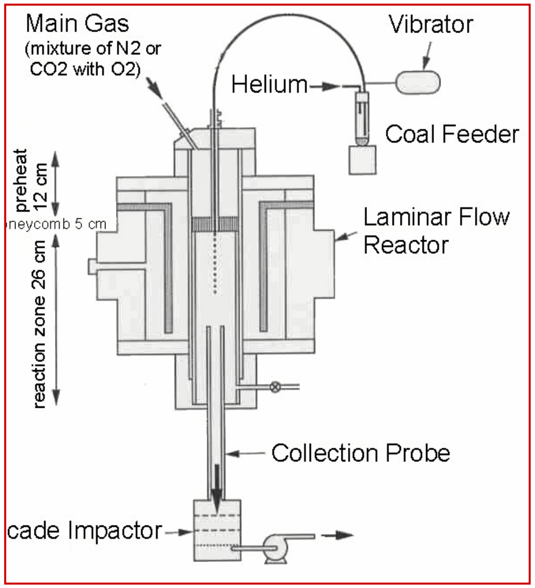
NEXT SOURCE EMISSIONS



Paved street dust

- Collecting dust for pilot study
  - Los Angeles
  - Albuquerque
  - New York/New Jersey
  - Phoenix
- Determine range of composition and acute toxicity
- Determine strategy for main study material
- Main study begins fall 2005



Simulated “downwind” coal emissions

- Workshop specified target composition
  - Simulate key coal emission-derived components
- Assembling “drop-tube” furnace system
- Pilot study comparing PRB and LSSA coals
- Main study begins fall 2006

SUMMARY

Recent Advances:

- Importance of vapor-phase SVOCs
- Relative toxicity of diesel and gasoline emissions
- Importance of high-emitters and oil-derived emissions
- Utility of multivariate analyses for composition vs health hazard
- Reduction of health impacts by low-sulfur fuel and catalyzed PM trap
- Initiated NERC study of gasoline emissions

Underway:

- Importance of ultrafines/nanoparticles, and oil emissions
- Comparative hazards of environmental PM samples
- Evaluation of new emission reduction technologies
- Validation of aerosol exposures of cultured cells
- Preparation for NERC street dust and coal emissions studies