Understanding The Health Implications Of Current And Potential Future Fuels

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Health Effects Institute

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Understanding the Health Effects of Exposure to Fuels and Vehicle Emissions

• What are the sources of exposure?
• What do we know about health effects?
• How are fuels and technology changing?
  • What might that mean for health?
• Concluding Thoughts
Major Vehicle/Fuel Emissions

- Carbon Monoxide
- Carbon Dioxide (Climate Change)
- Diesel Exhaust
- Particulate Matter (PM)
- Lead
- Nitrogen Oxides (NOx) and Hydrocarbons (HC)
  - Precursors to Ozone and PM
- Nitrogen Dioxide

- Air Toxics
  - Aldehydes
    - formaldehyde
    - acetaldehyde
    - others
  - Benzene
  - 1,3-butadiene
  - Metals
  - Polycyclic organic matter (e.g. PAHs)
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What are the Sources of Exposure?
## Estimated Average Contributions of Motor Vehicle Emissions to Ambient Levels of Major Air Pollutants in Developed Countries

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Percent Contribution</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon Monoxide</td>
<td>~90%</td>
<td>EPA (2000)</td>
</tr>
<tr>
<td>PM10</td>
<td>~20%- 25%</td>
<td>DETR (1999)</td>
</tr>
<tr>
<td>PM2.5</td>
<td>~25% - 30%</td>
<td>DETR (1999)</td>
</tr>
<tr>
<td>Nitrogen Oxides</td>
<td>~40%</td>
<td>EPA (2000)</td>
</tr>
<tr>
<td>Average Air Toxics</td>
<td>~21%</td>
<td>EPA (1999)</td>
</tr>
<tr>
<td>Urban Air Toxics</td>
<td>~42%</td>
<td>EPA (1999)</td>
</tr>
</tbody>
</table>
Contribution of motor vehicles primary emissions to ambient PM 2.5 in the Los Angeles Metropolitan Area (Schauer, 1996)

<table>
<thead>
<tr>
<th>Location</th>
<th>% Diesel Contribution</th>
<th>% Gasoline Contribution</th>
<th>Total % Vehicle Contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pasadena</td>
<td>18.8</td>
<td>5.7</td>
<td>24.5</td>
</tr>
<tr>
<td>Downtown L.A.</td>
<td>35.7</td>
<td>6.5</td>
<td>42.2</td>
</tr>
<tr>
<td>West L.A.</td>
<td>18.0</td>
<td>5.7</td>
<td>23.7</td>
</tr>
<tr>
<td>Rubidoux</td>
<td>12.8</td>
<td>0.7</td>
<td>13.5</td>
</tr>
</tbody>
</table>
**PM$_{2.5}$ and Total Carbon Source Contribution Estimates**

**PM$_{2.5}$**
- Gasoline ("cold starts") 24%
- Gasoline (hot stabilized) 3%
- Diesel Exhaust 10%
- Paved Road Dust 17%
- Wood burning (softwood) 2%
- Wood burning (hardwood) 3%
- Ammonium nitrate 13%
- Powerplant (coal) 13%
- Meat Cooking 7%
- Ammonium sulfate 4%
- Gasoline ("smoker") 3%

**Total Carbon**
- Carbon 25%
- Powerplant (coal) 26%
- Paved Road Dust 4%
- Diesel Exhaust 25%
- Gasoline (hot stabilized) 5%
- Wood burning (hardwood) 4%
- Ammonium sulfate 3%
- Meat Cooking 7%
- Gasoline ("cold starts") 6%

Source: Northern Front Range Air Quality Study
Sources of Ozone

Figure 12. National Trends in NO\textsubscript{x} Emissions, 1970–2003.

Mobile Sources of Air Toxics (MSATs)

- New Critical Review of the Literature on Exposure and Health Effects
- HEI Air Toxics Review Panel
  - Thomas Kensler, Chair, Johns Hopkins Bloomberg School of Public Health
- Selected and reviewed priority MSATs based on
  - Exposure
  - Health
  - Mobile Source Contribution
21 Mobile-Source Air Toxics (MSATs)

Priority MSATs selected based on ambient exposures (and role of mobile sources), and toxicity (particularly in humans)

- acetaldehyde
- acrolein
- arsenic cmpds
- benzene
- 1,3-butadiene
- chromium cmpds
- diesel PM and diesel exhaust organic cmpds*

- dioxin/furans
- ethylbenzene
- formaldehyde
- n-hexane
- lead cmpds
- manganese cmpds
- mercury cmpds

- MTBE
- naphthalene
- nickel cmpds
- POM
- styrene
- toluene
- xylene
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*Because of (a) all of the review activity of HEI and others on diesel and (b) the expected reductions in emissions with the 2007 and 2010 engine technologies, the Panel elected not to place it on the list of targeted air toxics – however, an expanded overview and summary is being developed.
Benzene Exposure: To What Extent are Mobile Sources a Significant Source?

- Urban roadside and urban in-vehicle higher than typical highest ambient levels measured
  - mobile sources likely to be important component of overall exposure
- Personal exposures to benzene appear to be in the same range as outdoor settings

Figure 8. Concentrations of benzene (µg/m³) at various locations. Data for figure are from Table 4.
Contribution of Mobile Sources to Overall MSAT Exposure

• 1,3-Butadiene > Benzene > Formaldehyde, Acetaldehyde > Acrolein

• Polycyclic Organic Matter (POM)
  • Depends on specific species
  • PAH: clear mobile source impact

• Naphthalene
  • Insufficient data, but likely low mobile source contribution
What Do We Know About Health Effects?
Particulate Matter (PM)

- High levels of PM (> 500 µg/m³) known to cause premature death
  - e.g. London 1952
- Studies in US, Europe, elsewhere have found association of PM with death at much lower levels (<50 µg/m³)
  - To date, no evidence of a “threshold” (safe level)
**Long Term PM Effects: “ACS” Study**

Approximately 5% increase in mortality for each 10 $\mu g/m^3$ PM$_{2.5}$

*HEI Reanalysis (Krewski, et al, 2000)*

- **Key Cohort Studies**
  - **American Cancer Society:** 550,000 individuals; 150 cities, many years
  - Detailed health, socioeconomic info on each subject
  - **2000 HEI reanalysis:** Validated results, identified robust associations of mortality with PM2.5, SO2, Sulfate
  - **Analysis recently extended as population ages, found new effect of PM2.5 on lung cancer, heart effects (Pope, et al)**

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**Risk of Cardiopulmonary Mortality**

(ACS 2 Pollutant Models)

- Relative Risk of Death
  - 0.8
  - 0.9
  - 1
  - 1.1
  - 1.2
  - 1.3
  - 1.4
  - 1.5

- **PM$_{2.5}^{}$**
- **Sulfate**
- **SO$_2$**
ACS-based Study In Los Angeles finds Larger Long-Term Effects on Mortality (Jerrett, et al 2005)

23,000 cases from ACS Cohort in LA. Mortality risk 3 times higher than ACS: 16% 10 µg/m³

Funded by HEI, in review
Short Term (Daily) PM Effects
National Morbidity, Mortality and Air Pollution Study (NMMAPS)
20 largest US cities
(Daniels et al HEI 2004)
A Key PM Question Going Forward: “Are all particles created equal?”

- PM the only complex mixture we regulate
- Some components and sources may be more toxic
  - And thus deserving of more attention
- Among the “prime suspects”:
  - Transition metals
  - Organic carbon compounds (e.g. PAHs) on diesel PM
  - Ultrafine PM?
- HEI National Particle Component Toxicity Initiative (HEI NPACT) underway
**Ozone**

- **Known to:**
  - Cause inflammation in respiratory tract
  - Reduce ability to breathe (lung function) for some
  - Increase hospitalization for asthma, other lung diseases

- **Recent multi-city evidence of effects on premature mortality**
- **Effects have been demonstrated for short term exposure**
  - *Long term effects are less certain*
Ozone Health Effects

- Some humans have been shown to have reduced lung function (measured as FEV1) after exposure to ozone.
Recent Multi-City Studies of Ozone and Mortality
Benzene

- **Occupational Studies: Cancer**
  - Follow-up of Pliofilm cohort (leukemia)
  - New Cohorts (e.g., Australian petroleum workers, gas & electric utility workers) leukemia at lower exposures

- **Supporting evidence from:**
  - Biomarkers
  - Genetics

- **Clear and widely accepted evidence from variety of occupational studies that risks of acute myeloid leukemia are increased**
  - Less certainty concerning other lymphohematopoietic cancers
Benzene Health Effects at Ambient Exposure Concentrations?

Community Studies of Cancer
As with other compounds, identifying effects in community studies is challenging
• Risk for childhood leukemia associated with proximity to petrochemical works and petrol stations in some studies
  • Not possible to single out benzene
  • Mixed results on association between traffic and childhood leukemia

Hematological outcomes
Recent studies reveal effects on hematological indices @ lower levels
• China: effects observed in lowest exposed group  (≤ 0.25 ppm [≤ 815 µg/m³]) compared w/ controls
• US: no assoc. between any hematologic indicator & mean benzene exposure between 0.14 and 0.60 ppm [46 and 1,960 µg/m³]

Thus, considerable uncertainty as to the lowest concentration that might be associated with hematologic effects
Acetaldehyde, Acrolein, and Formaldehyde

- **Acetaldehyde:**
  - *Irritating* (eye, skin, respiratory tract).
  - Data on carcinogenicity inadequate.
  - Current environmental concentrations well below levels causing irritation.

- **Acrolein:**
  - *Very irritating* to respiratory tract. Chronic inhalation studies: inflammation.
  - Environmental concentrations & personal exposures lower than concentrations known to cause irritation. (but not that much lower)

- **Formaldehyde:**
  - *Irritating* (eye, skin, respiratory tract).
  - Recently classified as *human carcinogen* (IARC): nasopharyngeal cancer at levels historically encountered in industry. Mechanism not fully understood.
  - Exposures today dominated by indoor exposures
“Traffic Effects”

- Growing number of studies looking at exposures and effects at roadside
  - High levels of some pollutants
  - Substantial populations potentially affected especially in urban areas
  - Exposure, Animal, and Epidemiology Studies
  - Crude Exposure metrics
    - Difficult to separate sources
- HEI Expert Review of over 400 studies underway
Testing Mobile and Other Source Contributions to Personal Exposure
RIOPA Study (Weisel, et al)
Association between proximity to truck traffic and symptoms (Brunekreef 1999)

- 2500 Children in 24 schools, located near freeways with varying traffic density (i.e. truck vs. car)

![Bar chart showing prevalence ratio for asthma, hayfever, phlegm, HD allergy, pet allergy, and wheeze last for >10,000 vs <10,000 trucks.]
Found elevated risk of Myocardial Infarction for those in traffic 2 hrs prior to event

- Limited Association with PM10, SO2
  - None with PM2.5, UF)
- Noise, stress could also play role
How are fuels and technologies changing?
How are fuels and technologies changing?

- Tighter standards for fuels and emissions
  - Reformulated Gasoline
  - Ultralow Sulfur Diesel
  - Tighter emissions standards
    - Auto Tier 2, CALLEV, Diesel on- and non-road, EPA Mobile Source Air Toxics Rule

- Alternative Fuels and Propulsion
  - Biofuels (ethanol, biodiesel)
  - Hybrid/electric
  - Hydrogen/fuel cell
Improvements in PM and NOx Diesel Emission Standards
(Both Fuel and Engine Requirements)
**Onroad Diesel Reductions 1975 - 2000**

(Source: HEI Tunnel Study in PA (2002))
Most PM filtration systems being considered for 2007 are the wall flow type shown on the left. Without regeneration to oxidize soot these devices quickly plug.

Catalyzed filtration systems like the J-M CRT® shown on the right reduce regeneration temperature by producing NO₂ from exhaust NO in an oxidizing catalyst upstream of filter.

The J-M CCRT® has a catalyzed washcoat on the filter as well to further reduce regeneration temperature.

NO₂ in the exhaust is an issue.

In most applications active regeneration is also required.
Continued Progress:
Impact of Mobile Source Regulations on Diesel PM2.5
(Source: EPA)
Reformulated Fuels:
47% reduction in ambient Benzene levels (US)

- Substantial reduction after 1995 RFG
- Largely through replacement with oxygenates
  - MTBE first
  - Now ethanol
- Further reductions mandated in 2007
  - Mobile Source Air Toxics (MSAT) Rule
Looking Ahead

• **Gasoline and Diesel Vehicles:**
  • Continued improvement in fuels, emissions
  • Hybrid and Plug in Electric

• **Natural Gas Vehicles**
  • Additional controls (catalysts)

• **Fuel Additives**
  • Biofuel blends (ethanol, biodiesel)
  • Metal additives? (iron, manganese, cerium)

• **Other Alternative Fuels**
  • Hydrogen (fuel cells)?
  • Gas-to-liquid fuels? (Fischer Tropsch)
Health Challenges with New Fuels and Technology

- Will the new technology continue to work?
- Are the new technologies really cleaner?
- Are there unintended consequences?
New Technologies: How Long Will They Last?  
(Seagrave, 2002)

- PM and semi volatile exhaust from normal and high emitting diesel and gasoline
- Subjected to a host of laboratory tests of inflammation, mutagenicity, toxicity
- **Results:**
  - Diesel and Gasoline results similar
  - Both High-Emitting Diesel and Gasoline showed substantially greater effect
- **Lube oil likely a key component**
New Technologies:  
How clean are they?

- Results of California bus engine testing
- Diesel w,w/o filter; natural gas (CNG) w,w/o catalyst

<table>
<thead>
<tr>
<th>Most Significant Emissions</th>
<th>HIGHEST</th>
<th>LOWEST</th>
</tr>
</thead>
<tbody>
<tr>
<td>1  NO\textsubscript{X}</td>
<td>Diesel base ~ Diesel/CRT &gt; CNG~CNG w/cat</td>
<td></td>
</tr>
<tr>
<td>2  Total PM Mass</td>
<td>Diesel baseline &gt;&gt; CNG &gt; CNG w/cat ~ Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>3  Total Ultrafine Particle Number*</td>
<td>Diesel baseline &gt; CNG ~ CNG w/cat ~ Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>4  Aldehydes**</td>
<td>CNG &gt;&gt; CNG w/CAT &gt; Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>5  Mutagenicity</td>
<td>CNG &gt;&gt; Diesel baseline ~ CNG w/CAT ~ Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>6  PAH Species***</td>
<td>Diesel baseline &gt; CNG &gt; CNG w/CAT ~ Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>7  NO\textsubscript{2}/NO\textsubscript{X}</td>
<td>Diesel/CRT &gt; Diesel baseline ~ CNG &gt; CNG w/CAT</td>
<td></td>
</tr>
<tr>
<td>8  CO\textsubscript{2}</td>
<td>Diesel/CRT ~ Diesel baseline ~ CNG ~ CNG w/CAT</td>
<td></td>
</tr>
<tr>
<td>Other Measured Emissions</td>
<td></td>
<td></td>
</tr>
<tr>
<td>9  Nonmethane Hydrocarbons</td>
<td>CNG &gt;&gt; Diesel baseline &gt; Diesel/CRT ~ CNG w/CAT</td>
<td></td>
</tr>
<tr>
<td>10 Other Toxic Hydrocarbons</td>
<td>CNG &gt; Diesel baseline &gt; CNG w/CAT ~ Diesel/CRT</td>
<td></td>
</tr>
<tr>
<td>11 CO</td>
<td>CNG &gt; CNG w/CAT &gt; Diesel baseline &gt; Diesel/CRT</td>
<td></td>
</tr>
</tbody>
</table>
### Table 1. Summary of Studies Examining MTBE Contamination of Drinking Water Sources

<table>
<thead>
<tr>
<th>Concentration Range (ppb)</th>
<th>California Public Water Sources&lt;sup&gt;1&lt;/sup&gt; (wells)</th>
<th>Maine Public Water Sources&lt;sup&gt;3&lt;/sup&gt; (wells)</th>
<th>Maine Private Water Sources&lt;sup&gt;3&lt;/sup&gt; (wells)&lt;sup&gt;4&lt;/sup&gt;</th>
<th>USGS/NAWQA Studies&lt;sup&gt;5&lt;/sup&gt; (wells)</th>
<th>USGS/EPA 12 Northeastern State Study&lt;sup&gt;6&lt;/sup&gt; (systems)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>N=5,195 MDL=5 ppb</td>
<td>N=793 MDL=0.1 ppb</td>
<td>N=946 (95% CI) MDL=0.1 ppb</td>
<td>N=2,743 MDL=0.2 ppb (censor level)</td>
<td>N=1,190 MDL=1 ppb&lt;sup&gt;7&lt;/sup&gt;</td>
</tr>
<tr>
<td>Non-Detects</td>
<td>~99%</td>
<td>~84.1%</td>
<td>~84.3%</td>
<td>~94.7%</td>
<td>~92.8%</td>
</tr>
<tr>
<td>MDL - 5 ppb</td>
<td>N/A&lt;sup&gt;2&lt;/sup&gt;</td>
<td>~14.6%</td>
<td>~12.8%</td>
<td>~4.5%</td>
<td>~5.0%</td>
</tr>
<tr>
<td>5-20 ppb</td>
<td>~0.3%</td>
<td>~0.9%</td>
<td>~1.5%</td>
<td>~0.4%</td>
<td>~1.3%</td>
</tr>
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<td>&gt; 20 ppb</td>
<td>~0.3%</td>
<td>~0.4%</td>
<td>~1.5%</td>
<td>~0.4%</td>
<td>~0.9%</td>
</tr>
</tbody>
</table>
## Fuel Additives: Ethanol

**Acetaldehyde, Acrolein, and Formaldehyde Exposures**

<table>
<thead>
<tr>
<th></th>
<th>Ambient</th>
<th>In-vehicle or near-roadway</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Acetaldehyde</strong></td>
<td>1 – 7 µg/m³</td>
<td>0.7 – 7 µg/m³</td>
</tr>
<tr>
<td><strong>Acrolein</strong></td>
<td>0.03 – 6 µg/m³</td>
<td>0.1– 6 µg/m³</td>
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<td><strong>Formaldehyde</strong></td>
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<td>36.1 – 55 µg/m³</td>
<td>4.3 – 438 µg/m³</td>
</tr>
<tr>
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<td>0.03 – 6 µg/m³</td>
<td>0.1 – 6 µg/m³</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>1 – 6 µg/m³</td>
<td>5 – 20 µg/m³</td>
</tr>
<tr>
<td>Formaldehyde</td>
<td>1 – 54 µg/m³</td>
<td>17 – 80 µg/m³</td>
</tr>
</tbody>
</table>
Metal Additives

• Since lead, no major metallic additives in US
• But substantial, and potentially growing, use in developing world
  • Relative low cost octane enhancement
  • Manganese and iron (ferrocene) most prominent
• Many metals known to cause neurotoxic and other effects in high doses, in workers
• Less is known at low doses
  • But metals are among the PM “Prime Suspects”
Concluding Thoughts

• **There Is No Perfect Fuel**
  • Need thorough lifecycle evaluation

• **It’s Not Just The Fuel**
  • Fuel/Engine systems work together
  • And the lube oil is likely important too…

• **Expect the Unexpected**
  • Unintended consequences WILL occur

• **Start Measuring Impacts Today**
  • We are changing the fuel supply as we speak
  • “Accountability” studies should get underway now
Thank You

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