HEI COMMUNICATIONS

Diesel Workshop: Building a Research Strategy to Improve Risk Assessment

March 7–9, 1999
Stone Mountain, Georgia

Health Effects Institute

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HEI

Workshop

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Building a Research Strategy to Improve Risk Assessment

DIESEL WORKSHOP
BUILDING A RESEARCH STRATEGY TO IMPROVE RISK ASSESSMENT
Stone Mountain, Georgia
March 7–9, 1999

SUNDAY, MARCH 7

Luncheon: 12 noon

Introductory Remarks, *Kathleen Nauss,* HEI

1:00 P.M.	**SESSION I: Risk Assessments of Diesel Emissions: Framework for Building a Research Strategy** **Chair,** *Dan Greenbaum,* HEI **Introduction** *Dan Greenbaum* **Overview of the Epidemiology and Toxicology Information That Supports Hazard Identification** *Paolo Boffetta,* International Agency for Research on Cancer **Information Gaps in Current Risk Assessments and Perspectives on Future Research Needs** *Vanessa Vu,* U.S. Environmental Protection Agency *Joe Mauderly,* Lovelace Respiratory Research Institute and Chairman of the EPA's Clean Air Scientific Advisory Committee **Invited Comments** *William Bunn,* Navistar *Janet Hathaway,* National Resources Defense Council **Discussion**
3:45 P.M.	**Break**
4:00–6:00 P.M.	**SESSION II: Chemical and Physical Properties of Diesel Engine Emissions** **Chair,** *Robert Sawyer,* University of California, Berkeley, and HEI Research Committee **Characterization of Diesel Particulate Matter: Impact of Measurement Techniques** *David Kittelson,* University of Minnesota **Characterization of Heavy-Duty Vehicle Emissions** *Nigel Clark,* West Virginia University **Characterization of Light-Duty Vehicle Emissions** *Peter Kohoutek,* Volkswagen AG, Germany (presenter: Michael Spallek) **Changes in Diesel Engine Emissions Over the Last Two Decades** *Barbara Zielinska,* Desert Research Institute **Panel Discussion: What Are the Research Needs for Emissions Characterization from the Perspective of Health Research?** **Panel:** *Lester Grant,* U.S. Environmental Protection Agency; *Bert Brunekreef,* University of Wageningen, The Netherlands
6:30 P.M.	**Reception and Dinner** **After Dinner Remarks** *Margo Oge,* Director of Office of Mobile Sources, US EPA

MONDAY, MARCH 8

8:00 A.M.	**SESSION III: Assessment of Exposure to Diesel Engine Emissions** **Chair,** *Philip Lorang,* Environmental Protection Agency, Office of Mobile Sources **Considerations in Assessing Exposure to Diesel Engine Emissions in Occupational and Ambient Settings** *Brian Leaderer,* Yale University and HEI Diesel Epidemiology Expert Panel **Comparison of Different Analytical Approaches for Measuring Constituents of Diesel Emissions in Australian Mines** *Alan Rogers,* OH&S, Pty. Ltd., Australia **Contributions of Diesel Engine Emissions to Ambient Particulate Matter** **Measurement Methods and Atmospheric Modeling Approaches** *Glen Cass,* California Institute of Technology and HEI Research Committee **Northern Front Range Ambient Air Quality Study** *Douglas Lawson,* National Renewable Energy Laboratory **Use of Models to Estimate General Population Exposures to Diesel Particulate Matter** *Alison Pollack,* Environ International Corporation
10:30 A.M.	**Break**
10:45 A.M.	**SESSION IV: What Do Published Epidemiology Studies Tell Us About Exposure-Response?** **Chair,** *John C. Bailar III,* The University of Chicago, HEI Review Committee and Diesel Epidemiology Expert Panel Chair **Introduction** *John C. Bailar, III* **Organizational Framework for Evaluating Epidemiologic Data for Use in Quantitative Risk Assessment** *G. Marie Swanson,* Michigan State University **Discussion of Exposure-Response Analyses in Published Epidemiology Studies** *Robert Sawyer,* University of California, Berkeley *David Hoel,* Medical University of South Carolina **Panel Findings and Recommendations** *John C. Bailar, III* **Questions and Discussion**
12:15 P.M.	**Luncheon and Afternoon Break**
3:00–5:30 P.M.	**SESSION V: What Will Epidemiology Studies Now Underway Tell Us About Exposure-Response?** **Co-Chairs,** *Charles Poole,* University of North Carolina, Chapel Hill, and HEI Diesel Epidemiology Expert Panel; *Gerald van Belle,* University of Washington and HEI Research Committee **Diesel Exhaust and Lung Cancer Mortality in German Potash Miners** **Cohort Study Design** *Robert Säverin,* Federal Institute of Occupational Safety and Health, Germany **Exposure Assessment** *Dirk Dahmann,* Institute for Research on Hazardous Substances, Germany **Results** *Robert Säverin* **Questions and Discussion** **Diesel Exhaust and Cancer in Danish Bus Drivers** **Study Design and Cancer Findings** *Jørgen Olsen,* Danish Cancer Society, Denmark **Biomarker Results** *Herman Autrup,* University of Aarhus, Denmark **Questions and Discussion**

MONDAY, MARCH 8 *(Continued)*

New HEI Research: Feasibility Studies to Identify Cohorts and Improve Exposure Assessment—Introduction to Poster Presentations
Diane Mundt, HEI

5:45 P.M. **Reception and Poster Presentations**

HEI Feasibility Studies
Paolo Boffetta, International Agency for Research on Cancer, France, and *John Cherrie*, Institute of Occupational Medicine, Scotland
Murray Finkelstein and *David Verma*, McMaster University, Canada
Eric Garshick, Channing Laboratory and *Thomas Smith*, Harvard School of Public Health
David Kittelson and *Gurumurthy Ramachandran*, University of Minnesota
Alan Gertler and *John Sagebiel*, Desert Research Institute
Barbara Zielinska and *Eric Fujita*, Desert Research Institute

7:00 P.M. **Dinner**

TUESDAY, MARCH 9

8:30 A.M. **SESSION VI: What Will Epidemiology Studies Now Underway Tell Us About Exposure-Response? (continued)**

Co-Chairs, *Charles Poole* and *Gerald van Belle*

Lung Cancer and Diesel Exhaust Among Non-Metal Miners in the United States

Cohort Study Design
Michael Attfield, National Institute for Occupational Safety and Health (NIOSH)

Case-Control Study Design
Debra Silverman, National Cancer Institute (NCI)

Industrial Hygiene Assessment
Dan Yereb, NIOSH

Historical Exposure Assessment
Mustafa Dosemeci, NCI

Questions and Discussion

10:30 A.M. **Break**

10:45 A.M. **Panel and General Discussion of Epidemiology Studies Presented in Sessions V and VI**

Panel: *Brian Leaderer*, Yale University; *Roger McClellan*, Chemical Industry Institute of Toxicology; *Kyle Steenland*, NIOSH; *Marie Swanson*, Michigan State University Cancer Center; *Charles Yarborough*, Caterpillar, Inc.

- How Will Ongoing Epidemiology Studies Contribute to Quantitative Cancer Risk Assessments of Diesel Emissions?
- What Additional Research Is Needed to Improve Quantitative Cancer Risk Assessments?

12:00 P.M. **Luncheon**

1:00 P.M. **SESSION VII: Consideration of Health Endpoints Other Than Cancer in Future Risk Assessments of Diesel Emissions**

Chair, *Michael Lipsett*, California Environmental Protection Agency and University of California, Berkeley

Introduction
Michael Lipsett

Presentations of Ongoing Research

Environmental Diesel Exhaust Exposure and Respiratory Health of Children in The Netherlands
Bert Brunekreef, University of Wageningen, The Netherlands

Inflammatory Effects of Diesel Emissions on Human Airways
Thomas Sandström, University Hospital of Umea, Sweden

Human Allergic Responses
David Diaz-Sanchez, UCLA School of Medicine

TUESDAY, MARCH 9 ***(Continued)***

Panel and General Discussion

Panel: *Thomas Sinks*, Centers for Disease Control and Prevention; *Frank Speizer*, Harvard Medical School and HEI Research Committee; *Jodi Sugerman-Brozan*, Alternatives for Community and the Environment

- What Health Endpoints Other than Cancer Should Be Considered for Quantitative Risk Assessment?
- What Research Approaches Would Advance our Understanding of the Quantitative Relationships Between Exposure to Constituents of Diesel Emissions and Noncancer spiratory Effects?

Closing Comments, *Dan Greenbaum*, HEI

4:00 P.M. **Adjourn**

INTRODUCTION

The Health Effects Institute (HEI) has supported research on the health effects of exposure to diesel emissions for many years. In the early 1980s, this research focused on the role of the organic constituents of diesel exhaust particles in cancer development, through studies of mutagenicity, metabolism, and carcinogenesis. However, an animal bioassay involving exposure of rats to either diesel particles or carbon black produced similar incidence of lung tumor responses in both groups, focusing attention on the role of particles rather than adsorbed chemicals.

A 1995 HEI Special Report, *Diesel Exhaust: A Critical Analysis of Emissions, Exposure, and Health Effects,* raised questions about the relevance of animal data for quantitative cancer risk assessment. The report noted that epidemiologic studies showed consistent small increases in risk for exposed workers, but concluded that the absence of concurrent exposure measurements limited the utility of those studies for quantitative risk assessment.

As part of the Institute's continued interest in diesel engine emissions and health effects and in an effort to understand how current research or possible new research could be useful for quantitative risk assessment, HEI initiated the Diesel Epidemiology Project in 1998. This multifaceted effort included the work of a Diesel Epidemiology Expert Panel (the Executive Summary of the Panel's report is included in these proceedings), a set of 6 feasibility studies (the abstracts of posters presented are included here), and a Diesel Workshop: Building a Research Strategy to Improve Risk Assessment, held March 7–9, 1999. These proceedings summarize the workshop discussion.

WORKSHOP GOAL

The goal of the workshop was to build a research strategy to improve quantitative risk assessment (QRA) of diesel engine emissions. The agenda was designed according to a risk assessment framework to focus discussion on data gaps and research needs. Information learned at the workshop will be used to help HEI and others plan future research of the health effects of diesel emissions.

WORKSHOP PLANNING COMMITTEE

Glen Cass
California Institute of Technology

Aaron Cohen
Health Effects Institute

Michael Lipsett, California Environmental Protection Agency; University of California, Berkeley

Philip Lorang
U.S. Environmental Protection Agency

Charles Poole
University of North Carolina at Chapel Hill

Jonathan Samet
Johns Hopkins University

Robert Sawyer
University of California, Berkeley

Michael Spallek
Volkswagen, AG

Vanessa Vu
U.S. Environmental Protection Agency

Charles Yarborough
Caterpillar, Inc.

HEI WORKSHOP COORDINATORS

Kathleen M. Nauss
Director for Scientific Review and Evaluation

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Staff Scientist

RISK ASSESSMENT ISSUES

Participants in the opening session presented a framework for building a research strategy to improve QRA of diesel exhaust. HEI President Daniel Greenbaum posited two ways of looking at diesel engines: potential problems of adverse health effects and the promise of new technology. He also reviewed the current state of risk assessments of diesel emissions, referring to the work of the International Agency for Research on Cancer, the World Health Organization, the California Environmental Protection Agency (EPA), and draft documents of the U.S. EPA. As mentioned by Greenbaum, and subsequently also by Paulo Boffetta, investigators worldwide consistently have found an elevated risk for lung cancer associated with occupational exposure to diesel engine emissions; the overall increase found is approximately 30%. Other cancers do not appear to be associated with an increased risk from exposure, but they have not been as well studied.

Epidemiologic studies of lung cancer have been conducted primarily in occupational settings. Concurrent exposure data in these studies are not available, however, and information on diesel constituents has not been available to date. Although the association between diesel exhaust and lung cancer is biologically plausible, the mechanism responsible for lung cancer development in humans is unknown.

As discussed by Vanessa Vu of the U.S. EPA, risk assessments require making numerous assumptions and have a range of uncertainties. For diesel exhaust, the hazard has been identified, but not well characterized, with an unknown risk of cancer for low-level exposure. She noted that, at this time, the major uncertainties include the lack of epidemiologic data in ambient settings, information on the role of diesel exhaust constituents, the value of rat lung tumor data in predicting human response, and the lack of understanding of mode of action. For non-cancer effects, there is a lack of exposure-response data for chronic respiratory outcomes in exposed workers, and rat data are used to set the toxicity. Joe Mauderly stressed the need to look at diesel health effects in the context of all particulate matter (PM).

Industry and non-governmental organization representatives William Bunn and Janet Hathaway, respectively, discussed risk assessment information gaps and research needs from their perspectives. Bunn thought that information gaps included the lack of (1) "concurrent" exposure measures in epidemiologic research; (2) particle characterization; (3) evaluation of emissions from current engines; (4) an understanding of why occupational studies, which cover a wide range of exposure levels, yield similar risks; and (5) improved control of smoking in epidemiologic studies. He thought the research needs included a major occupational epidemiologic study, understanding effects of diesel and all particles combined, development of exposure indices, evaluation of the diesel mixture, and studying non-cancer endpoints.

Janet Hathaway pointed out that gaps and uncertainties for assessing risk of diesel emissions do not differ from those that exist for many other environmental agents, and that some occupational exposures (e.g., those of truck drivers), were similar to ambient exposures. In her view, assumptions used to develop quantitative estimates of risk of exposure to diesel exhaust should be refined as new data become available. Again, however, a mechanistic understanding of diesel-induced lung cancer is difficult because a relevant animal model is lacking. Additionally, increasing fine particles in diesel exhaust may in fact enhance its biological effect. She emphasized that it is important to understand mechanisms of exposure affecting health so that technological changes designed to protect health do not in fact make it worse. Non-cancer effects, including asthma, cannot be ignored — however, such outcomes are not only researched less, but they are also funded less substantially than lung cancer. Finally, she noted that interactions between diesel emissions and other allergens need to be considered — the public health impact of understanding mechanisms, especially for asthma, is important, given that improved understanding can lead to better control.

Daniel Greenbaum opened the discussion following the session presentations by noting that diesel exhaust is a moving target, in that we are not really sure what the most important focus of research should be — total mass, fine particles, or polycyclic aromatic hydrocarbons (PAHs). The question then is how do we project what is most important into the future? The need to distinguish diesel exhaust from the larger particulate matter (PM) debate is not clearly agreed on. Much of the discussion during Session 1 focused on whether separating diesel PM from ambient PM was possible or even necessary. Some participants thought that "hot spots"— that is, areas where diesel contributions to pollution are high — could be considered for more specific human diesel-exposure studies. It was also suggested that the impact on health was the real issue when examining air pollution effects, and it was not necessarily relevant if the cause was diesel particulate matter (DPM) defined as separate, or in addition to, ambient PM. A "holistic" approach was suggested as well — rather than studying one component at a time, "air pollution" should be considered, and research should focus on cancer and non-cancer health outcomes.

There were differences of opinion on the importance of cigarette smoking as a confounder in epidemiologic studies of diesel exhaust exposure. Some participants thought the issue had been adequately addressed, and others thonght that the association between smoking and lung cancer is so great that it could affect the magnitude of the association (relative risk) between diesel exhaust and lung cancer. The possibility of a synergistic effect between smoking and diesel engine exhaust was also suggested.

CHARACTERIZATION OF DIESEL EMISSIONS

The second session included presentations about current information on the physical and chemical characteristics of diesel exhaust as well as changes in emissions from heavy-duty (HD) and light-duty vehicles over the last 20 years. David Kittleson noted that one of the problems in characterizing exposure is understanding what happens to constituents of diesel exhaust when they leave the tailpipe. One challenge is to design a dilution and sampling system that produces accurate measurements of exposure. Significant gas-to-particle conversion takes place on exhaust cooling after leaving the tailpipe, with a thousand-fold dilution occurring within a few seconds. On emission, new particles form by nucleation, pre-existing particles grow, and nucleation/adsorption are competing processes. These processes need to be understood and accounted for in determining human exposures. Kittleson thought that sampling conditions should mimic atmospheric dilution in order to obtain samples that represent human exposure.

The impact of after-treatment additives on emissions from HD vehicles is essentially unknown, according to Nigel Clark. He noted that continuous PM data are generally not available, and although correction factors for speed and engine degradation can be applied, there is little information on cold-start emissions or the effects of terrain, location, altitude, temperature, and maintenance. Clark raised the important point that in-use emissions from laboratory measurement data do not accurately reflect real-use emissions. Alternative fuels, improved diesel formulations, and blends with natural gas–derived Fischer-Tropsch liquids, if used in diesel engines, offer potential advantages for improved emissions.

In his discussion of unregulated light-duty diesel engine emissions, Michael Spallek indicated that tests at Volkswagen using a formulation of diesel known as "Swedish diesel fuel" in vehicles substantially reduced the ozone-forming potential in emissions compared with using a European "reference" diesel fuel. He further noted that emissions of PAHs adsorbed to diesel particulates from current diesel engines are 95% less than those emitted in the 1980s; a further reduction in PAHs was shown when Swedish fuels were compared with the reference diesel fuel.

Discussing historical trends in diesel emissions over the last 20 years, Barbara Zielinska noted that a comparison of HD diesel emissions over time is not straightforward. Differences in testing procedures (engine dynamometers, chassis dynamometers, and tunnel studies) and the units to express emission rates provide the greatest challenges. She indicated that several factors affecting emissions from HD vehicles include vehicle weight, driving activity, fuel composition, engine exhaust after-treatment, and vehicle age.

Zielinska also pointed out the wide variability seen in the organic carbon (OC)/elemental carbon (EC) ratios in emissions data depend on the model year of the vehicle. Although PAH emissions show a declining trend as a function of model year, PAH and nitro-PAH composition in emissions have not changed significantly in the last 20 years. Exhaust after-treatments and improved fuel reformulation have had impact on emissions, however, but the possible health effects of such changes are unknown.

In the discussion, the point was made that most of the PM in diesel emissions has a mean aerodynamic diameter of less than 2.5μm. Little is known, however, of what happens to diesel PM once it is out of the tailpipe — for instance, how long are particles in the air? How are emissions transformed and dispersed? How far will they travel? Besides the PM, other substances emitted (such as carbon monoxide and nitrogen oxides) need to be understood. When determining health endpoints, there is a need to consider contributions of source emissions.

Concern was expressed not only about the size of particulate matter, but also about the number of particles and their composition, including transition metals. Needed are studies across engines and fuel types to characterize emissions, focusing on particle changes and on tradeoffs to health as well as ozone levels and climate. It appears that new on-road data available give a picture of what is emitted that is different from previous findings. Consistency of measurements was generally agreed to be vital to comparability of investigations.

EXPOSURE TO DIESEL EMISSIONS

Assessment of exposure is one of the most critical components for QRA. Brian Leaderer discussed currently used or proposed markers of diesel exhaust emissions including EC, submicron particles, PAHs, extractable mass, NO_x, CO, CO_2, and particle phase nicotine (environmental tobacco smoke correction). He noted, however, that diesel engines are not the only source of these exposures, particularly EC, which is the most commonly used diesel marker. Using EC as a diesel marker needs consideration of other potential area sources contributing EC. He indicated that stating the limitations of methods used and specifying uncertainties were important when assessing exposure.

Alan Rogers's presentation focused on various analytic approaches for measuring exposures of diesel emissions in Australian mines and subsequently modeling results. He reported considerable variability in EC — from 30% to 80% of diesel submicron mass in personal-exposure samples. The EC/total carbon ratios, however, tend to be consistent over equipment that differs both in type and age.

Glen Cass discussed characterization of fine particle emissions from diesel engines and subsequent air quality modeling methods used to determine how various sources produce the particle mixture. There are multiple sources of carbon particle emissions, and particle size varies substantially by source. When source apportioning, potential contributors to aerosol composition need to be investigated and characterized; in particular, area measurements of restaurant emissions, tire dust, wood smoke, and gasoline vehicle emissions, for example, should be collected with ambient measures of PM.

Douglas Lawson presented findings from the collaborative Northern Front Range Air Quality Study (NFRAQS). A copy of the full report is available on-line at *http://nfraqs.cira.colostate.edu*. One objective of the study, which was conducted along Colorado's Front Range, was to identify sources or contributors to $PM_{2.5}$ in the NFRAQS region. Some important sources of $PM_{2.5}$ in the Denver metropolitan area were gasoline exhaust (28%), dust and debris (16%), and diesel exhaust (10%). Particulate ammonium nitrate and sulfate were also large contributors. As part of the discussion, it was mentioned that more time needs to be spent looking at "fugitive emissions" — that is, pollutants from paved road, tires, and braking dust — not from vehicle tailpipes.

In developing models of emissions, on-road HD emissions are the major consideration, because they are the largest source of diesel PM in the United States; however, more light-duty vehicle data should be included in updates, according to Alison Pollack. She noted that emission models are generally based on vehicle certification data and not real-world testing. As a result, there is a need for both in-use emissions and improved activity estimates. Additionally, understanding how various engines, roads, temperatures, and variations in humidity affect emissions is needed. Current models are based on very little data, and activity estimates are based on a large number of assumptions. Even fewer data are available for off-road models, which include over 300 equipment types. There is a need to understand the uncertainties. The discussion that followed focused on how we can incorporate new in-use emissions data into existing models.

EXPOSURE-RESPONSE: REPORT OF HEI'S DIESEL EPIDEMIOLOGY EXPERT PANEL

The application of exposure assessment in epidemiologic studies of diesel exhaust has been limited. However, two epidemiologic studies for which industrial hygiene data were retrofitted to provide estimates of past exposure have been considered for use in quantitative risk assessments of diesel emissions. As part of the HEI's Diesel Epidemiology Project, the Institute's Diesel Epidemiology Expert Panel was, at the time of the workshop, in the process of finalizing its Special Report. Included with these proceedings is the Executive Summary of the Panel's report, *Diesel Emissions and Lung Cancer: Epidemiology and Quantitative Risk Assessment*. A copy of the full report is available at *http://www.healtheffects.org*.

HEI DIESEL FEASIBILITY STUDIES

Another aspect of the HEI Diesel Epidemiology Project is six feasibility studies funded in 1999 in response to RFA 98-3. Three of these studies are designed to identify new cohorts that might be appropriate for an epidemiologic study of diesel exhaust. The focus of the other three studies is new methods of exposure assessment and characterization. These six studies were presented in a poster session.

EXPOSURE-RESPONSE: ONGOING STUDIES

The purpose of this session was to become familiar with ongoing epidemiologic studies of diesel exhaust, particularly for their potential contribution to QRA. There are, in fact, a limited number of epidemiologic studies currently underway that examine the association of diesel exhaust and lung cancer. German investigators are following a cohort of potash miners for lung cancer mortality, and preliminary findings of a study of Danish bus drivers and tramway workers were presented.

Robert Säverin reported the findings of the German potash miners cohort study. Workers were followed from 1970 to 1994 for the development of lung cancer. Two exposure groups were compared for lung cancer risk — production (highly exposed) and workshop (lowest exposure). Results showed an increased risk for lung cancer, based on 11 and 6 deaths respectively. Dirk Dahmann discussed the exposure assessment aspects of this study, which included personal and stationary samples.

Jørgen Olsen presented the study of Danish bus drivers and tramway workers. The study was designed to identify the incidence of cancer among these workers, and to compare rates of cancer observed to those expected in the general population. The exposure of interest was air pollution, as measured by respirable dust, NO_2, and PAH levels, not specifically diesel exhaust. Olsen also presented preliminary results of a nested case-control study. Herman Autrup discussed preliminary biomarker results, looking at DNA adducts in bus drivers (exposed) and postal workers (unexposed). He noted that the markers were not specific for compounds in diesel exhaust.

One of the largest epidemiologic studies of diesel-exposed individuals currently underway is being conducted in the United States by the National Institute of Occupational Safety and Health (NIOSH) and the National Cancer Institute (NCI). This is a cohort study of U.S. non-metal miners, with a nested case-control component as well as current-exposure assessment measurements and detailed historical exposure reconstruction for study participants.

Michael Attfield presented the cohort mortality study aspect, which will identify any causes of death, including lung cancers, that may be elevated. Incidence of causes of death will be compared by extent of workers' diesel exhaust exposure. The cohort includes underground miners (exposed) as well as surface workers who are essentially not exposed to diesel exhaust. The retrospective follow-up period will range up to 48 years. The study will be done in two phases — the first using job category to determine exposure level, the second using quantitative exposure-response determined through various metrics. The first report is expected in 2003.

Debra Silverman described the nested case-control component of the NIOSH/NCI study. Cases will include all lung cancer deaths on death certificates identified in the follow-up period. Histologic confirmation of lung cancers identified is planned. Controls will be selected from among the cohort members who were alive prior to the day the case died. Extensive next-of-kin interviews will be conducted to obtain detailed information on lifestyle (including smoking), employment, and medical history.

Daniel Yereb described current exposure assessment of diesel emissions, including personal and area sampling methods. Mustafa Dosemeci outlined the detailed retrospective exposure assessment procedures. These procedures focus on elemental carbon, respirable combustible dust, and diesel particulate matter as primary direct surrogates of diesel exhaust. Other direct surrogates (e.g., NO_X, hydrocarbons, and total dust) and indirect surrogates (e.g., diesel equipment and fuels used, mining method, and ventilation) have been identified. The historical exposure assessment will include several steps that will identify sources of historical information on exposures, equipment, and processes, as well as standardize job profiles and incorporate monitoring data. Dosemeci indicated that this information will be used to develop exposure indices for all study subjects.

The discussion following these presentations focused on how these new studies might contribute to QRA. The proposed methods in the miners studies should be able to connect source characterization and exposure assessment better than previous efforts in epidemiologic studies. Also, there will be the possibility of a more careful evaluation of markers of diesel, both for low exposure and other sources of carbon. Although the miners studies involve exposure levels that are considerably higher than general population ambient exposures, it may be feasible to integrate exposure assessment and characterization over a broad range.

Another point was that extrapolating the results of occupational studies to the general population required too many assumptions. Because the findings from these studies presented are from male workers, heavily exposed, and likely to be smokers, shifting an exposure down to general population levels from occupational exposure levels requires a degree of caution. Also, the mining environment is different from the ambient air, and may not be directly comparable.

The identification of diesel as a probable hazard would most likely be strengthened if a dose-response relationship were seen in epidemiologic studies. In examining this relationship, more attention needs to be paid to sources of uncertainty and sensitivity in model selection. The shape of the dose-response curve will depend on a number of factors, including whether the evaluation utilizes attributable risk versus individual risk, how the extrapolation to levels in the general environment occurs, consideration of susceptible groups, and the impact of smoking. It appears that specific biomarkers for either exposure or effect, which might be useful, are not yet available.

Another perspective offered in the session discussion involved consideration of the body of epidemiologic information in order to provide a range of elevated risk. Because improvements in epidemiologic studies move us forward in small steps, an improvement might include use of incidence data, providing the occurrence of all new health events in a defined time period; this type of design would also allow for interviewing and obtaining direct information from study subjects regarding exposures. Exposures change, and the risk measured and studied in these epidemiologic studies does not reflect the exposures at the time that results are available. An important consideration in moving forward with the research in this area is determining how uncertainties will be measured and interpreted.

NON-CANCER HEALTH EFFECTS

The final session focused on non-cancer health outcomes and the research currently underway in this area. Although there is little epidemiologic research currently examining the non-cancer health effects of diesel exhaust emissions, as emphasized by Michael Lipsett in his introductory remarks to the session, interest was high concerning what in fact was being done. Studies of respiratory effects in children, inflammatory effects in human airways, and human allergic responses were presented.

Bert Brunekreff presented preliminary findings of a study of school children in the Netherlands involving air monitoring and health measurements. Extremely preliminary analyses suggest a relationship between some respiratory symptoms and truck (not car) traffic density.

When examining the inflammatory effects on human airways, Thomas Sandstrom posed the question of what in fact is causing the symptoms — is it the adsorbed organic chemicals or the particles? Research on inflamed human airways also raises the question of the possible mechanisms for asthma, allergies, and chronic obstructive lung disease, and whether there is a connection with cardiovascular disease. David Diaz-Sanchez reported that human allergic response, following exposure to a known allergen such as ragweed, plus diesel exhaust particles, produces an enormous increase in IgE in response to the challenge. IgE production for ragweed is decreased with drug pre-treatment; however, treatment with drugs does not decrease the diesel exhaust particles/allergen response. The mechanisms involved here appear to require additional research to understand.

Leading the discussion of non-cancer endpoints, Thomas Sinks presented some "guiding principles" to keep in mind in defining these endpoints. These include a need for determining a sensitive and specific outcome that is unique to diesel, a response that varies in a known pattern, an understanding of the natural history of the disease, knowledge of the incidence and prevalence of the effects, and a minimally invasive technique to detect the effects. Continuing the discussion, Frank Speizer noted that something less than total PM might be important in studying non-cancer effects — that is, PM may be a measure of "dirty air" and the proportion of diesel particulates may not matter. As with lung cancer research, there is a need to develop measures of exposure better than those currently available, perhaps finding the key component of dirty air. The final panelist, Jodi Sugerman-Brozan, thought those residents in urban hot spots, where buses and trucks idle heavily, should be studied. Especially important in such studies are children who are likely to be more sensitive. She also noted that opportunities to bring residents and scientists together were important.

In looking to new research in this area, standardized protocols and endpoints will be useful, with the inclusion of objective data. Community-based surveillance is an additionally useful component. As changes in diesel emissions and fuels occur, a "natural experiment" is introduced for looking at non-cancer endpoints and the effectiveness of interventions.

The workshop will help to influence, along with other information, future directions for HEI research under development. A description of current HEI diesel-related activities is available in HEI's Diesel Program Summary, and future plans for possible diesel research activities are discussed in HEI's Strategic Plan: 2000–2004. Both can be obtained from the HEI website at *http://www.healtheffects.org*.

SUMMARY OF RESEARCH DIRECTIONS AND WAYS TO FILL DATA GAPS

Improve understanding of health effects, including:

- biologic mechanism responsible for lung cancer development in humans
- risk to health of low-level diesel exposures
- risk to health of current emission levels
- non-cancer effects, including asthma, allergies, COPD, and diesel/allergen interactions
- health effects from after-treatments to diesel exhaust

Improve understanding of emissions, including:

- diesel exhaust constituents and changes in those constituents with new technologies over time
- changes in emission characterization from emissions out-of-the-tailpipe to inhaled
- characterization of diesel particles
- differences in emissions that result from cold starts; over different terrains; in different locations; at various altitudes, temperatures, and humidity levels; and after varying degrees of mechanical maintenance
- differences in emissions across engines and fuel types, focusing on particle changes and tradeoffs to health, ozone levels, and climate

Improve understanding of exposures, including:

- measurements for epidemiologic studies
- sources of uncertainty and sensitivity in building mathematical models of exposure
- development of biomarkers of diesel exhaust exposure, specific among PM pollutants

SESSION I

Risk Assessments of Diesel Emissions: Framework for Building a Research Strategy

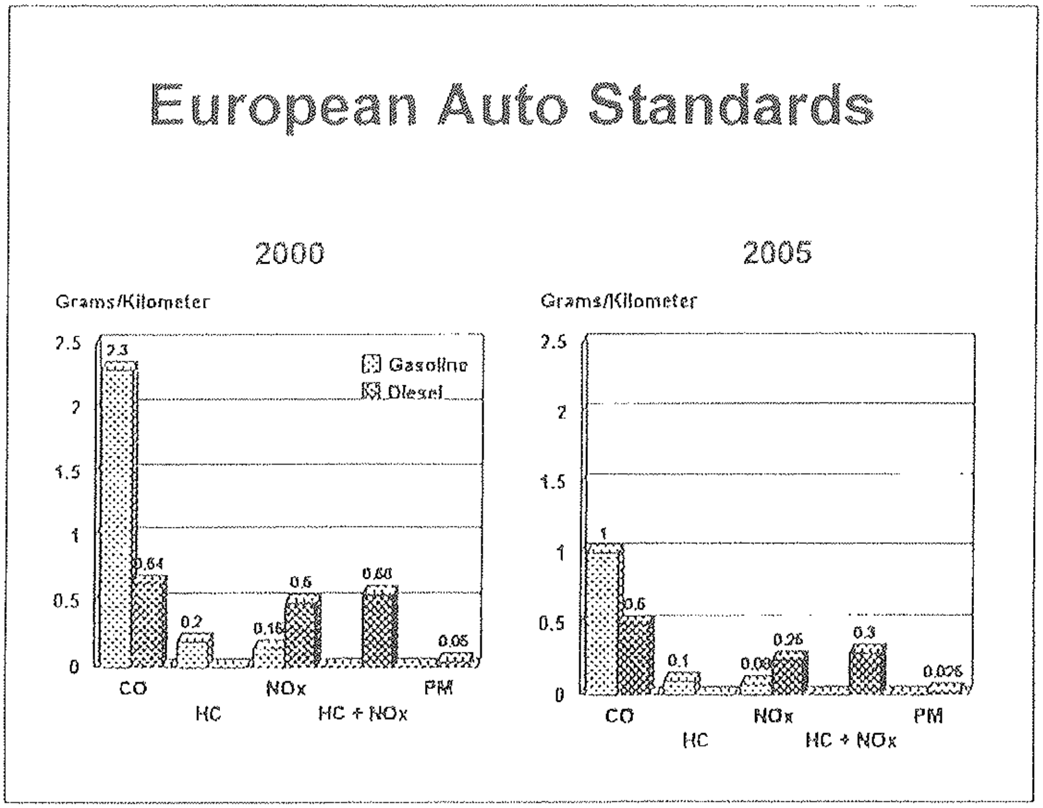
Daniel Greenbaum, Chair

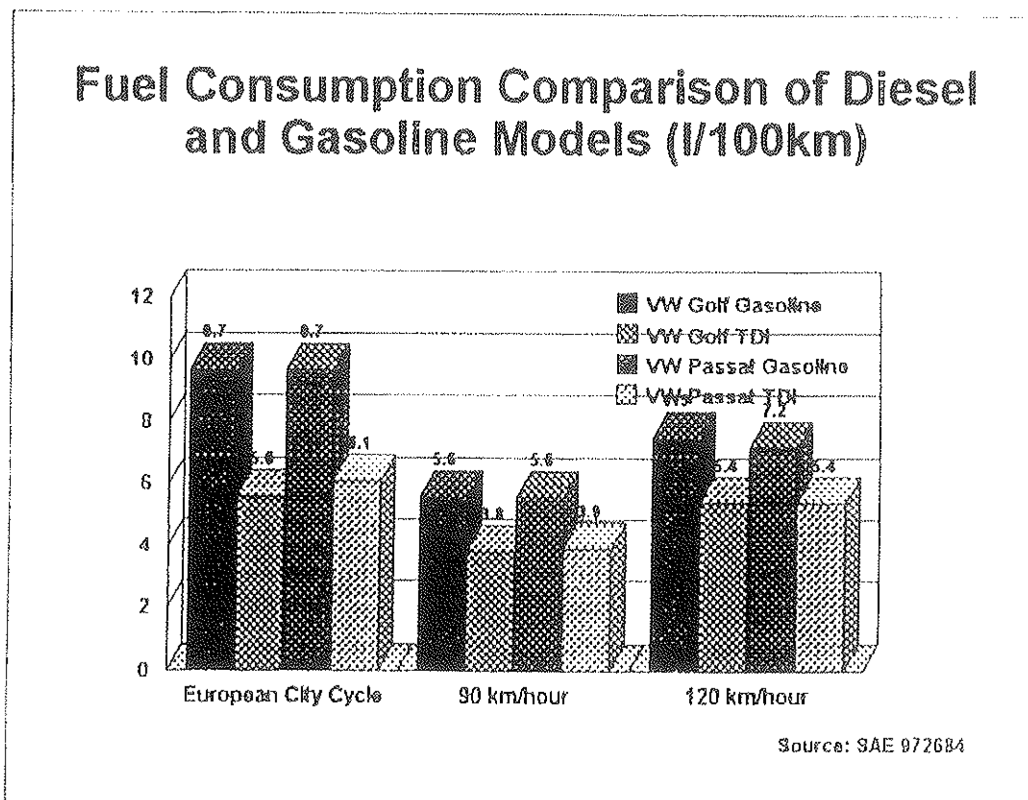
Paolo Boffetta

Vanessa Vu

Joe Mauderly

The introductory session set the stage for how HEI and others—including both government and industry—can improve risk assessments of diesel emissions. An overview of the existing toxicologic and epidemiologic data on the cancer risks of exposure to diesel emissions was presented, as well as the gaps in this information. Representatives of government, industry, and environmental groups presented their perspectives on research needs.


European Auto Standards
2000
2005
Grams/Kilometer
Gasoline
Diesel
CO
HC
NOx
HC + NOx
PM


Fuel Consumption Comparison of Diesel and Gasoline Models (l/100km)
VW Golf Gasoline
VW Golf TDI
VW Passat Gasoline
VW Passat TDI
European City Cycle
90 km/hour
120 km/hour
Source: SAE 972684

CURRENT STATE OF DIESEL RISK ASSESSMENT

- WHO International Agency for Research on Cancer (IARC - 1989 Final)
- WHO International Programme on Chemical Safety (1996 Final)
- California EPA (1998 Final)
- U.S. EPA (1998 Draft)

HEI

- IARC, 1989
  - Rat data of cancer "sufficient"
  - Human Epi data "limited"
  - Considered diesel a "probable" human carcinogen
- WHO, 1996
  - Similar to IARC
  - Epi studies considered "inadequate for a quantitative estimate of human risk"

HEI

- California EPA (1998)
    - Rat data not sufficient for quantitative estimates
    - Human epi data can be used for quantitative risk estimates(QRA)
    - "causal association of diesel exhaust and lung cancer" is a "likely and reasonable explanation"
    - CARB named diesel exhaust a *Toxic Air Contaminant* (Fall, 1998)

HEI

- U.S. EPA (Draft - 1998)
    - Rat data "sufficient" for risk estimates
    - Human epi data "limited"
    - Proposed diesel exhaust a "probable" human carcinogen
    - CASAC review late 1998
    - Risk Assessment being revised currently

HEI

- Many Agencies
  - Assessing Risk,
  - Taking Risk Management Actions (e.g. occupational protections, emissions controls)
- At the same time, A Continuing Need for Improved Science
  - To better identify degree and nature of hazard
  - To better calculate estimates of risk (QRA)

HEI

- This Workshop:
  - To inform future decisions by HEI and others on the most important research to pursue in the long and short term
  - To base future research decisions on understanding of:
    - what we know now, and the needs of the Risk Assessment Agencies
    - results of HEI Expert Panel on Epidemiology and QRA
    - ongoing epi studies, and new HEI feasibility studies
    - emerging studies of non-cancer effects

HEI

Cancer Hazard Overview

Paolo Boffetta

Overview of the epidemiology and toxicology information that supports cancer hazard identification of diesel emissions.

Paolo Boffetta, International Agency for Research on Cancer, Lyon, France

Epidemiological studies of diesel exposed workers, which are relevant to the evaluation of hazard of lung cancer, have investigated five exposure circumstances. Ten studies addressed the risk among truck drivers: the combined relative risk (RR) estimate is 1.49 (95% confidence interval [CI] 1.36-1.65); three studies investigated heavy equipment operators (combined RR 1.11, 95% CI 0.89-1.38); six studies included railroad workers (combined RR 1.44, 95% CI 1.30-1.60); five studies included bus workers (combined RR 1.24, 95% CI 0.93-1.64); finally, four studies classified workers in various occupations as exposed to diesel emissions (combined RR 1.16, 95% CI 1.01-1.32). When the 28 risk estimates are combined, the summary RR is 1.33 (95% CI 1.24-1.44). These results are reasonably consistent within and between groups of studies; evidence of an increase in lung cancer risk with duration of exposure to diesel emissions or with estimated cumulative exposure can be derived from a number of studies. Although the individual studies might suffer from bias, it is unlikely that biases explain the overall picture: fact, studies with stronger design, such as cohorts with an internal reference group, suggest a larger increase in risk than other studies, and there is no evidence of publication bias. The available evidence suggests that tobacco smoking does not act as a confounder; the possible confounding effect of other risk factors, such as diet and socio-economic status, however, has not been adequately addressed.

Inhalation studies in rats show an increased incidence of lung tumours, with a stronger effect among females. However, particles similar to those in diesel emissions, such as carbon black and titanium dioxide, exert a similar carcinogenic effect on female rats, and there is no evidence of carcinogenicity of filtered diesel emissions. The relevance of the results of the inhalation studies in rats to cancer hazard identification in humans has therefore been questioned. Data from inhalation experiments in other species are limited, and no clear evidence of carcinogenicity is available. Positive results are available from a limited number of animal studies based on lung implantation, skin painting and subcutaneous administration.

In conclusion, the human data point towards a causal association between exposure to diesel emissions and increased lung cancer hazard: a few unresolved methodological issues and the lack of clear evidence from animal inhalation studies, however, suggest caution in reaching a final conclusion.

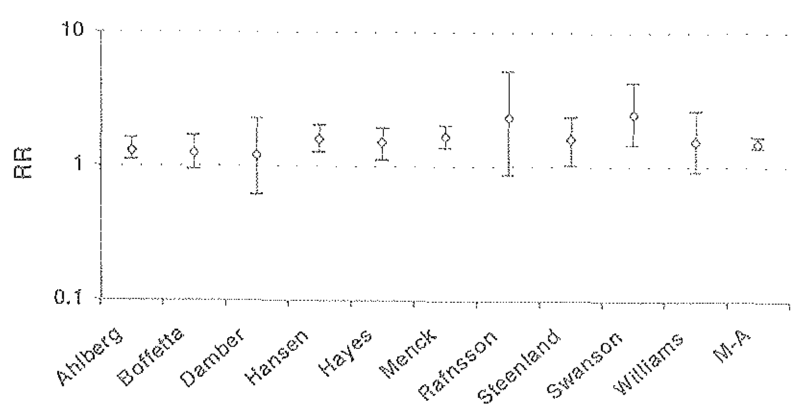
The results of epidemiological studies of bladder cancer suggest a positive association (summary RR 1.29, 95% CI 1.12-1.49, based on 21 risk estimates). However, the data suffer from three limitations: (i) the overall results depend on the inclusion of studies of truck drivers, (ii) there is no clear evidence of a dose-response relationship and (iii) the results of cohort studies do not suggest an increased risk. Although an increased hazard of bladder cancer among workers exposed to diesel emissions is plausible, the available evidence does not support a causal interpretation. Results on other cancers are limited and no conclusions can be drawn: an increased risk of kidney cancer has been reported in two studies, but the overall evidence is negative; positive results have been reported in single studies for cancers of the colon, liver, pancreas and prostate, and for malignant melanoma.

Epidemiological studies of diesel exhaust exposure

- Truck drivers
- Equipment operators
- Railroad workers
- Bus workers
- Studies based on job-exposure matrix

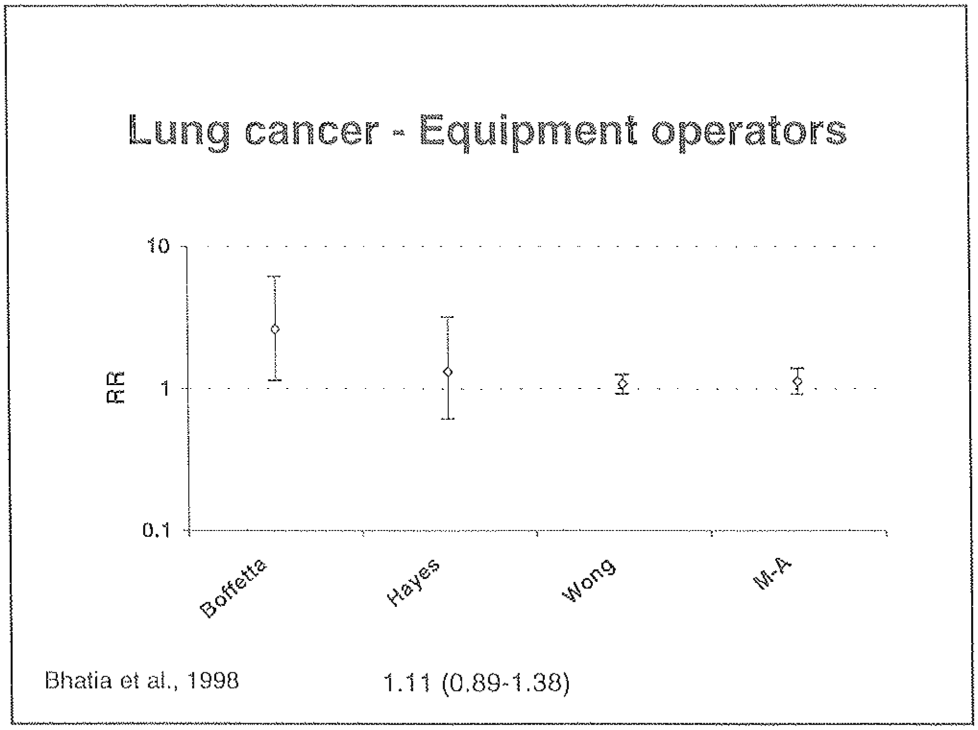
Recent reviews: Cohen & Higgins, 1995; Bhatia et al., 1998

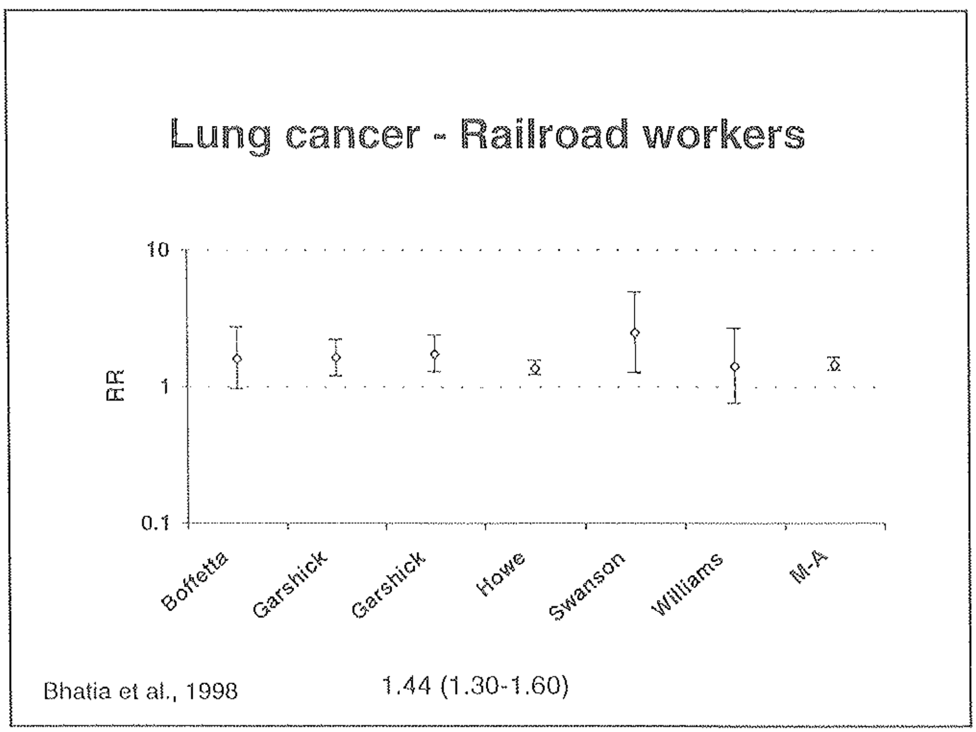
Lung cancer - Truck drivers

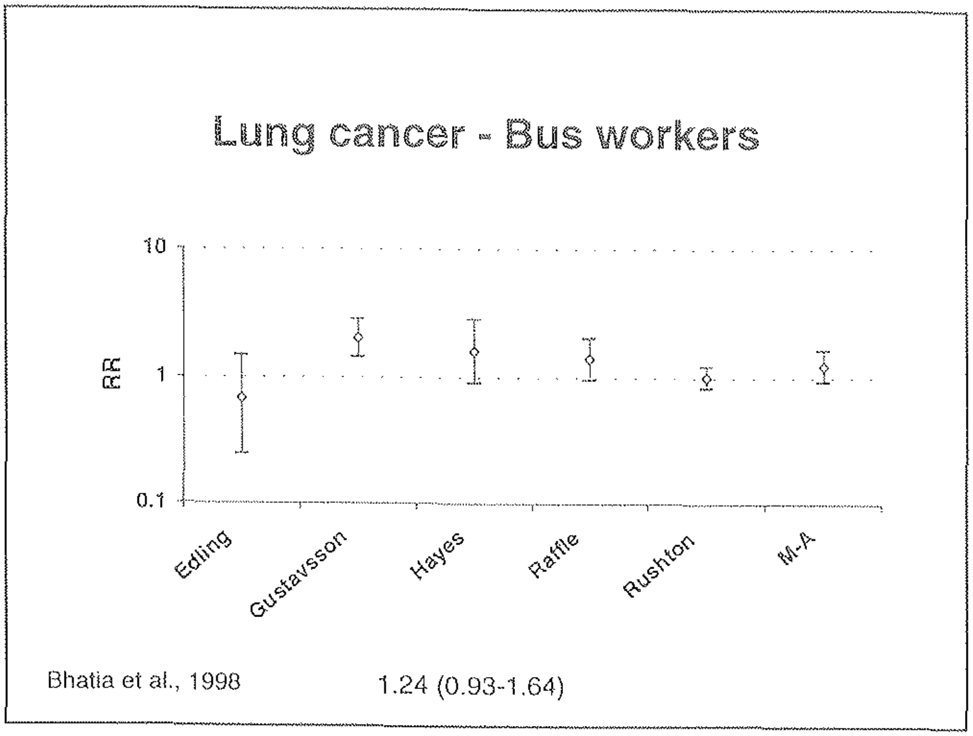


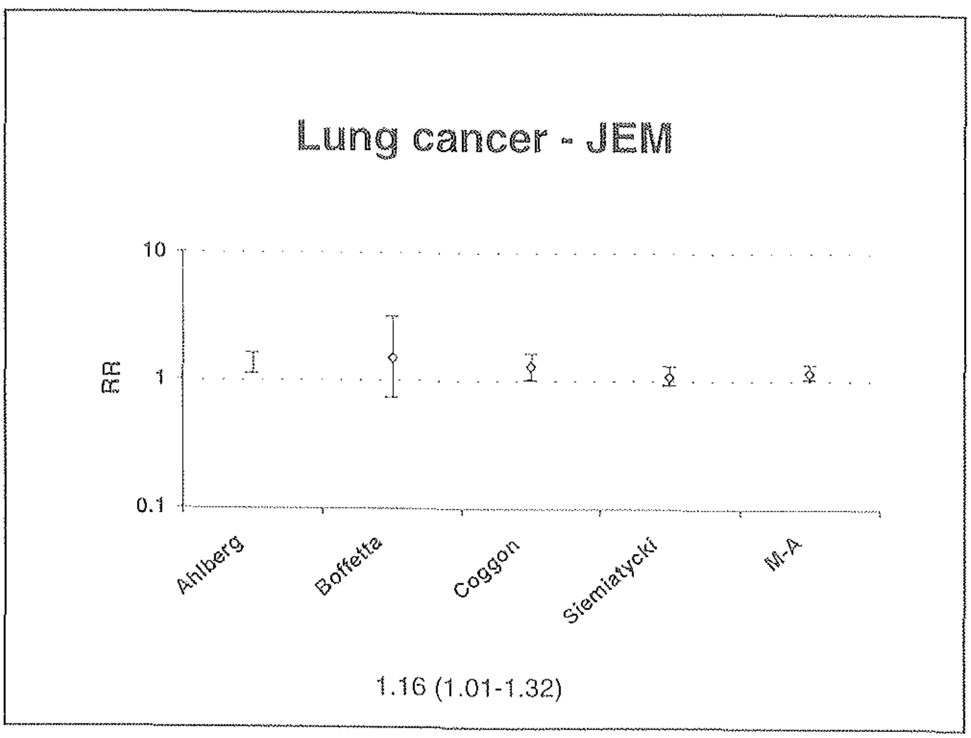
Bhatia et al., 1998

1.49 (1.36-1.65)

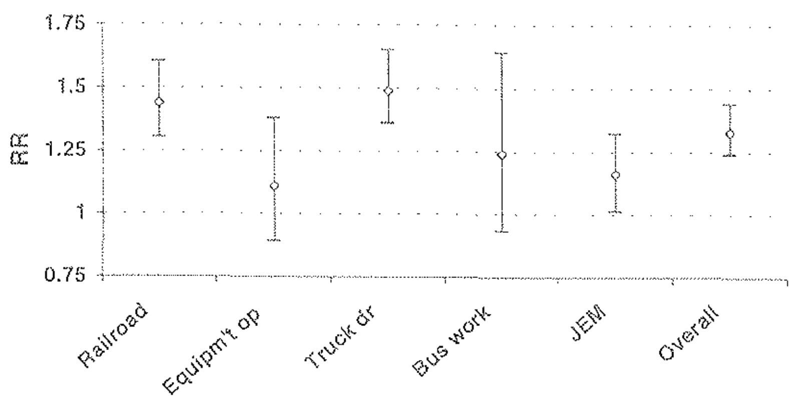

Lung cancer - Equipment operators
10
RR
1
0.1
Boffetta
Hayes
Wong
M-A
Bhatia et al., 1998
1.11 (0.89-1.38)


Lung cancer - Railroad workers
10
RR
1
0.1
Boffetta
Garshick
Garshick
Howe
Swanson
Williams
M-A
Bhatia et al., 1998
1.44 (1.30-1.60)


Lung cancer - Bus workers
10
RR
1
0.1
Edling
Gustavsson
Hayes
Rafle
Rushton
M-A
Bhatia et al., 1998
1.24 (0.93-1.64)


Lung cancer - JEM
10
RR
1
0.1
Ahlberg
Boffetta
Coggon
Siemiatycki
M-A
1.16 (1.01-1.32)

Lung cancer - Summary meta-analysis



Bhatia et al., 1998

1.33 (1.24-1.44)

Risk of lung cancer

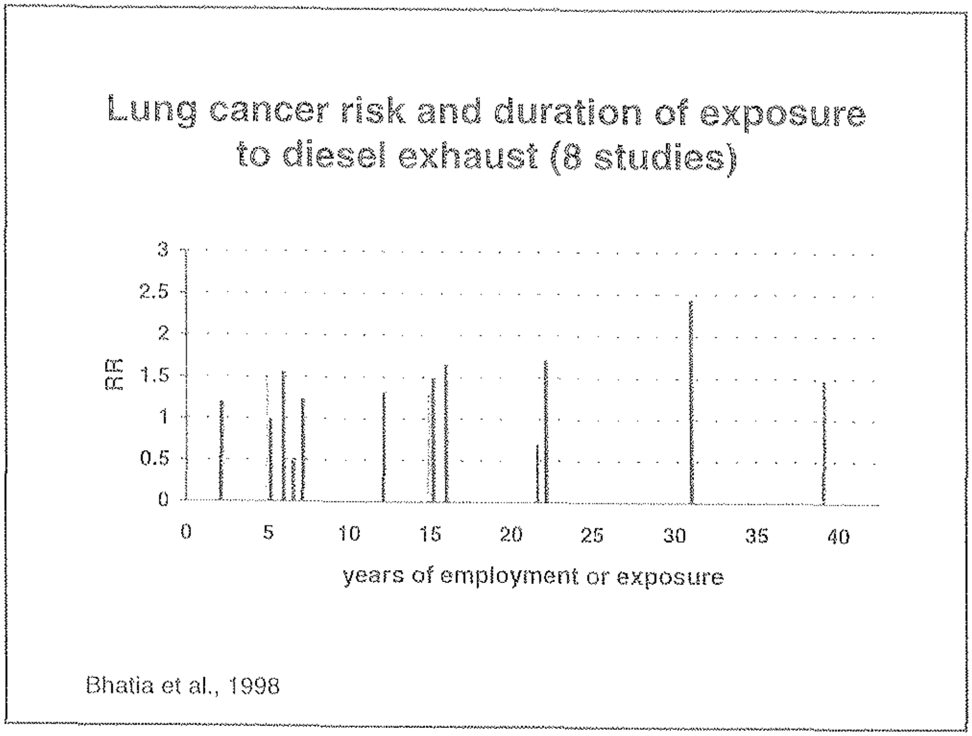
- Presence of an association
  - consistency of results
  - dose-response
  - biological plausibility
- Explanation
  - chance
  - bias
  - confounding
  - causality

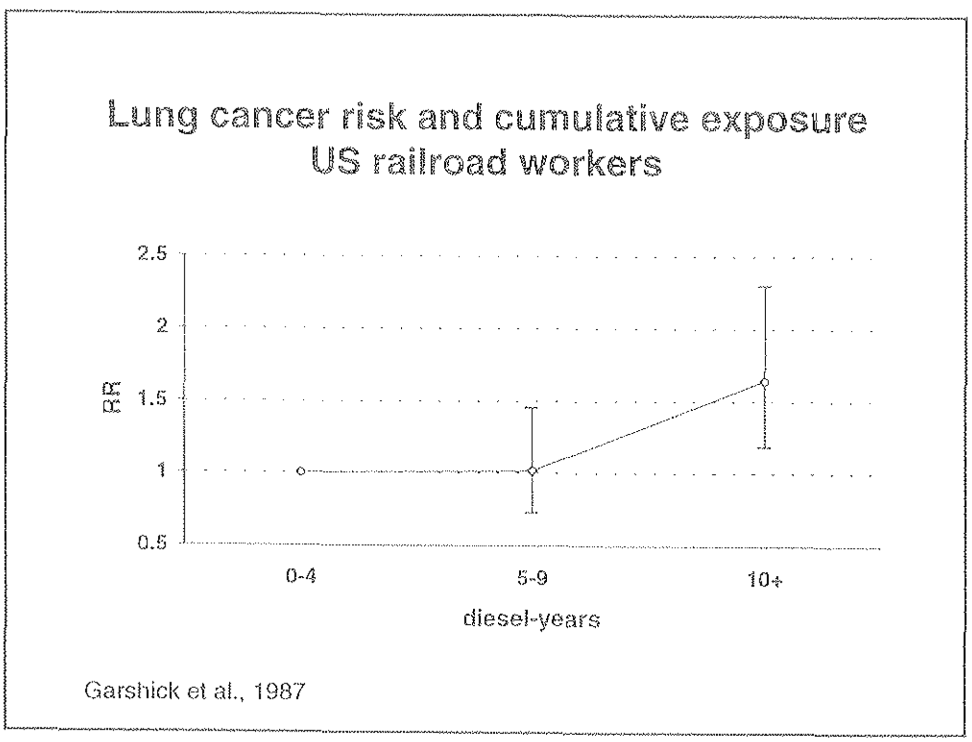
Consistency of results

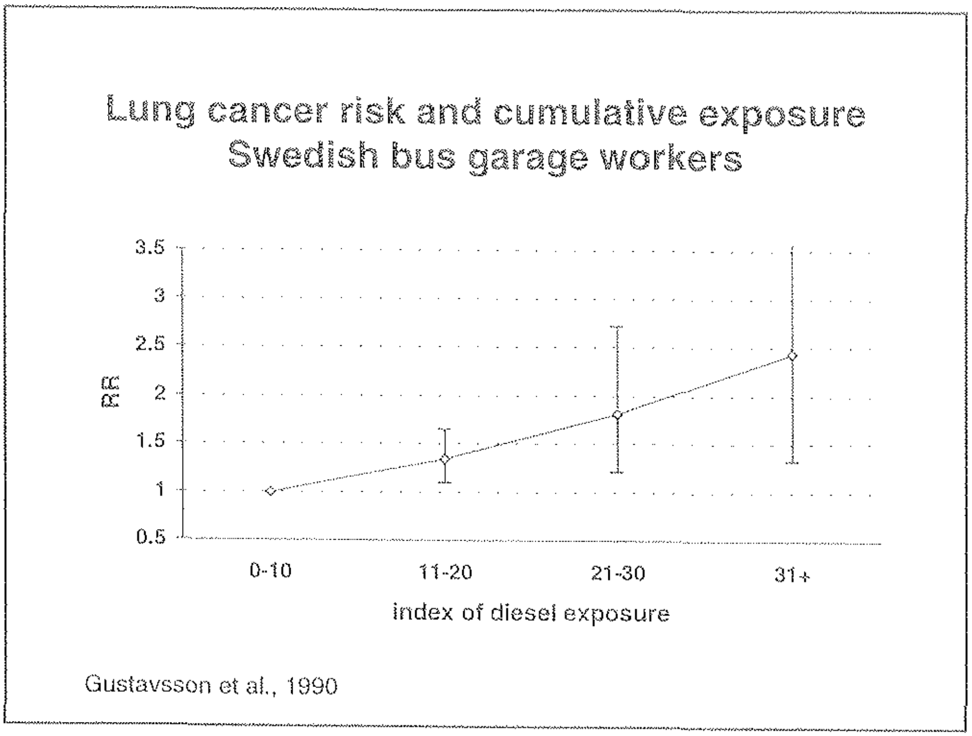
- Limited statistical evidence of heterogeneity within groups of studies
- Heterogeneity in pooled estimates among groups of studies

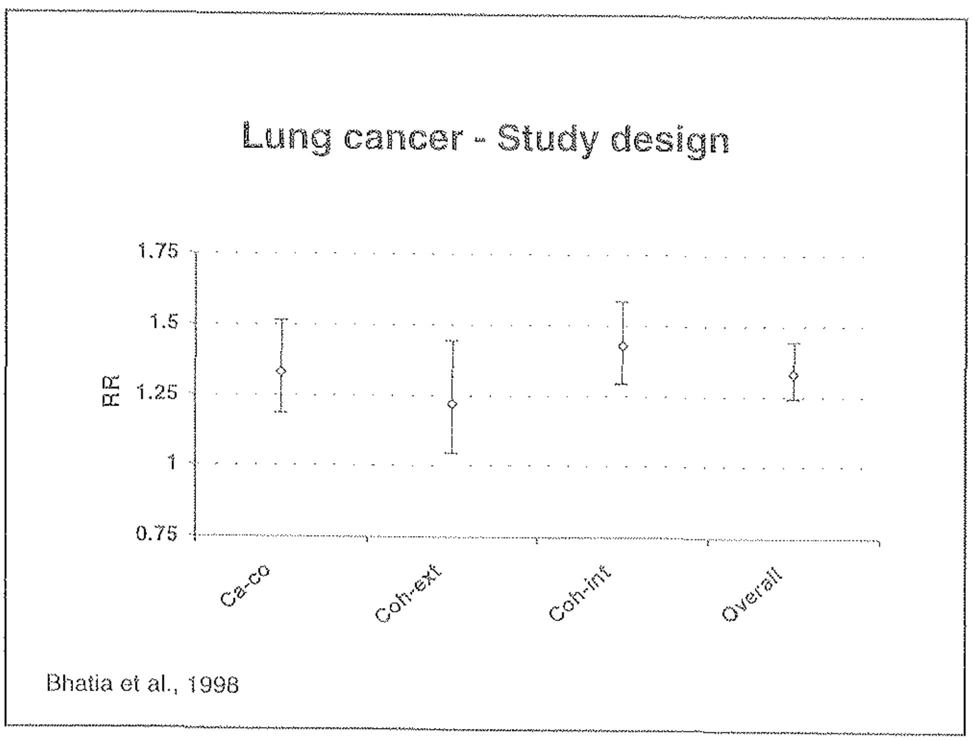
Heterogeneity of results

Bhatia et al., 1998


Lung cancer risk and duration of exposure to diesel exhaust (8 studies)
RR
3
2.5
2
1.5
1
0.5
0
0
5
10
15
20
25
30
35
40
years of employment or exposure
Bhatia et al., 1998


Lung cancer risk and cumulative exposure US railroad workers
RR
2.5
2
1.5
1
0.5
0-4
5-9
10+
diesel-years
Garshick et al., 1987


Lung cancer risk and cumulative exposure
Swedish bus garage workers
3.5
3
2.5
2
1.5
1
0.5
RR
0-10
11-20
21-30
31+
index of diesel exposure
Gustavsson et al., 1990

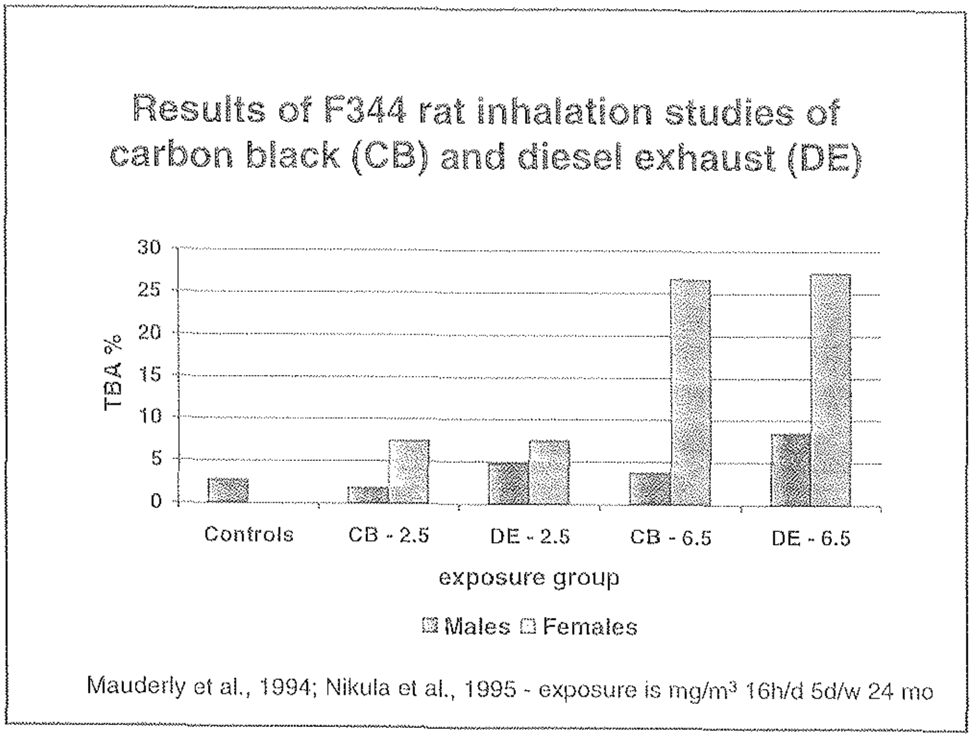

Lung cancer - Study design
1.75
1.5
1.25
1
0.75
RR
Ca-co
Coh-ext
Coh-int
Overall
Bhatia et al., 1998

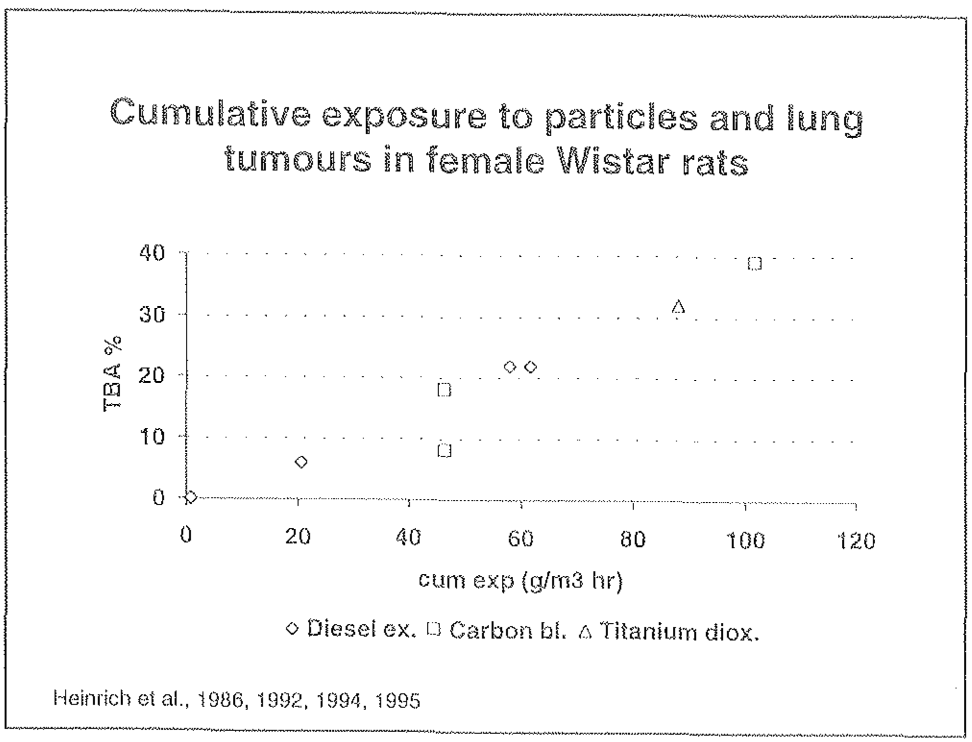
Biological plausibility

- Comparison of magnitude of risk with studies of tobacco smoke (1-2 cigarettes/day)
- Target organs
- Evidence from experimental studies

Animal carcinogenicity

- Inhalation (rats, mice, other species)
- Lung implantation
- Subcutaneous administration
- Skin painting

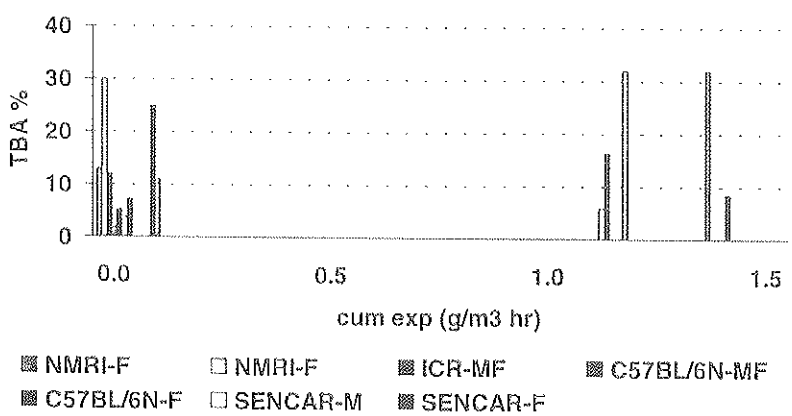

Results of F344 rat inhalation studies of carbon black (CB) and diesel exhaust (DE)
30
25
20
15
10
5
0
TBA %
Controls
CB - 2.5
DE - 2.5
CB - 6.5
DE - 6.5
exposure group
Males
Females
Mauderly et al., 1994; Nikula et al., 1995 - exposure is mg/m³ 16h/d 5d/w 24 mo


Cumulative exposure to particles and lung tumours in female Wistar rats
40
30
20
10
0
TBA %
0
20
40
60
80
100
120
cum exp (g/m3 hr)
Diesel ex.
Carbon bl.
Titanium diox.
Heinrich et al., 1986, 1992, 1994, 1995

Inhalation studies in rats conclusions

- Increased incidence of lung cancer
- Females are more sensitive
- Particles similar to diesel exhaust exhert a similar effect (in females)
- Unclear role of organic fraction
- No increased incidence of other tumours

Cumulative exposure to diesel exhaust and lung tumours in mice

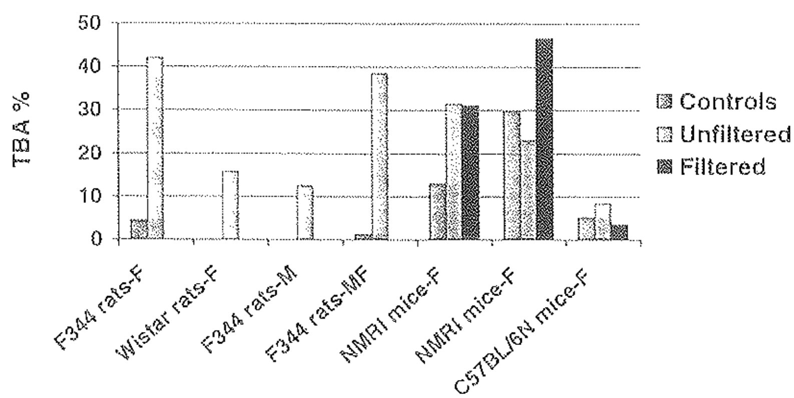


Heinrich et al., 1986, 1995; Takemoto 1986; Pepelko 1983

Inhalation studies in mice conclusions

- Weak suggestion of increased incidence of lung cancer
- Unclear role of particles vs. organic fraction

Effect of particle removal



Heinrich 1986, 1995; Ishinishi 1988; Brightwell 1989 - concentatrions 3.0-6.6 mg/m³

Lung implantation

- Excess of lung cancer in one study of female rats
- Effect due to fractions containing 4- to 7-ring PAHs and nitro-PAHs

Grimmer 1987

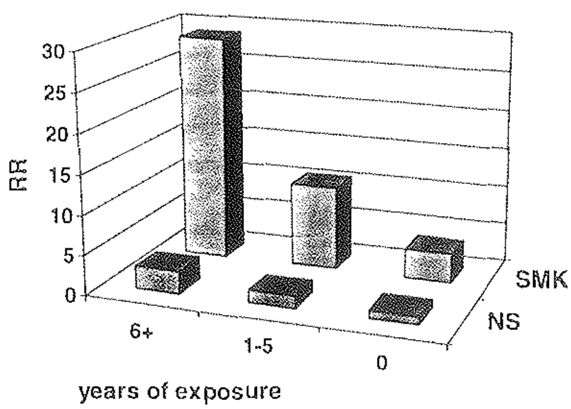
Other experimental studies

- Excess of skin tumours in studies of skin painting and tumour initiation using particle extracts
- Excess of tumours at injection site in one subcutaneous administration study

Animal carcinogenicity
Conclusions

- Excess of lung tumours in rats, of questionable relevance to humans
- No clear evidence in other species
- Positive results in other experimental settings

Interaction of diesel exposure and tobacco smoking - Swedish dock workers



Emmelin et al., 1993

Chance

- Only some of the individual studies have statistically significant results
- $p>0.05$ for meta-analyses of studies of
  - heavy equipment operators
  - bus workers
  - JEM
- $p<0.05$ for overall meta-analysis and groups
  - truck drivers
  - railroad workers

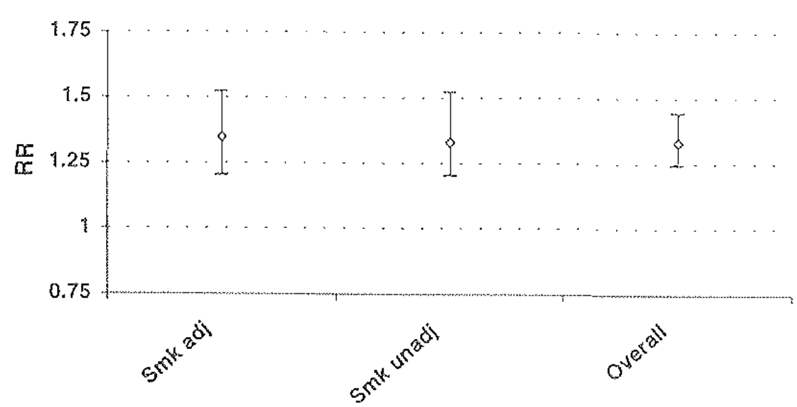
Bias

- Meta-analysis of subsets of studies argues against bias
- No evidence of publication bias

Confounding

- Heterogeneity of results among groups (truck drivers)
- Lack of evidence of confounding by tobacco smoking
- Other potential confounders
  - diet
  - other occupational exposures
  - other socio-economic factors

Lung cancer - Adjustment for smoking

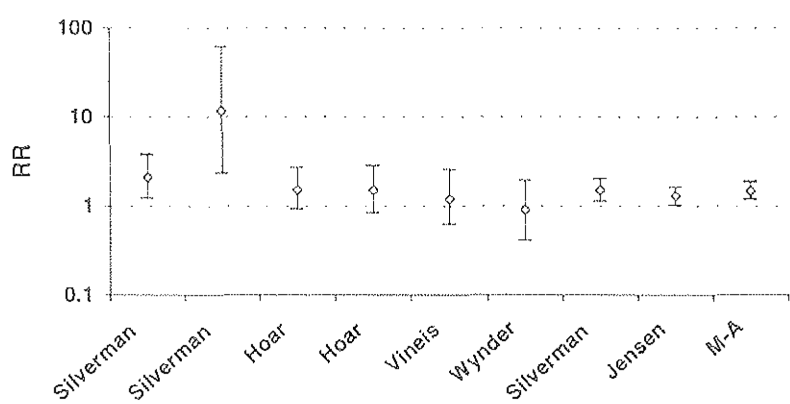


Bhatia et al., 1998

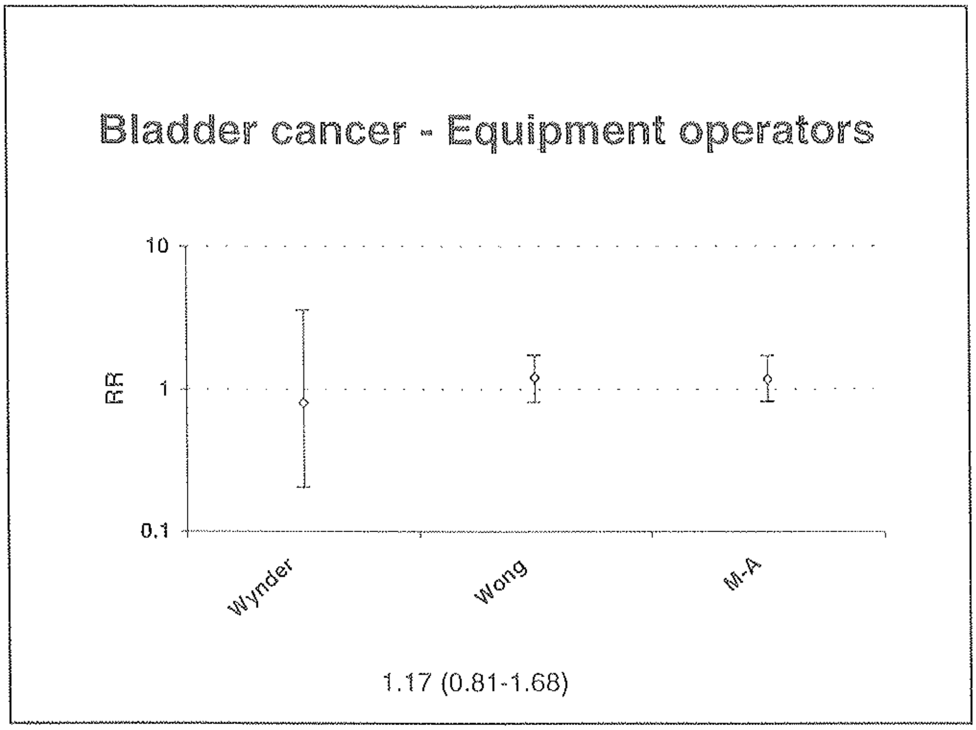
Lung cancer and diesel exhaust exposure: A causal association?

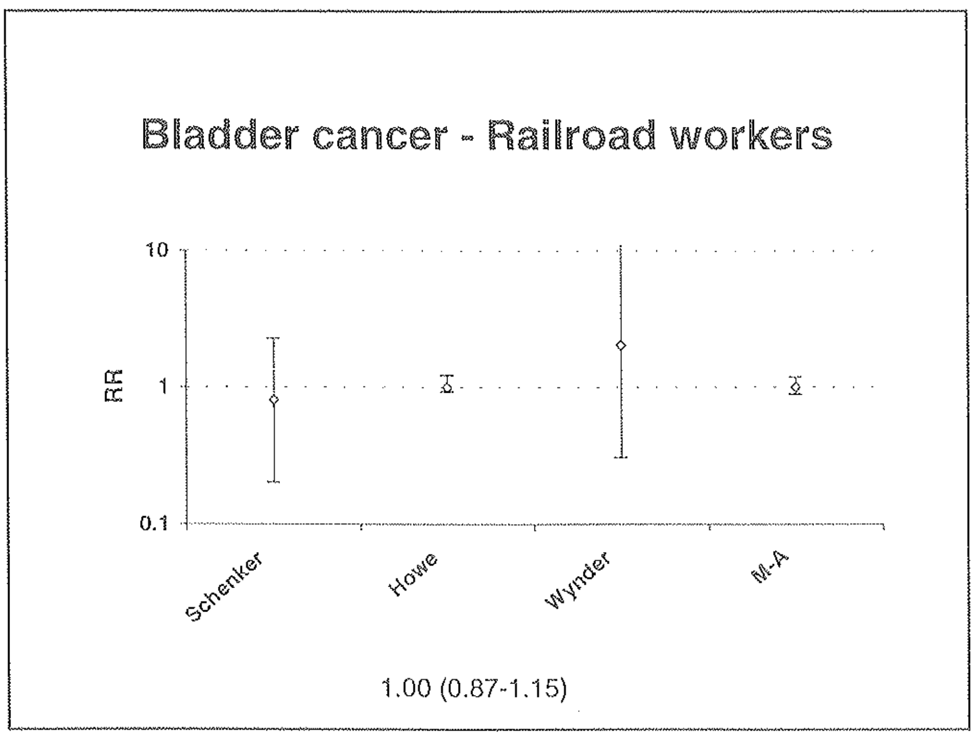
- Unresolved issues and areas for future research:
  - most data from small studies with limited exposure data
  - possible confounding factors other than tobacco smoke
  - lack of evidence from molecular epidemiological studies

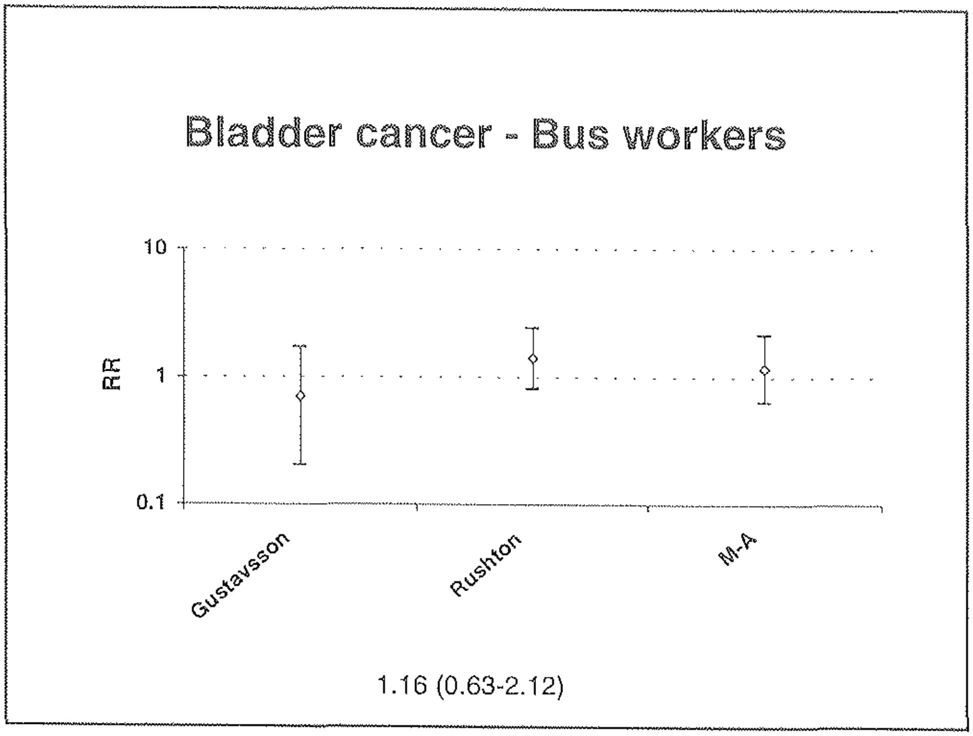
Bladder cancer - Truck drivers

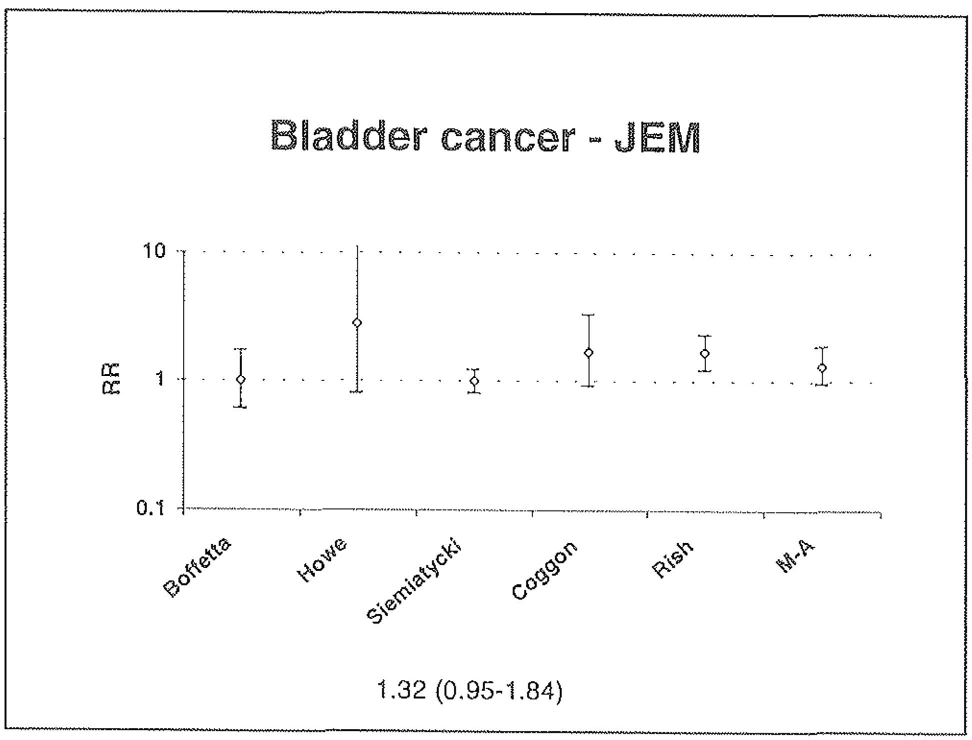


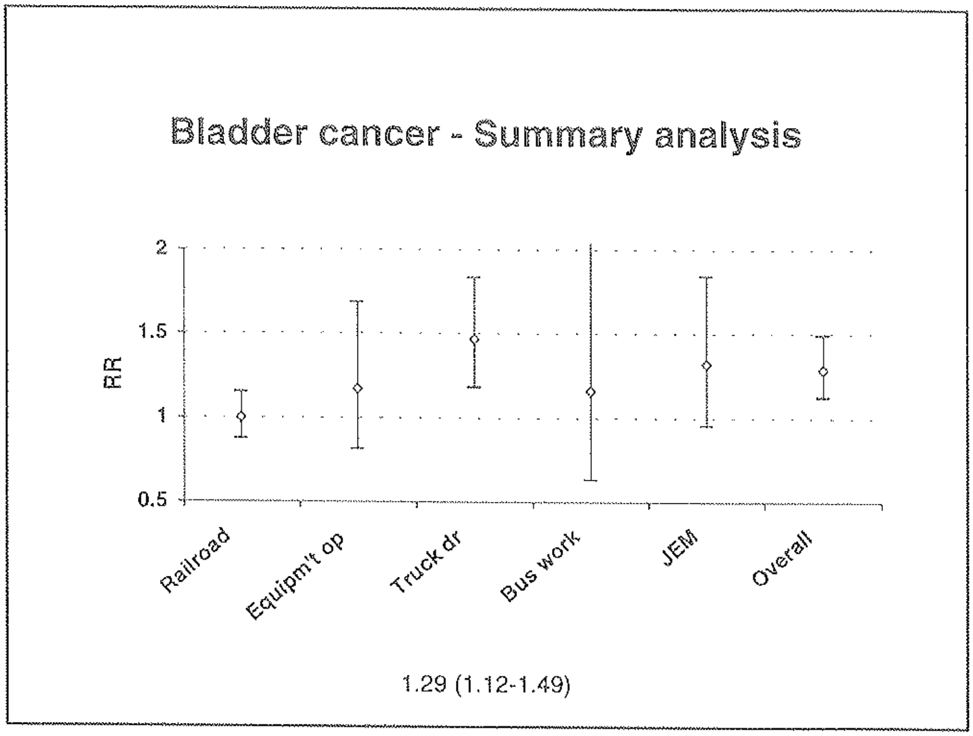
1.47 (1.18-1.83)

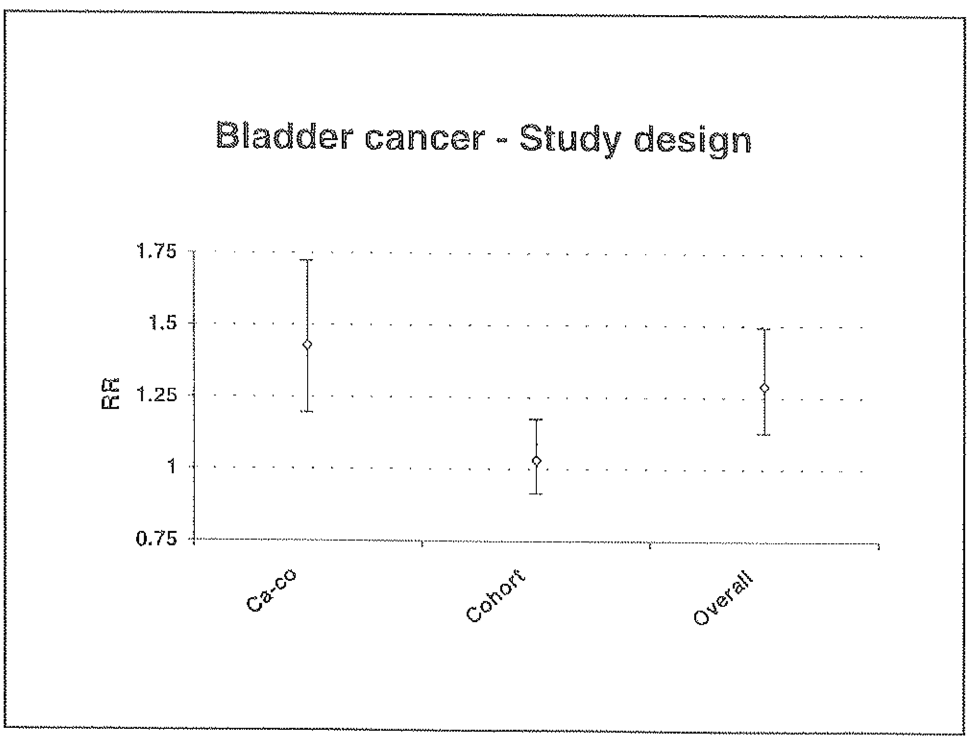

Bladder cancer - Equipment operators
10
1
0.1
RR
Wynder
Wong
M-A
1.17 (0.81-1.68)


Bladder cancer - Railroad workers
10
1
0.1
RR
Schenker
Howe
Wynder
M-A
1.00 (0.87-1.15)


Bladder cancer - Bus workers
10
RR
1
0.1
Gustavsson
Rushton
M-A
1.16 (0.63-2.12)


Bladder cancer - JEM
10
RR
1
0.1
Boffetta
Howe
Siemiatycki
Coggon
Rish
M-A
1.32 (0.95-1.84)


Bladder cancer - Summary analysis
2
1.5
RR
1
0.5
Railroad
Equipm't op
Truck dr
Bus work
JEM
Overall
1.29 (1.12-1.49)

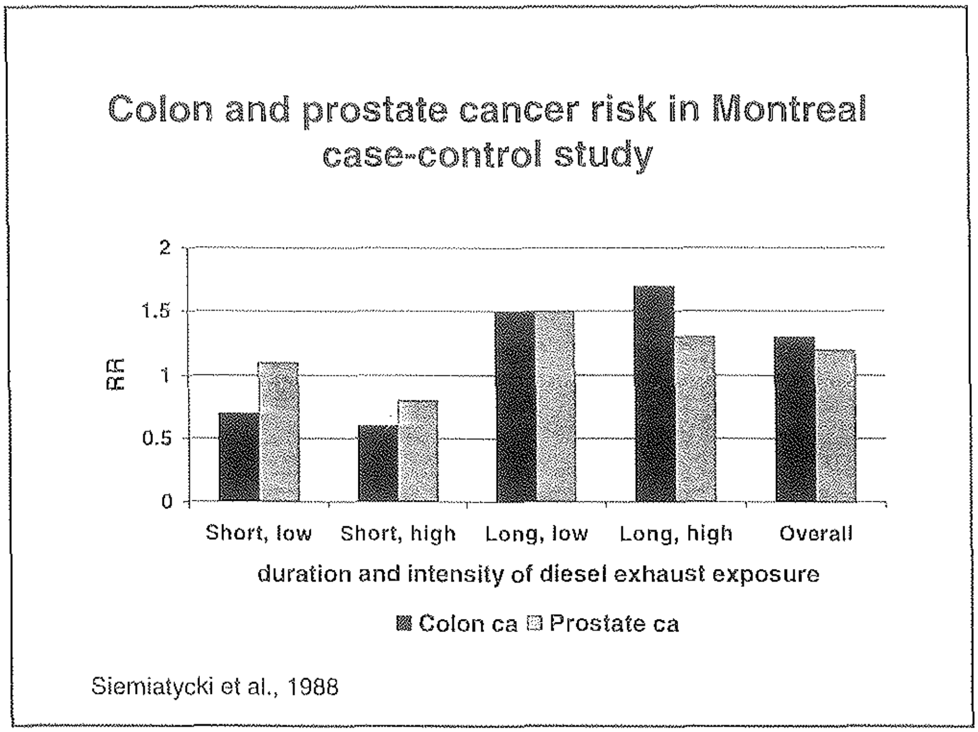

Bladder cancer - Study design
1.75
1.5
RR
1.25
1
0.75
Ca-co
Cohort
Overall

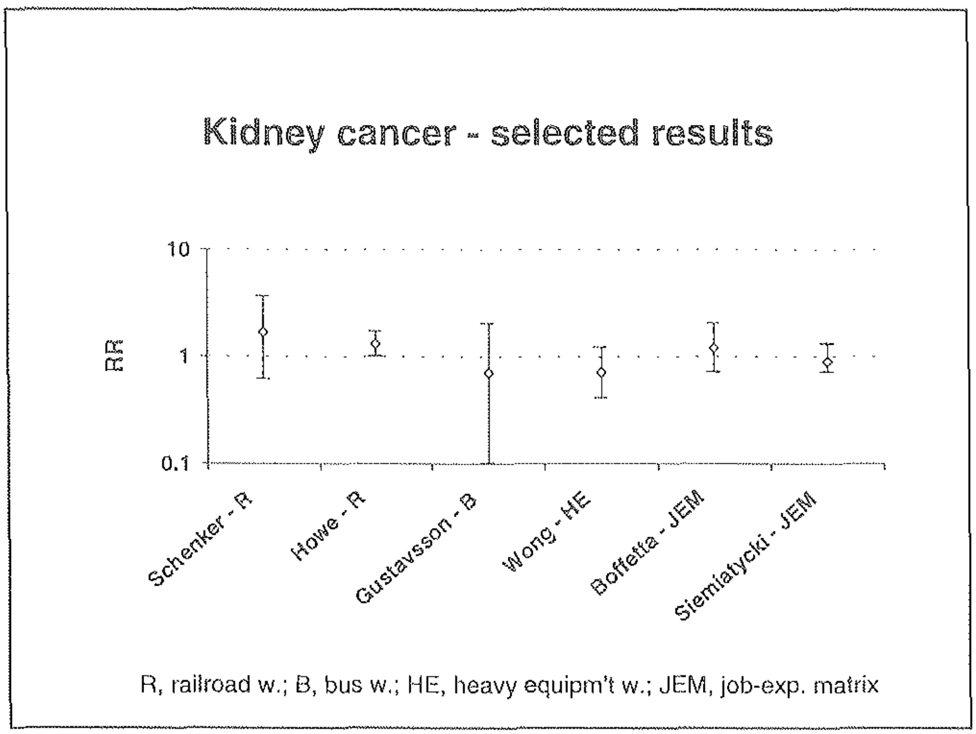
Bladder cancer - Evaluation of the evidence

- Limited database
  - statistical power of available studies
  - dose-response analyses
- Overall results dependent on inclusion of studies of truck drivers
- RR of cohort studies < case-control studies

Other cancers

- Colon cancer
  - Siemiatycki 1988
- Prostate cancer
  - Siemiatycki 1988
- Kidney cancer
- Other neoplasms reported at increased risk in one study
  - melanoma, pancreatic cancer, liver cancer


Colon and prostate cancer risk in Montreal case-control study
RR
2
1.5
1
0.5
0
Short, low
Short, high
Long, low
Long, high
Overall
duration and intensity of diesel exhaust exposure
Colon ca
Prostate ca
Siemiatycki et al., 1988


Kidney cancer - selected results
RR
10
1
0.1
Schenker - R
Howe - R
Gustavsson - B
Wong - HE
Boffetta - JEM
Siemiatycki - JEM
R, railroad w.; B, bus w.; HE, heavy equipm't w.; JEM, job-exp. matrix

Perspectives on Research Priorities

Vanessa Vu

Recommendations For Further Research To Improve The Characterization Of Health Risks From Exposures To Diesel Engine Emissions- *Vanessa T. Vu, U.S. Environmental Agency, Washington DC, USA*

Diesel exhaust (DE) consists of a complex mixture of substances formed in the combustion process of a diesel engine. The mixture includes compounds in a vapor phase and very fine particles with a carbon core coated by condensed organic substances. Many organizations have reviewed the epidemiology, toxicology, and experimental studies of DE and concluded that DE may cause acute and chronic respiratory effects, and has the potential to cause lung cancer in humans. However, considerable uncertainties remain about the suitability of existing data in conducting quantitative risk estimates for the general population exposed to current ambient levels of DE. This presentation identifies near-term and long-range research priorities to improve and reduce uncertainties in future risk assessments of DE.

Near-Term Research Needs

A major uncertainty is the representativeness of the available exposure-response data from worker population exposed to DE in the past vs. current environmental (ambient) population exposures in characterizing the possible cancer risks today. There have been some qualitative and quantitative changes in diesel emissions overtime as a result of changes in engine technology and fuel reformulation. Research is needed to

- better characterize DE among engine types (heavy duty truck, light duty, construction/off road, and locomotive/marine engines) and ages of engines, and fuel formulations including particle concentration, size, fraction of organics, concentrations of particular organic; the collected data would allow a direct comparison of effects of engine type, changing in engine technology and fuel reformulation; the characteristics of fresh DE versus aged DE should also be compared;

- obtain more complete characterization of exposure measurements in ongoing or planned occupational studies with regard to information on all components of exposure (i.e., magnitude, frequency and duration);

- further assess the potential carcinogenic effects of DE in human populations in nonoccupational setting that may have higher exposure to DE.

Available data are limited for the purpose of establishing a reliable assessment of potential health risk for chronic noncancer respiratory effects from exposures to DE. Current toxicity values (RfC) rely on dose response data from animal studies. Available studies also suggest that DE may have immunological properties that may elicit or exacerbate existing respiratory allergies.Research is needed to

- further assess the potential acute and chronic health effects in human populations exposed to DE;

- further investigate the possibility that DE either induces allergic responses, or synergises with allergens present in ambient air.

Long-Range Research Needs

The presence or absence of an exposure-dose-response threshold is another source of uncertainty. This is due to the lack of complete understanding of how DE may cause adverse health effects in exposed humans and laboratory animals. Although there are hypotheses about the specific mechanisms by which DE might cause cancer and other diseases, no specific biological pathways or specific constituents of DE have been firmly established as the responsible agents for below particle overload effects. The particle overload argument in the rat model is an important factor for high dose exposure but it does not rule out concurrent mode of action events for the organics or combined particle and organic effects. Also, the exposure-dose-response relationships for DE induced cancer and noncancer health effects have not been fully established. Research is needed to

- assess the most biologically important physical and chemical characteristics and constituents of DE, including the relative role of particulate emissions and their size and distributions, and the adsorbed organic compounds and their bioavailability in causing adverse health outcomes;

- further investigate the underlying mechanisms by which DE induces lung cancer and noncancer effects through toxicological studies (including development of appropriate animal and/or in vitro models for mechanistic investigations) and studies in humans;

- develop biologically relevant dose metrics for DE that better link exposure-dose-effect relationship, and appropriate biomarkers of exposures, effects, and susceptibility;

- differentiate the toxicologic properties of DE from ambient fine PM particle having similar particle distributions but without diesel specific organics.

In order to improve the characterization of potential human exposures to DE, further research is needed to

- measure population exposures to the most biologically important constituents of DE;

- identify human subpopulations that are potentially most susceptible to adverse health effects from DE exposures;

- assess the quantitative relationships between ambient concentrations and actual human exposures (i.e., internal dose and/or biologically effective dose) to the most toxicologically relevant DE constituents.

DIESEL ENGINE EMISSIONS

RESEARCH PRIORITIES

Vanessa T. Vu, Ph.D.
National Center for Environmental Assessment
U.S. Environmental Protection Agency

HEI DIESEL WORKSHOP
Stone Mountain, Georgia
March 7-8, 1999

OBJECTIVES

- Discuss major uncertainties in current health risk assessments of Diesel Exhaust (DE)
- Identify further research needs & priorities to improve future risk assessments

OUTLINE

- Overview of the risk assessment process
- Sources of uncertainty in current DE risk assessments
- Research strategy to improve future risk assessments

Exposure-Dose-Response Relationships

Emission Source → Fate & Transport → Environmental Concentration → Human Exposure → Internal Dose → Target Tissue Dose →

→ Biologically Effective Dose → Molecular & Cellular Interactions → Early Biological Effect → Altered Structure/ Function → →Disease

Risk Assessment Process

- Identify and characterize health effects of concern
- Mode of action (MOA) hypotheses for extrapolation options
- Measures of tissue dose; perform dose-response evaluation
- Identify exposed human populations; estimate human exposures
- Nature and extent of risks; confidence & major uncertainties

Hazard Identification & Characterization

- *Questions*
  - Is the environmental agent capable of causing an adverse effect in humans? If so, under what exposure conditions?
  - How does the agent produce its effect? Are there mechanistic data to support this hypothesis? Other mechanistic hypotheses?
- *Default Assumptions*
  - Effects found in an exposed human population are predictive of other populations including sensitive subpopulations
  - Effects in animals indicate potential effects in humans

Exposure-Dose-Response Assessment

- *Questions*
  - What are the quantitative relationships between exposure and biologically effective dose and adverse effects in animals? In humans?
- *Two Step Process*
  - Model data in range of observation
  - Evaluation in range of human exposure of interest
- *Default Procedures*
  - Data on most sensitive species to be used in D/R assessment
  - Nonlinear dose-response curve or existence of a threshold for noncancer health effects
  - Linear extrapolation for carcinogenic effects

Exposure Assessment

- *Questions*
  - What environmental exposures occur or are expected to occur for human populations?
  - What is the resulting dose to the target tissue?
- *Process*
  - Characterize emissions, environmental transport and fate
  - Estimate human exposure and dose

Diesel Exhaust: Hazard Characterization
Major Findings & Uncertainties

- Carcinogenic potential in humans
  - *Epidemiologic studies showing increases in the risk of lung cancer under occupational exposure settings*
  - *Chronic exposures to high concentrations of DE produce lung tumors in rats*
  - *DE constituents (whole particle, particle extracts, and gaseous fractions) cause changes in genetic materials*
  - *Whether DE can cause carcinogenic effects at low exposures remains to be determined*
- Acute and chronic respiratory effects
  - *suggestive evidence in exposed workers; contribution from ambient fine PM not known; extensive evidence in animals*
- Possible immunological effects
- Knowledge gaps about mode(s) of action
  - *Role of DE constituents not firmly established*
  - *Several mechanistic hypotheses (e.g., particle specific mechanisms, gases, adsorbed organics on the particles)*

Diesel Exhaust
Exposure-Dose-Response Assessment: Major Uncertainties

Carcinogenic Effects

- **Model in the range of observation**
  - *Exposure-response data in occupational studies-* **actual historical exposure measurements not available; mass of DE particulate as surrogate for exposure of dose**
  - *Rat lung tumor data may not be predictive of human response-* **effects appear to be due to "non-specific" particle effects at high exposures**
- **Extrapolation to ambient exposures**
  - *Low dose linearity is assumed-* **lack of good understanding of MOA, inference of mutagenicity/genotoxicity of DE constituents**
  - *Extrapolation of occupational data to ambient exposure-* **qualitative and quantitative changes in DE overtime (change in engine technology, fuel reformulation)**

Diesel Exhaust
Exposure-Dose-Response Assessment: Major Uncertainties

Noncancer Health Effects

- **Lack of exposure-response data for chronic respiratory effects in exposed workers**
- **Toxicity value (RfC) based on rat data**
  - **Rats may be more biologically sensitive to DE particles than humans**
  - **Possible effects due to other DE constituents not considered**

Diesel Exhaust
Human Exposure-Dose Assessment: Major Uncertainties

- **Ambient levels of DE particulate may not be the most appropriate dosimeter for human exposures**
  - *many DE constituents may have biological and toxicological properties*
  - *lack of understanding of the quantitative relationships between ambient concentrations and actual human exposures (i.e., internal dose) to DE constituents*

- **Accuracy of estimates of human exposures to DE constituents**
  - *variations due to many factors (e.g., diesel vehicles, engine types and age, emissions control, fuel quality, traffic flow, location of individual relative to emission sources)*
  - *DE constituents from other sources (e.g. gasoline emissions, cigarette smoke)*

- **Knowledge gaps about susceptible subpopulations- personal and environmental factors that affect DE susceptibility unknown**

Research Strategy
Near-Term Research Needs

- **Improve exposure-dose-response assessment in the range of observation (cancer risks)**
  - *more reliable exposure measurements in ongoing and planned occupational studies- complete characterization of exposure measurements (e.g. magnitude, frequency, duration of exposure)*
  - *new prospective studies to evaluate potential cancer risks in non-occupational setting that may have higher exposure to DE*

- **Improve scientific bases for the use of D/R data from worker populations exposed to DE in the past to estimate cancer risks from exposures to current environmental population exposures**
  - *complete characterization of DE among engine types (heavy duty truck, light duty, construction, locomotive engines) & ages, fuel formulations*
  - *particle concentration and size, organic fraction, concentration of specific organic substances; compare fresh DE and age DE*

Research Strategy

Near-Term Research Needs (continued)

- **Further evaluate other health effects of DE**
  - acute and chronic respiratory effects in human populations and sensitive subpopulations
  - immunological effects- possible allergic responses or synergistic effects with allergens in ambient air

Research Strategy

Long-Range Research Needs

- *Issues*
  - Scientific uncertainties associated with linear low dose extrapolation for cancer risks as default procedure
  - Shape of dose-response curve at low dose
  - Presence or absence of threshold effects

- *Proposed Research*
  - Assess biologically important physical and chemical DE constituents in causing adverse health effects- *relative role of DE particulate and particle size, adsorbed organic compounds and their bioavailability in causing adverse health effects*
  - Elucidate underlying mechanisms of DE induced carcinogenicity and toxicity- *development of animal and/or in vitro models for mechanistic investigations*

Research Strategy
Long-Range Research Needs (continued)

- **Better characterize human exposures to DE and strengthen human exposure-dose-effect relationships**
  - **Develop biologically relevant dose metrics for DE-** *biomarkers of exposures, effects, and susceptibility*
  - **Measure population exposures to the most biologically important constituents of DE**
  - **Identify susceptible human populations**
  - **Assess the quantitative relationships between ambient concentrations and actual human exposures (i.e., biologically effective dose) to the most biologically relevant DE constituents**

Uncertainties Concerning Environmental Lung Cancer Risk
Joe Mauderly

UNCERTAINTIES CONCERNING THE ENVIRONMENTAL LUNG CANCER RISK FROM INHALED DIESEL ENGINE EMISSIONS

Joe L. Mauderly

Lovelace Respiratory Research Institute, Albuquerque, NM

The lung cancer risk from environmental exposure to inhaled diesel engine emissions is a matter of considerable uncertainty and debate. A risk is plausible because diesel soot contains known carcinogenic chemicals and is ubiquitous in the environment. Retrospective epidemiology of workers suggests that jobs presumed to have high diesel exposures were associated with small increases in cancer risks, and high-dose laboratory assays confirm the cancer potential of the material. On the other hand, estimating the magnitude of cancer risk is problematic and can not be done with high confidence. Neither the actual exposures nor the shape of the exposure-response function are known for the occupational groups. Lower-level environmental exposures are not well-known, and it is not clear if an exposure threshold for cancer risk might exist. Laboratory studies clearly demonstrate increased risk among heavily-exposed rats, but not other species, and strong evidence indicates that the rat's lung response to heavy particle exposure should not be extrapolated quantitatively to predict human risk.

In May, 1998, a Clean Air Scientific Advisory Committee panel reviewed EPA's latest draft diesel health assessment document and determined that the draft was not an acceptable summary of current knowledge. That conclusion was based on several perceived inadequacies in the document, including needs for updating information, use of rat data for quantitative cancer risk estimates, lack of portrayal of diesel soot health risks as an integral part of ambient particulate matter health risks, and inadequate discussion of the occupational exposure-response issue. The panel's conclusion was not based on disagreement with the values estimated for cancer risk, but neither was there agreement with the values presented in the draft. There was mixed opinion among the panel about whether or not a useful quantitative estimate of risk could, in fact, be developed at all, and if so, which of several approaches should be used. The majority of panel members favoring a quantitative estimate preferred using occupational epidemiological data as a starting point.

Placing the likely health risks from diesel engine emissions in their proper perspective is a very important issue with large societal stakes. It is important that we neither grossly overestimate nor grossly underestimate the risk, and it is also important to place the risk in perspective among the risks from other man-made and natural air pollutants. Three considerations regarding risk have been given inadequate attention.

First, we need to estimate future risk, not past risk. Diesel emissions are changing, and our present data were derived from exposures that do not reflect future emissions. Old diesel engines are still in use, but emissions are evolving. Compression ignition technology will be important for future internal combustion engines of all sizes; however, those engines and their emissions will hardly resemble past engines and emissions.

While many of the chemical species of concern continue to be found in contemporary emissions, the amount emitted per unit of work is falling. Presumably, risk would be lowered in parallel to shrinking emissions. From on the trajectory of current regulations and technological developments, one might assume that the "soot" which formed the black diesel smoke of the past will not be a concern at all for new engines very far in the future. It is not inconceivable that particulate emissions might be virtually eliminated from compression ignition engines in the future. To the extent that particulate emissions remain, they are more likely to be ultrafine condensed droplets of organic matter, perhaps on nuclei of other materials.

Second, health risks from particulate diesel emissions need to be viewed as a subset of the total risk from environmental particulate matter (PM), rather than as a separate risk. It should, but often doesn't, go without saying that the cancer burden from environmental diesel particles can not be greater than the burden from fine PM. There is little, if anything, unique about diesel soot or other diesel emissions; the same chemicals are emitted from other sources. The population has been exposed to diesel emissions for a long time; thus, any cancer burden from environmental diesel particles must be contained within (and can not exceed) the burden from environmental PM. Beginning with the environmental PM cancer burden would seem to be a more logical starting point for estimating environmental diesel-related cancer risk than would cantilevering exposure-response slopes downward from uncertain, but much higher, occupational exposures. As we continue to improve our understanding of exposure-response relationships for environmental PM and cancer, we need to remember that the truth about risks from environmental diesel particles must lie within those relationships, not outside them.

Third, it seems that more effort might be expended exploring the possibility of working from the bottom up rather than the top down in framing the magnitude of cancer risk from diesel emissions. Most effort has been directed toward projecting exposure-risk slopes downward from occupational cancer incidence data (ie, "top down"). Both the actual exposures and the exposure-response slopes of the occupational data continue to be argued hotly, which affirms their large uncertainty. This uncertainty provides a very unsteady fulcrum for the very long dose-response "lever" necessary to reach the low environmental soot levels, such that small changes in slope at the top markedly influence the level of risk at the bottom. A couple of first-order approaches can be suggested to frame risk from the other ("bottom up") perspective. First, what is the likely maximum cancer burden attributable to environmental diesel soot? That is, if we deduct the portions thought to be due to active and passive smoking, radon, and other pollutants and factors, how large could the soot-induced cancer burden be? Of course, estimates for all causes contain uncertainties, but to the extent we understand other lung cancer risks, the residual should form an upper bound for risk from diesel soot. Second, the portion of the total, or inhaled, "dose" of carcinogens encountered in the environment attributable to diesel soot would frame an estimate of the portion of cancer risk attributable to soot. If risk is incurred by soot-borne organic mutagens and carcinogens, it is relatively straightforward to estimate the lifetime dose. Comparing that to doses from other sources would place the relative risk from diesel soot in a useful perspective.

UNCERTAINTIES CONCERNING THE ENVIRONMENTAL LUNG CANCER RISK FROM INHALED DIESEL ENGINE EMISSIONS

Joe L. Mauderly

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TOPICS

Plausibility & problems

What CASAC really said

The issue is future risk, not past risk

The environmental diesel soot issue is a subset of environmental PM issue

Can we estimate risk from the bottom up instead of the top down?

A LUNG CANCER RISK IS PLAUSIBILE

Diesel exhaust contains traces of known mutagens and carcinogens, and is ubiquitous

Environmental particulate matter is associated with lung cancer risk, and diesel soot is a subclass

Occupational epidemiology suggests that jobs presumed to have high exposures were associated with increased risk

High-dose bioassays confirm cancer potential of soot-borne Organic fraction

UNDERSTANDING THE SIZE OF THE RISK IS PROBLEMATIC

Occupational epidemiology:

Exposures?
Exposure-response curve?

Environmental epidemiology:

Exposures?
Are there enough cancers to go around?

Laboratory data:

We know the rat risk well, but so what?
Other data provide comparative potency

CASAC – WHAT DID THEY SAY?

(EPA-SAB-CASAC-99-01, October, 1998)

"The February 1998 draft is not an acceptable summary of current knowledge ---"

1. Information in some sections was not current; especially section describing diesel emissions.
2. Quantitative estimates of human environmental cancer risk from rat data are not warranted.
3. Failed to acknowledge PM database and likely relationship of diesel soot risk to PM risk.
4. Failed to address occupational dose-response dilemma.
5. Treated all risk estimates as of equal validity and weight.

CASAC PANEL OPINION ON CANCER RISK ESTIMATION

(N = 13)

	Y	N	A
Include some form of quantitative estimate?	8	3	2
If so, include some form of estimate from:			
Epidemiology?	8	3	2
Animal data?	5	8	
B(a)P?	4	4	5
Comparative potency?	7	6	

IT AIN'T YOUR DADDY'S DIESEL!

Compression ignition is not a sin!

But having unprotected ignition may be.

Engines and fuels have changed – and they are still changing.

"Thumbprints" are one thing, but how big is the thumb?

Future risk is not about "soot" as we've known and loved it.

What will we do when CI and SI look the same?

Ultrafines have always been there, and may or may not be important.

Are they solid or droplets?

We don't have laboratory health data on "new" emissions.

But as soon as somebody tells us what it is, we'll study the stuff!

CE-CERT STUDY

(U.C. Riverside, 1998)

Cummins L10-310 engine (6 cyl, turbo, 4-stroke, DI)

Heavy-Duty Transient cycle

Three fuels + only variable

	Pre-'93	Reform.	Low-A
Sulfur (ppm)	349.7	71.3	<0.1
Nitrogen (ppm)	352	205	<1.0
Aromatics (wt%)	31.0	22.3	4.3
PAH (wt%)	5.9	4.1	0.7

CE-CERT STUDY – RESULTS

(Hot Start)

		Pre-'93	Reform.	Low-A
PM	g/Bhp-hr	.218	.182 (-17)	.181 (-17)
NOx	"	4.74	4.61 (-3)	4.41 (-7)
THC	"	0.52	0.50 (-4)	0.48 (-8)
PAH (27)	mg/Bhp-hr	1.56	0.98 (-37)	0.75 (-52)
NitroPAH (5)	µg/Bhp-hr	4.85	4.40 (-9)	4.04 (-17)
Mutagenicity	Rev/Bhp-hrx10^6	7.4	4.7 (-36)	3.8 (-49)

THE BODY COUNT

FROM ENVIRONMENTAL DIESEL PARTICLES

CAN'T BE LARGER THAN

THE BODY COUNT FROM ENVIRONMENTAL PM

ASSOCIATION BETWEEN AIR POLLUTION AND LUNG CANCER IN THE SIX CITIES STUDY

(Dockery et al. NEJM 329:1754, 1993)

8096 adults, 1429 deaths after 14-16 yr follow-up

Adjusted (age, gender, smoking, education, body mass) mortality rate ratio for most vs least polluted city:

All causes	1.26 (1.08-1.47)
Non-cancer cardiopulmonary	1.37 (1.11-1.68)
Lung Cancer	1.37 (0.81-2.31)

ASSOCIATION BETWEEN LUNG CANCER AND PM IN THE AHSMOG STUDY

(Beeson et al., EHP 106:813, 1998)

California Adventist males, '77-'92, 2278 deaths, 16 lung cancer cases

PM_{10} In Excess Of $\mu g/m^3$	Interquartile Increment (days/yr)	Relative Risk	Lower C.I.
40	139	4.50	1.31
50	149	4.96	1.54
60	132	4.72	1.69
80	78	3.43	1.71
100	43	2.95	1.71

WORKING FROM THE BOTTOM UP

How Big Could The Risk Be?

(Hint: it's not 1 per 250 people)

If 90% of lung cancers are due to active smoking, some are due to radon, some are due to ETS, some are due to other pollutants, and some to other causes - *how many could be due to diesel soot?*

If lung cancers are due to soot-borne organic species, *what portion of the total intake* of such material is from diesel soot?

LIFETIME DOSE OF PAH FROM ENVIRONMENTAL EXPOSURES

Assume: Breathe 15 LPM for 80 yrs
Average soot concentration = 3 $\mu g/m^3$
Soot = 50% organic
Organics = 10% PAH
20% lung deposition, 100% bioavailable

Lifetime dose of PAH = 18 mg

SUMMARY

We'll probably never estimate the past lung cancer risk with a high degree of confidence.

What we need to do is estimate the future risk.

We need to get the diesel emissions risk issue in perspective in regard to everything else that's out there.

To the extent that there is environmental risk from soot, the best investment is to get the old smokers away from the people!

SESSION II

Chemical and Physical Properties of Diesel Engine Emissions

Robert Sawyer, Chair

David Kittelson

Nigel Clark

Michael Spallek

Barbara Zielinska

The session included presentations on the impact of different measurement techniques on the characterization of diesel particulate matter. Methods for characterizing emissions from heavy-duty and light-duty vehicles were presented, followed by a discussion of how diesel engine emissions have changed over the past 20 years.

Characterization of Diesel Particulate Matter
David Kittelson

Characterization of Diesel Particulate Matter: Impact of Measurement Techniques

David B. Kittelson
Department of Mechanical Engineering
University of Minnesota

HEI Diesel Research Strategy Workshop
7 March 1999

Nearly all the mass emitted by engines is in the submicron diameter range. Most of the particle number emitted is in the nanoparticle (or nuclei mode) range, $D_p < 50$ nm, while most of the particle mass (and surface area) is in the accumulation mode range, 50 nm $< D_p <$ 1000 nm. Nuclei mode particles are typically hydrocarbons or sulfate, while accumulation mode particles are mainly carbonaceous soot agglomerates.

Ever tightening emission standards for new diesel engines have led to dramatic reductions in particle mass emitted by these engines. However, a number of recent studies suggest that at similar mass concentrations nanometer size particles are much more dangerous than micron size particles. In addition, a recent HEI study (1) showed that a new diesel engine with low particle mass emissions produced much higher concentrations of tiny particles than a similar engine of older design and much higher mass emissions. These results have led to questions about whether mass based emission standards should be supplemented with particle size, surface area, or number concentration standards. It is important to note, however, that emission of high concentrations of nanometer sized particles is not a new phenomenon. Examination atmospheric particle size measurements made on or near roadways reveals that high concentrations of particles in the nanometer size range have been observed for more than 30 years (2-6). While exposure to such particles may not be new, the possible adverse health effects of these particles makes their accurate measurement essential.

Accurate measurement of nanometer sized particles depends upon choice of appropriate instruments and design of a dilution and sampling system that accurately simulates the atmospheric dilution process. The choice of instruments is fairly straightforward because there are only a few commonly used commercial instruments.

Probably the most commonly used sizing instrument is the TSI scanning mobility particle sizer (SMPS). It has a sizing range from about 7 to 700 nm and measures a complete size distribution in two minutes. Particles are sized by their electrical mobility equivalent diameter. The instrument requires a steady sample during the measurement period so that its usefulness in measuring transients is limited. However, some investigators (7) have used the instrument in a single size mode. This allows transients to be studied one size at a time. A size distribution may be reconstructed by measuring several transient cycles and combining the results. New versions of the SMPS have recently become available that size particles as small as 3 nm.

The primary particle detection device in the SMPS is the condensation particle counter. A CPC counts all particles larger than its lower size limit, typically 3 to 10 nm diameter, depending on the instrument configuration. CPCs have response times of only a few seconds and thus can be used as stand-alone instruments to examine the variation of number concentration during transients. Some models of the CPC have inlet concentration limits of only 10^4 or 10^5 particles/cm^3 and require highly diluted samples.

Another sizing instrument that is starting to be applied to engine exhaust particle size measurements is the electrical low-pressure impactor (ELPI). The ELPI sizes particles in the 30 nm to 10 μm range by aerodynamic diameter. The great advantage of this instrument is its transient response; it can produce a complete size distribution in 1-2 s. However, the instrument does not respond to particles smaller than 30 nm, the size range that often contains most of the particle number. Also, response in the upper end of the size range may be compromised by the presence of the large concentrations of nanoparticles common in engine exhaust aerosols.

Dilution and sampling may have a very strong influence on engine exhaust size distributions in the nanoparticle size range. The influences of dilution and sampling are only poorly understood. However, it is clear that a significant amount of particulate matter is formed by gas-to-particle conversion during exhaust dilution from particle precursors present in the vapor phase in the tailpipe. These precursors include sulfuric acid and fuel and oil residues. Typically more than 90% of the number and about 30% of the mass of exhaust particulate matter forms during dilution. Thus a significant fraction of the particulate matter associated with engine exhaust is not present in the tailpipe. This particle formation process is much faster than and distinct from the formation of secondary particulate matter that results from atmospheric reactions of reactive species present in the exhaust.

Several processes are involved in the gas-to-particle conversion that takes place during dilution. As exhaust dilutes and cools, the ratio of partial pressure to vapor pressure of condensable species, the saturation ratio, goes through a maximum. In most systems the only species that is likely to become super-saturated enough during dilution for homogeneous nucleation (gas-to-particle conversion to form new particles) is sulfuric acid. Sulfuric acid particles may nucleate by heteromolecular nucleation of sulfuric acid - water nuclei. These nuclei may grow by coagulation (collision with other nuclei), absorption of sulfuric acid and water, and (this is only a hypothesis) growth of nuclei by absorption of hydrocarbons to form particles of mixed composition. Simultaneous competitive processes may suppress growth. These include adsorption of sulfuric acid and hydrocarbons onto carbon agglomerates and coagulation of nuclei with carbon agglomerates. Thus carbon (soot or carbonaceous agglomerates) in the exhaust will tend to suppress the formation and growth of nanoparticles. The potential of an engine to form nanoparticles will depend upon the relative concentrations of volatile particle precursors (sulfuric acid and hydrocarbons) and solid carbon. Consequently a clean engine with low soot emissions may have more potential to form nanoparticles than an engine with higher soot emissions. But this is only the potential - formation also depends strongly upon dilution conditions!

Recent laboratory studies (8,9) have demonstrated the extreme sensitivity of nanoparticle formation to dilution conditions. When a modern, medium-duty diesel engine was run under stabilized, steady-state conditions, changes of up to two orders of magnitude in number emissions were observed as dilution conditions were varied over the range of dilution ratios, temperatures, and residence times. Essentially all of the changes in number emissions were due to changes in the nanometer size range. Dilution had little influence on the solid carbonaceous particles in the accumulation mode diameter range. Even larger changes in number emissions, sometimes more than three orders of magnitude, were observed when the engine was equipped with an exhaust filter that removed most of the carbonaceous particles. Thus number emissions and particle size are extremely sensitive to dilution conditions. This makes it essential that we develop dilution and sampling systems that accurately mimic the atmospheric dilution process. We must understand the tailpipe to nose process.

Nanoparticles have been observed on and near roadways for many years. These particles are likely to have been formed by nucleation during dilution. Coagulation will cause these nuclei to grow quickly into the accumulation mode diameter range during atmospheric aging, especially if a large accumulation mode is present. Consequently the highest number weighted exposures will occur on or near roadways where particles are young before significant coagulation has occurred.

If nanoparticles are a problem, emissions spark ignition engines may have to be considered. Spark ignition engines typically emit smaller particles than diesel engines and are an important source of fine particles and nanoparticles. A recent study in Colorado concluded that up to 2/3 of the fine particle mass emitted by vehicles was from spark ignition engines. New gasoline direct injection engines emit much higher particle concentrations than conventional engines and may approach diesel levels under some conditions (10).

References

1. Bagley, S. T., K.J. Baumgard, L.D. Gratz, J.H. Johnson, and D.G. Leddy. 1996. Characterization of Fuel and Aftertreatment Device Effects on Diesel Emissions. Health Effects Institute Research Report No. 76, 88 pp.
2. Weingartner, E., et al. 1997(a). Aerosol Emission in a Road Tunnel. Atmosph. Environ. 31(3):451-462.
3. Whitby, K. T. and B. K. Cantrell. 1975. Atmospheric Aerosols - Characteristics and Measurement. ICESA Conference Proceedings, IEEE #75-CH 1004-1 ICESA, paper 29-1, 6 pp.
4. Whitby, K. T., et al. 1975. Characterization of California Aerosols - 1. Size Distributions of Freeway Aerosol. Atmosph. Environ. 9:463-482.
5. Wilson, W. E., et al. 1977. General Motors Sulfate Dispersion Experiment: Summary of EPA Measurements. JAPCA 27(1):46-51.
6. Kittelson, D. B., et al. 1988. Characterization of Diesel Particles in the Atmosphere. Coordinating Research Council AP-2 Project Group Fiual Report.
7. Rickeard, D., et al. 1996. Exhaust particulate Size Distribution: Vehicle and Fuel Influences in Light Duty Vehicles. In Diesel Engine Combustion and Emission Control (SP-1206) SAE 961980, p. 87-111.
8. Abdul-Khalek, I., D.B. Kittelson, and F. Brear, "The Influence of Dilution Conditions on Diesel Exhaust Particle Size Distribution Measurements," SAE Paper No. 1999-01-1142, International Congress and Exposition, Detroit, MI, March 1-4, 1999.
9. Abdul-Khalek, Imad S., David B. Kittelson, and Fred Brear, "Diesel Trap Performance: Particle Size Measurements and Trends," SAE Paper No. 982599 and SP-1399, presented at the SAE International Fall Fuels & Lubricants Meeting & Exposition, San Francisco, CA, October 19-22, 1998.
10. Graskow, B.R., D.B. Kittelson, M.R.Ahmadi, and J.E. Morris, SAE Paper No. 1999-01-1144, "Exhaust Particulate Emissions from Two Port Fuel Injected Spark Ignition Engines," International Congress and Exposition, Detroit, MI, March 1-4, 1999.

Characterization of Diesel Particulate Matter: Impact of Measurement Techniques

David B. Kittelson
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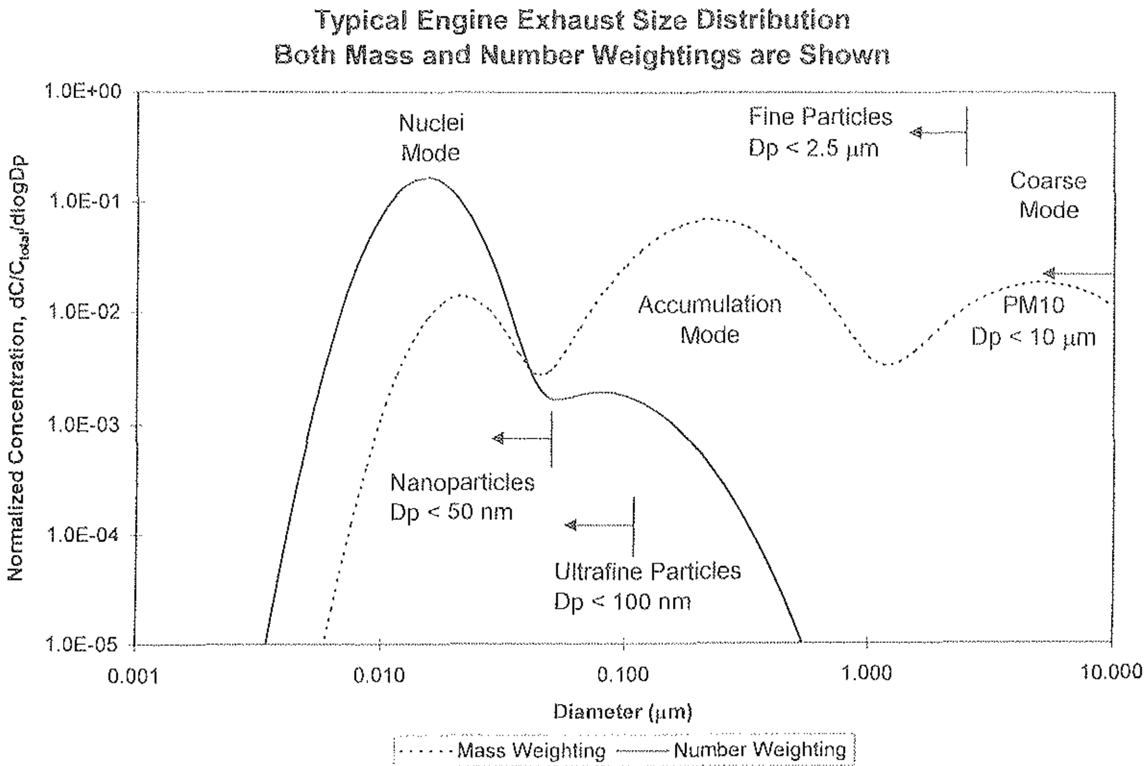
7 March, 1999

Outline

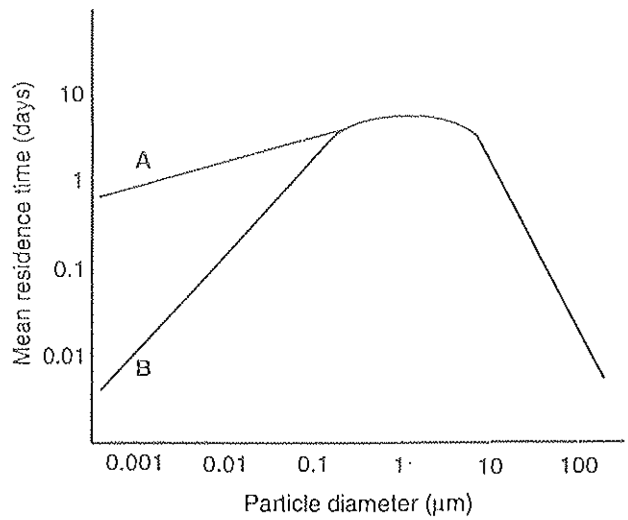
- Typical Engine Exhaust Size Distribution
- Why Measure Size?
- Background -- Historical Perspective
- Typical Sizing Instruments
- Nature of particles
- Sampling Issues
- Conclusions



Why Measure Size?

- Health effects
- Behavior in atmospheric
  - Residence time
  - Visibility
  - Surface reactions
- Performance of aftertreatment devices
- Evidence that low (mass) emission engines may emit much smaller particles and higher number concentrations

Figure 2.7 The Residenc‹ me of Atmospheric Particles Entering the Boundary Layer.



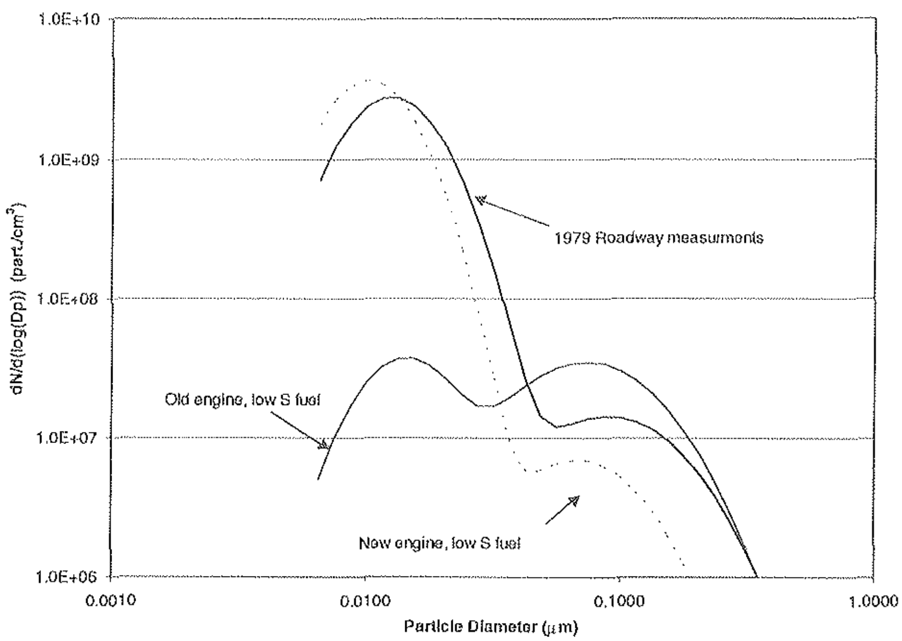
Source: Adapted from Jaenicke, 1993.
Note: Curve A represents the deposition of aerosol particles to the ground, while curve B also includes the effect of coagulation, which reduces the number of small particles without removing their substance from the air.

"Airborne Particulate Matter in the United Kingdom," Third Report of the Quality of Urban Air Review Group, The University of Birmingham, Edgbaston, England, prepared at the request of the Department of the Environment, UK (May 1996).

A Recent HEI Study Gave a Surprising Result for a Low Emission Engine

- Particle concentrations and size distributions were measured for a 1988 and a 1991 engine
  - When run on very low sulfur fuel particle number emissions were much higher, 30 -100 times, from the new engine although mass emissions were about 3 times lower
  - The 1988 engine produced high number emissions, but not as high as the 1991 engine when run on a 1988 type fuel (higher sulfur)
  - This raised concerns that new engines might be producing large numbers of nanoparticles while still meeting mass emission standards and focused attention on nanoparticle emissions
- However reviews of measurements made on and near roadways in the 70's and 80's show high nanoparticle emissions. **High nanoparticle emissions may be a problem but are not a new development!**

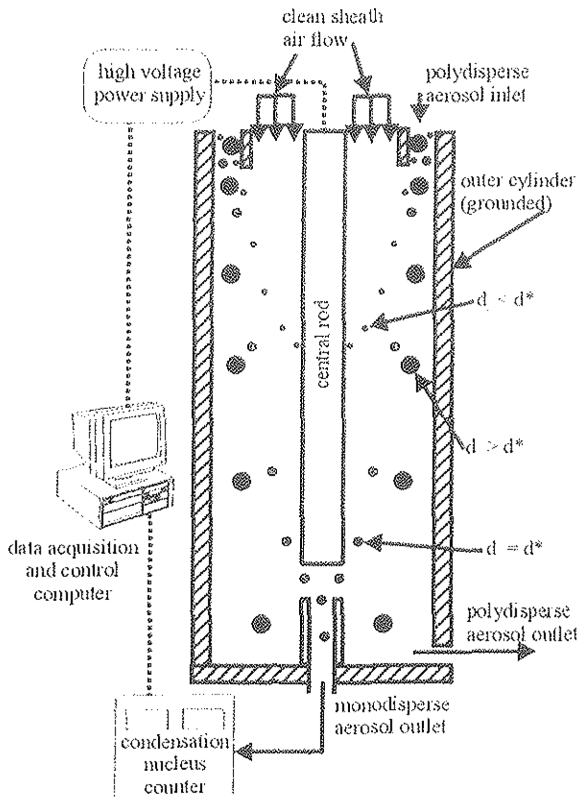
Number Size Distribution Data from HEI Report and 1979 CRC Roadway Study



Particle Sizing

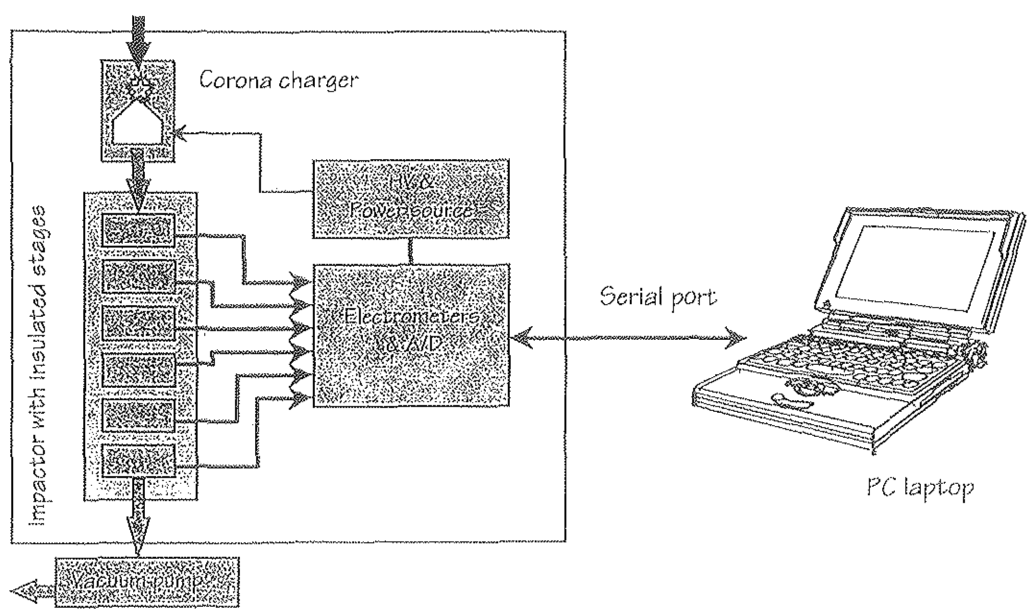
- Electron microscope
- Mechanical mobility
  - Aerodynamic diameter (size, shape. density)
    - » Inertial impactors (10 nm -> 10 μm)
    - » Aerodynamic particle sizer (0.5 - 10 μm)
  - Stokes diameter (size and shape)
    - » Electrical mobility diameter - EAA, DMA, SMPS (3 - 700 nm)
    - » Diffusion diameter - Diffusion battery (10 - 200 nm)
- Light scattering (size, shape, refractive index) (100 nm -> 10 μm)

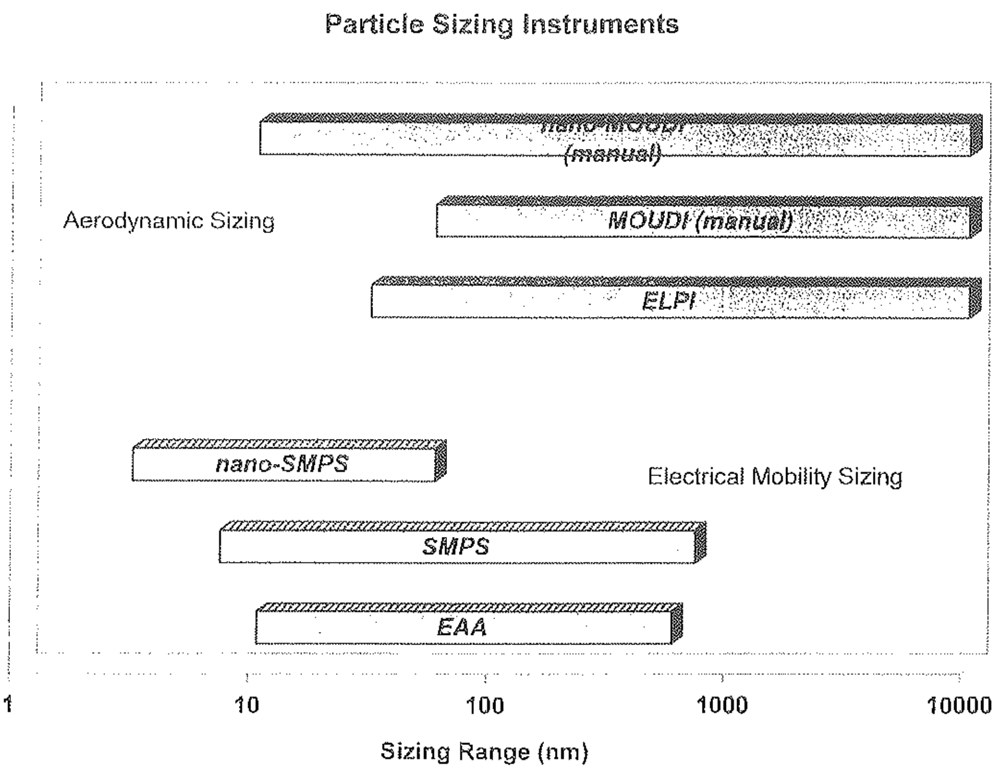
Scanning Mobility Particle Sizer (SMPS)



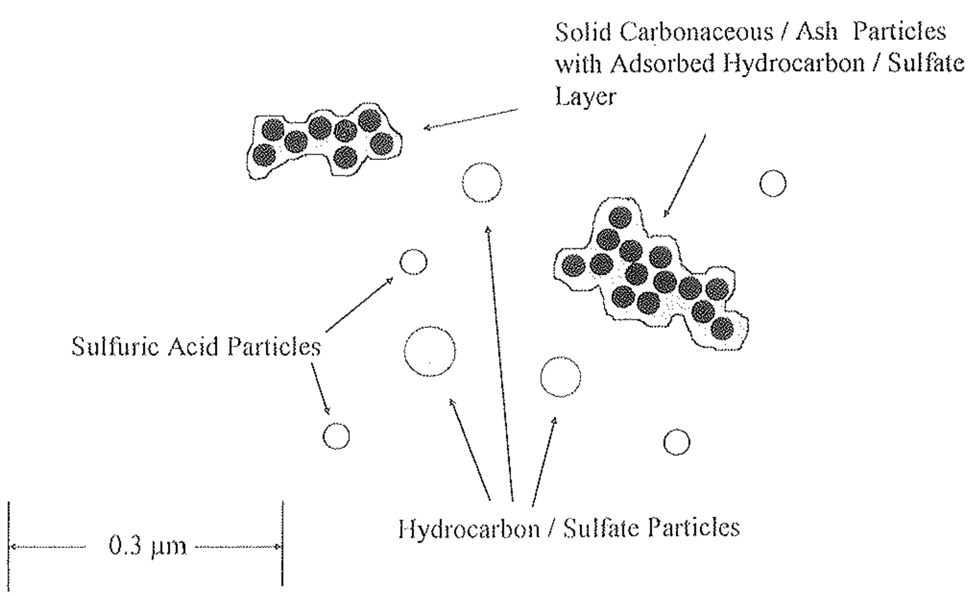
Electrical Low Pressure Impactor

- Combines electrical detection with aerodynamic size classification





Particles consist mainly of highly agglomerated solid carbonaceous material and ash and volatile organic and sulfur compounds.



Sampling Problems - Wall Interactions

- Losses
  - Thermophoresis
  - Inertial
  - Diffusion
    - Particles
    - Particle precursors
- Additions
  - Reentrainment
  - Outgassing

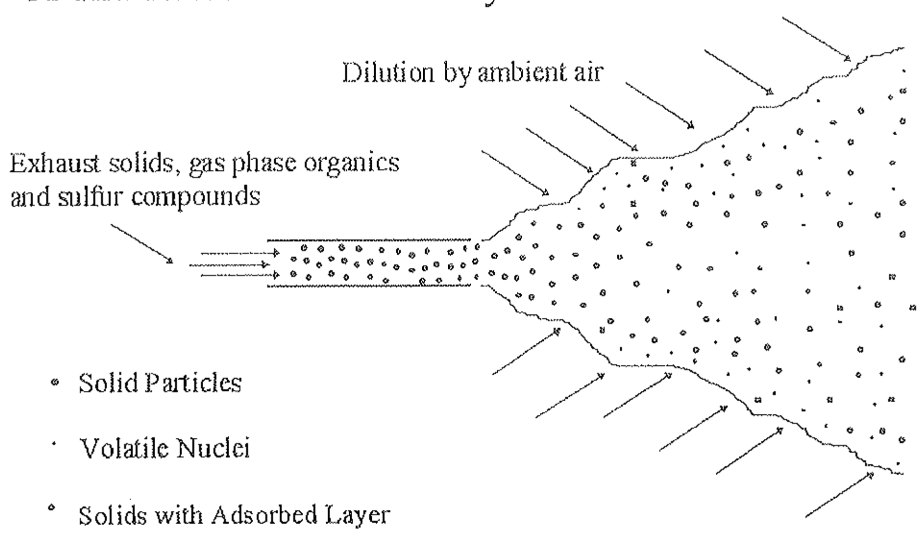
Sampling Problems - Homogeneous

Processes sensitive to correct simulation of atmospheric dilution

- Gas to particle conversion
  - Nucleation
  - Adsorption / condensation
- Coagulation

Atmospheric Dilution Leads to Nucleation, Absorption, and Adsorption

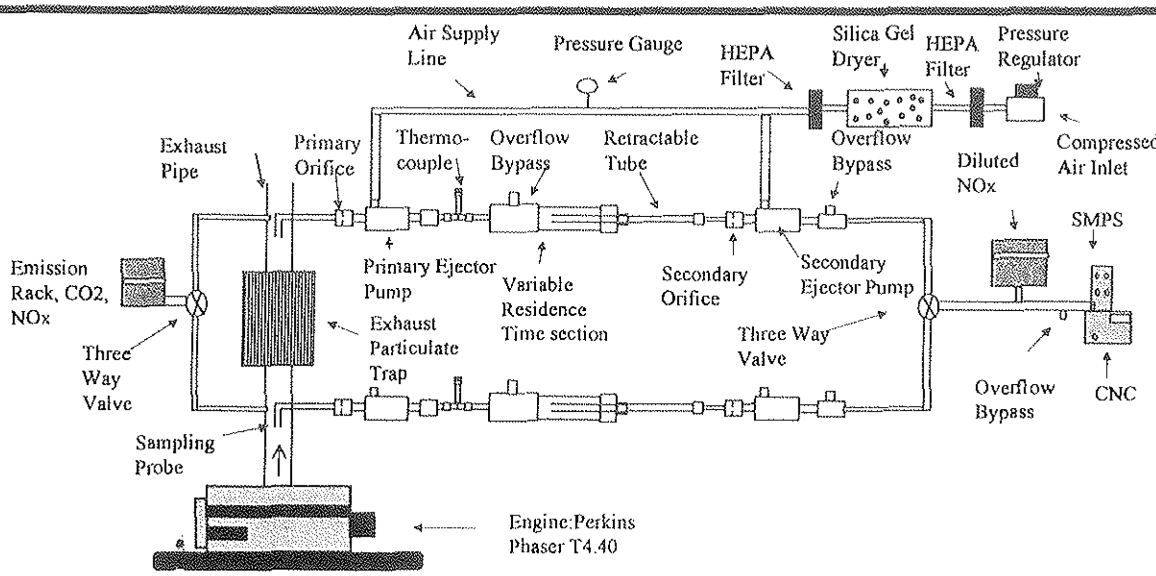
A dilution ratio of 1000 may be reached in 1 - 2 s

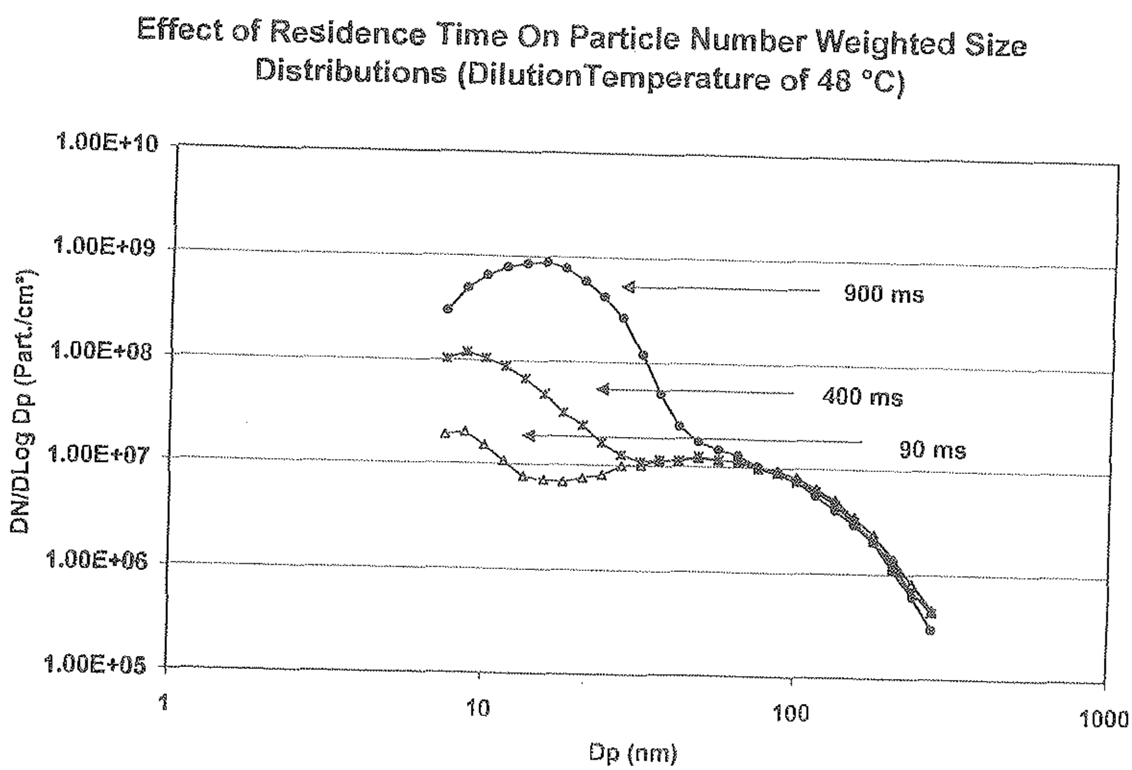


Significant Gas to Particle Conversion Takes Place as the Exhaust Dilutes and Cools

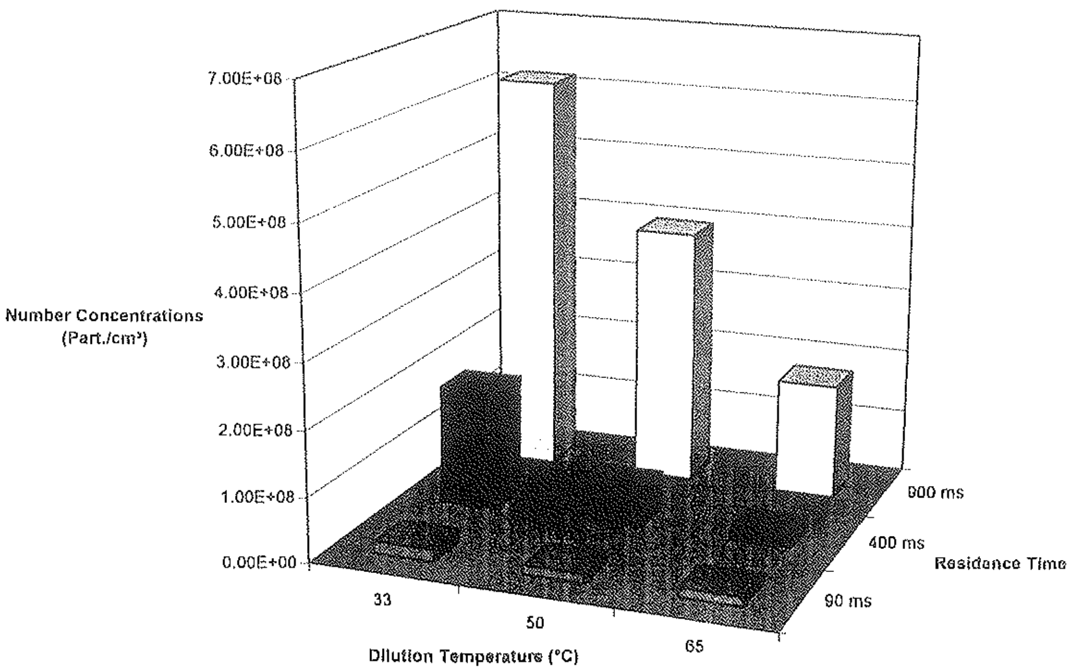
- More than 90% of the particle number may form as nanoparticles
- From 5 to more than 50% of the particle mass may form as nanoparticles and adsorbed material
- This process is very sensitive to dilution conditions

Experimental Setup and Variable Residence Time Micro-Dilution System





Effect of Residence Time and Dilution Temperature On Particle Number Concentrations
(Mode 8, Primary Dilution Ratio of 12, Total dilution of about 700)



Conclusions - 1

- The most difficult problem associated with the characterization of submicron diesel particulate matter is the design of the dilution and sampling system, not the choice of appropriate instruments.
- A significant amount of particulate matter (e.g. 90 % of the number and 30% of the mass) is formed during exhaust dilution from material present in the vapor phase in the tailpipe (e.g., sulfuric acid, fuel and oil residues).
  - New particles are formed by nucleation. This is likely to be the source of most of the ultrafine and nanoparticles (and particle number) associated with engine exhaust.
  - Preexisting particles grow by adsorption or condensation.
  - Nucleation and adsorption are competing processes. Soot agglomerates provide a large surface area for adsorption that suppresses nucleation. Thus, diesel engines with low soot mass emissions, may have high number emissions.

Conclusions - 2

- Nucleation and adsorption depend on dilution rate, (or residence time at intermediate dilution ratio), humidity, temperature, and relative concentrations of carbon and volatile matter.
  - Changes of more than two orders of magnitude in nanoparticle concentration may occur as dilution conditions are varied over the range that might be expected for normal ambient dilution, e.g., 0.1 to 2 s dilution time scales.
  - Even larger changes may occur downstream of exhaust filters or with very clean engines where exhaust carbon concentrations are low
- Coagulation may dramatically reduce number concentrations if exhaust is not diluted rapidly.
- Sampling systems should mimic atmospheric dilution to obtain sample streams representative of human exposure for size analysis.

Conclusions - 3

- Currently most of the particles in the nanoparticle size range are volatile. However, as engines become cleaner, metallic ash particles from the lubricating oil (or fuel if metallic additives are present) may become more important.
- Spark ignition engines typically emit smaller particles than diesel engines and are an important source of fine particles and nanoparticles.
  - A recent study in Colorado concluded that up to 2/3 of the fine particle mass emitted by vehicles was from spark ignition engines
  - New gasoline direct injection engines emit much higher particle concentrations than conventional engines and may approach diesel levels under some conditions
- Nanoparticles are formed under roadway dilution conditions - and have been for many years!

Characterization of Heavy-Duty Vehicle Emissions
Nigel Clark

EXTENDED ABSTRACT FOR HEI DIESEL WORKSHOP

CHARACTERIZATION OF HEAVY DUTY VEHICLE EMISSIONS

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West Virginia University
Morgantown WV 26506-6106

The national Class 8 truck fleet is growing steadily and diesel engines are expected to capture substantial portions of the light truck and sport utility market over the next decade. Particulate Matter (PM) and Oxides of Nitrogen (NO_x) have traditionally been regarded as the emissions of concern from heavy duty diesel vehicles, although certain organic compounds within the hydrocarbon (HC) fraction of the exhaust are now also receiving attention. Diesel engine technology in the USA has evolved rapidly over the last decade, due to increasingly stringent federal and California standards, and enabled largely by the revolution in electronic controls. In-cylinder fuel injection is now accomplished through electronic measurement and at higher pressures than previously employed, so that mass emissions of PM have been reduced, even if the number count of ultrafine particles remains in question. NO_x emissions have been reduced through retarded fuel injection, in-cylinder combustion advances and use of high air to fuel ratios. Exhaust gas recirculation is now emerging as a NO_x control measure. Aftertreatment devices such as traps and catalytic converters hold promise for reduction of PM and HC, but NO_x reduction strategies for the exhaust stream have not yet achieved great success. Emissions have also been assuaged through the introduction of low sulfur diesel fuel, and low aromatic fuel in California. Research has shown that paraffinic and oxygenated fuels with high cetane ratings offer further emissions advantages.

Despite these advances, knowledge of the quantitative contribution of heavy duty automotive diesel exhaust to the atmospheric inventory is poor, and the current models are ill founded. Accurate inventory is needed both for epidemiological studies and in development of plans directed at air quality improvement. Recently, data have revealed that many electronically controlled engines emitted high NO_x under cruise conditions as a fuel economy measure. Current inventory models do not account for this contribution, and are based on certification data that is not representative of emissions from vehicles in real use. Chassis dynamometer data have shed light on real world truck and bus emissions. Whether they are expressed in units of g/bhp-hr, g/mile or g/gallon of fuel, emissions remain strongly dependent on the test cycle that is employed, which implies that emissions in the real world vary widely with vehicle vocation and behavior. In particular, PM and carbon monoxide emissions depend strongly on transient behavior of the engine. Present models also fail to consider the effect of engine tampering and malmaintenance on emissions, and the effect of terrain, altitude, cold starting and weather conditions on emissions. Laboratory emissions measurements fail to take into account the nature of dilution and effect of temperature and humidity encountered across the nation. Several approaches exist to improve modeling efforts, but data on the whole in-use spectrum of trucks, buses and non-road diesel vehicles are scarce. There is hope for the development of transient instantaneous emissions models, including neural net models, that can be employed to predict the emissions for various types of truck activity documented in the field. Improved instantaneous emissions measurement procedures are needed because standard emissions analyzers have slow response times. The Tapered Element Oscillating Microbalance is now producing useful data on continuous PM mass emissions for use in advanced inventory models.

Alternative fuels have offered some advantages in replacing diesel. Spark ignited natural gas lean burn engines have low PM and NO_x potential but suffer loss of fuel economy. Dual fuel natural gas engines have emissions benefits with good fuel economy, but require that two fuels are carried on board. Compression ignition alcohol fueled engines emit unburned alcohol and aldehydes, but reduce NO_x and PM. Soy-derived biodiesel offers lowered PM emissions relative to diesel but is an expensive fuel. Potential lies in the use of improved diesel formulations, and blends with natural gas-derived Fischer-Tropsch liquids. There is at present no competitor, either in efficiency or economy, to the diesel engine for heavy duty transportation, although lightly hybridized diesel-electric vehicles may serve to reduce the national emissions inventory.

CHARACTERIZATION OF HEAVY-DUTY VEHICLE EMISSIONS

Health Effects Institute Diesel Workshop
March 7, 1999

Presented by:
Nigel N. Clark

Department of Mechanical and Aerospace Engineering
College of Engineering and Mineral Resources
West Virginia University

with thanks to David L. McKain and colleagues
at West Virginia University

DIESEL ENGINE EMISSIONS

Particulate Emissions

Unburned fuel and lubricants
Adverse respiratory effects
For modern diesel engines: $PM \cong PM_{10} \cong PM_{2.5}$

Oxides of Nitrogen

Consist of NO and NO_2. NO_2 is higher under low load conditions
Contributes to ground level ozone (smog) and secondary particulate formation

Hydrocarbons

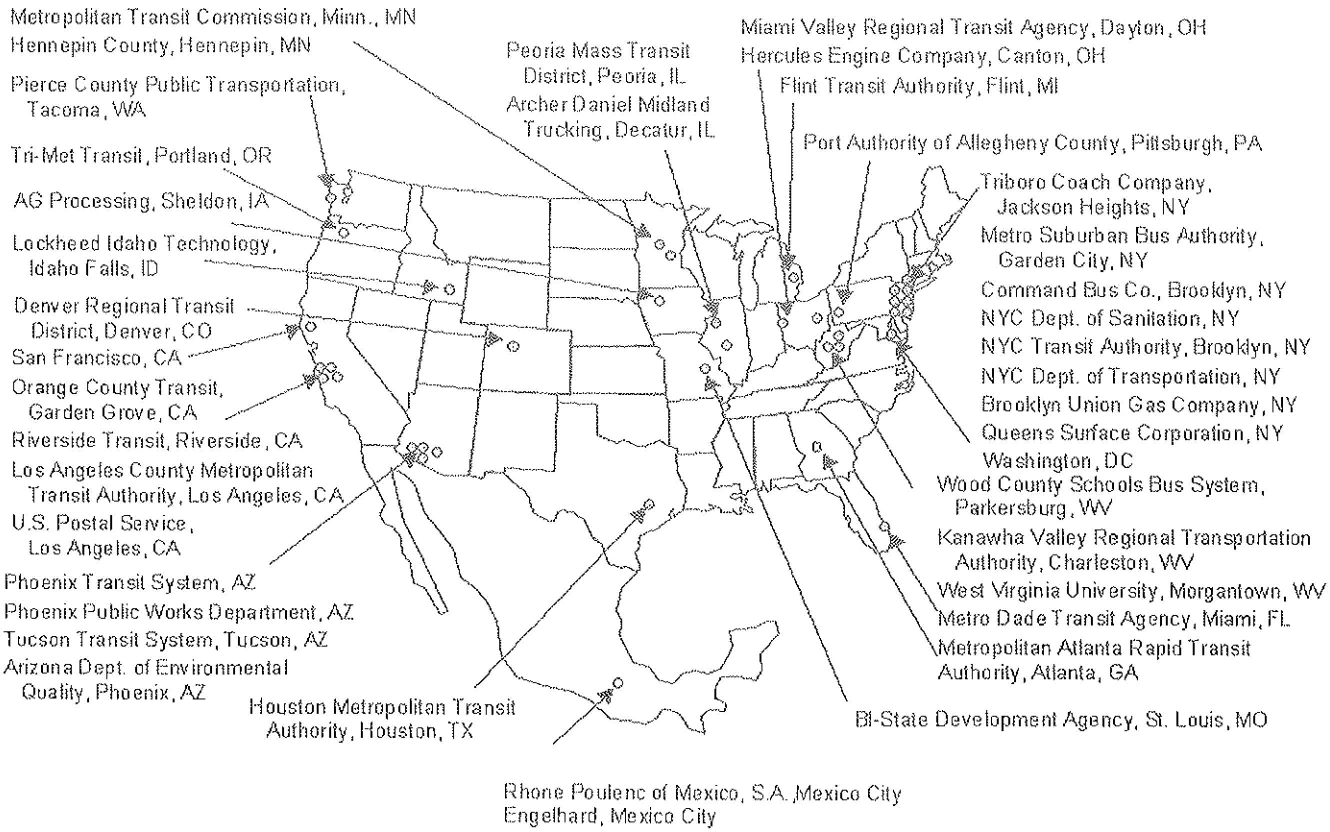
Unburned fuel and lubricants
Contains some toxic species
Contributes to smog

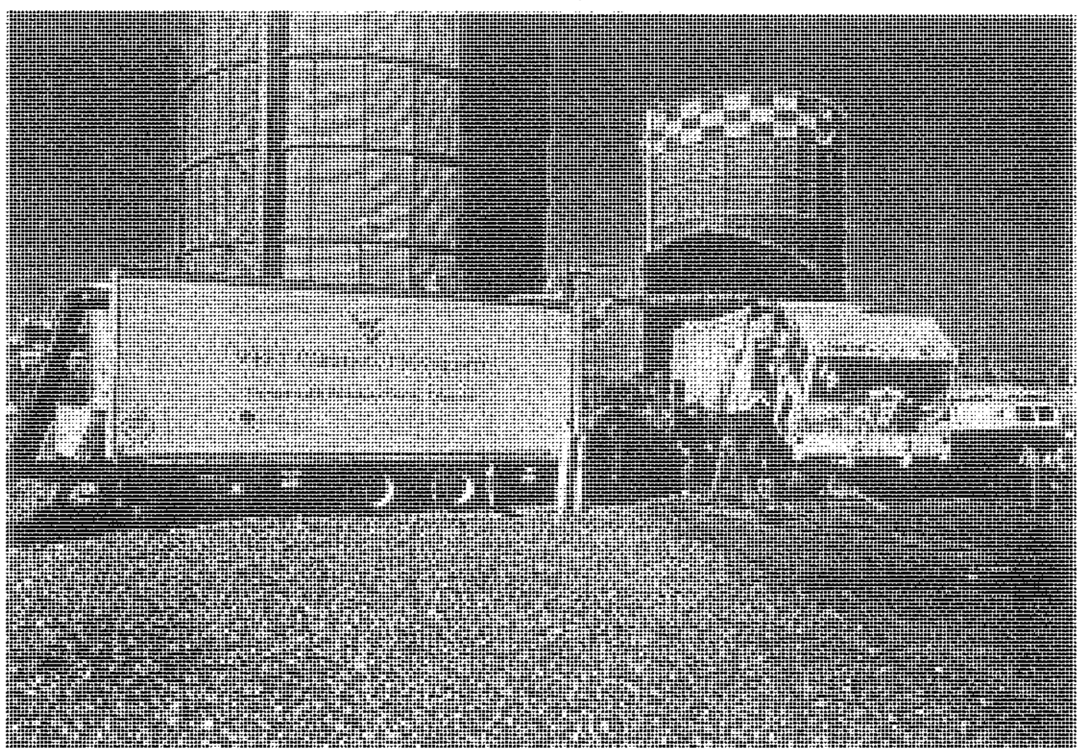
Carbon Monoxide

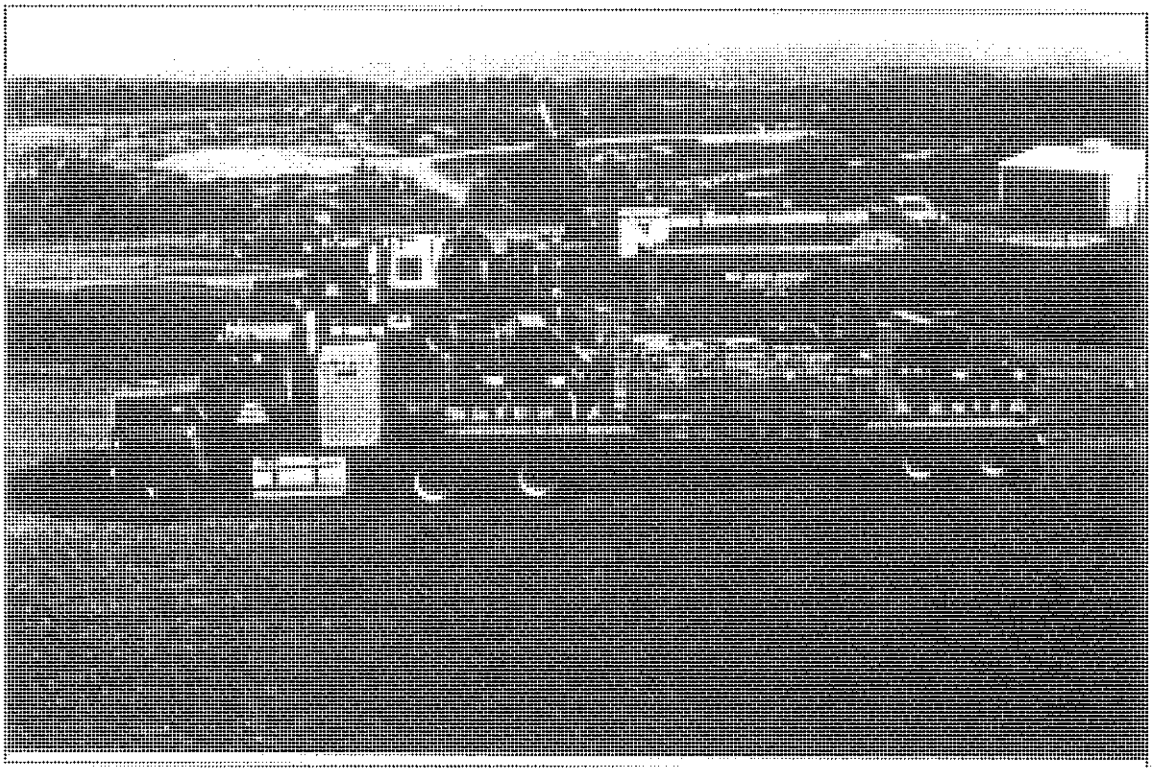
Far lower than for gasoline vehicles
Adverse respiratory effects

TRANSPORTABLE HEAVY DUTY VEHICLE EMISSIONS TESTING LABORATORY

(Lyons, Clark, Gautam)





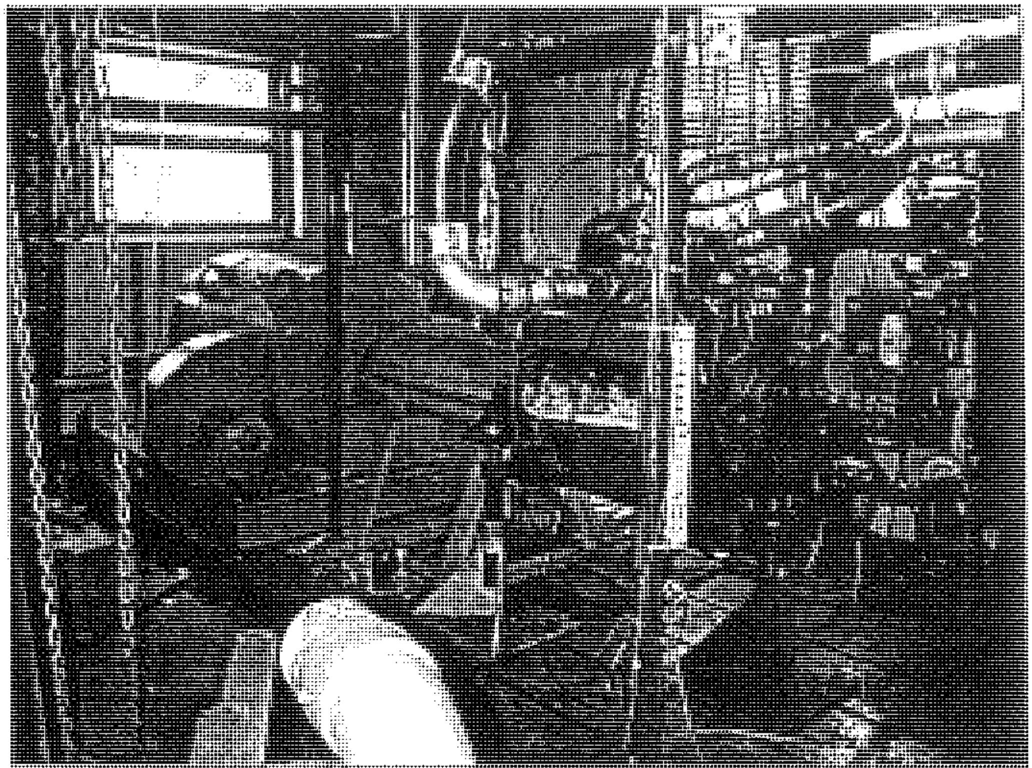


HEAVY-DUTY COMPRESSION IGNITION ENGINE EMISSIONS STANDARDS FOR TRUCKS AND URBAN BUSES

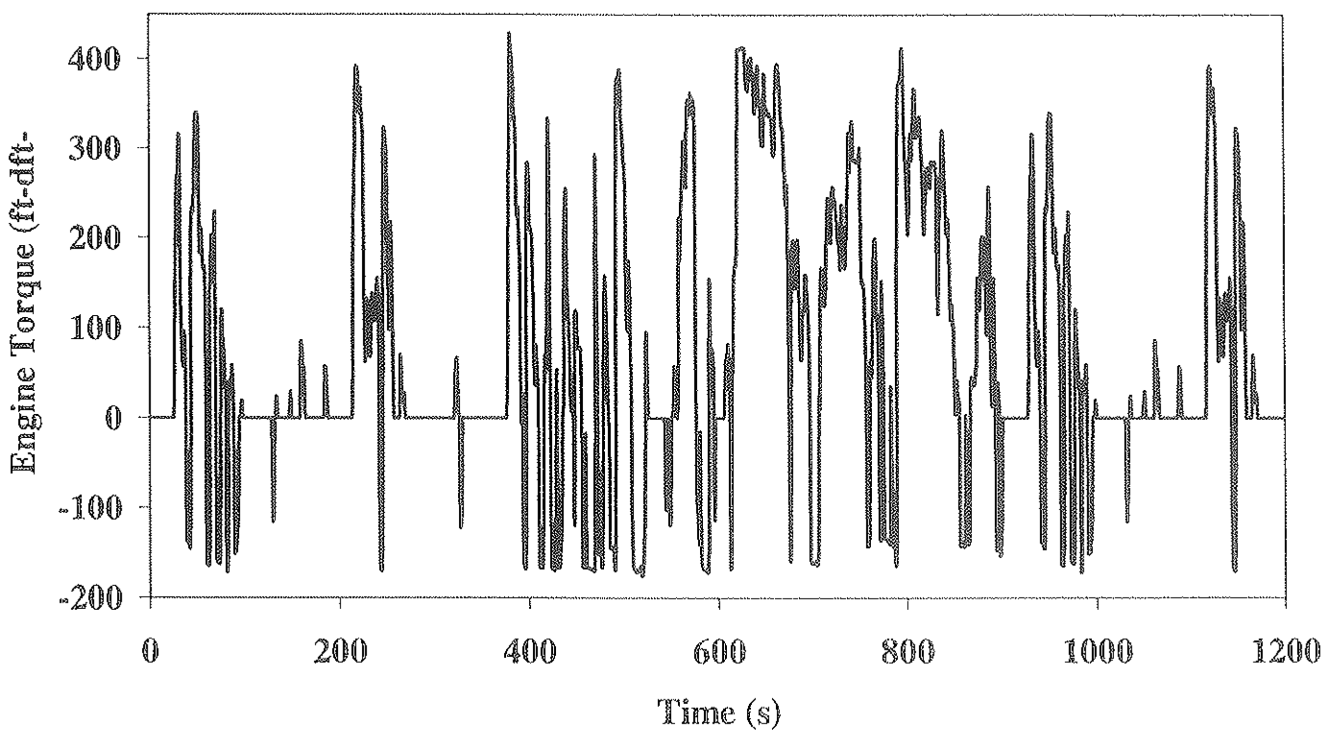
	CO (g/bhp-hr)	HC (g/bhp-hr)	$NMHC + NO_X$ (g/bhp-hr)	NO_X (g/bhp-hr)	PM (g/bhp-hr)
1985-87	15.5	1.3		10.7	No standard
1988-89	15.5	1.3		10.7	0.60
1990	15.5	1.3		6	0.60
1991-93	15.5	1.3		5	0.25/0.10[1]
1994-97	15.5	1.3		5	0.10/0.07[2]/0.05[3]
1998+	15.5	1.3		4	0.10/0.05[3]
proposed	15.5		2.4 or 2.5 with a limit of 0.5 on NMHC		0.10/0.05[3]

[1]1993 Urban Buses, [2]1994-95 Urban Buses, [3]1996- Urban Buses

- The transient federal test procedure does not represent all vehicle use
- Laboratory conditions do not reflect weather, altitude or typical exhaust dilution characteristics



Engine mounted to 550 hp DC dynamometer at the WVU Engine and Emissions Research Laboratory



Engine torque schedule for certification testing heavy duty engines (percent torques have been translated using an engine torque map)

ADVANCING DIESEL ENGINE TECHNOLOGY

HIGHER POWER IN APPLICATION
- Class 8 truck power has risen 25% in last 5 years

SUV APPLICATIONS
- Diesel pickups have taken the market by storm
- Diesel SUV and vans are planned widely

HIGHER POWER DENSITY
- Rising in-cylinder pressures
- Turbocharger boost replacing displacement

ELECTRONIC CONTROLS WITH HIGHER INJECTION PRESSURES
- Wide authority over injection timing and pulsewidth
- Reduction in PM mass emissions
- Uncertain effects on PM number count

ADVANCES IN BOOST CONTROL
- Wastegating and variable geometry turbochargers

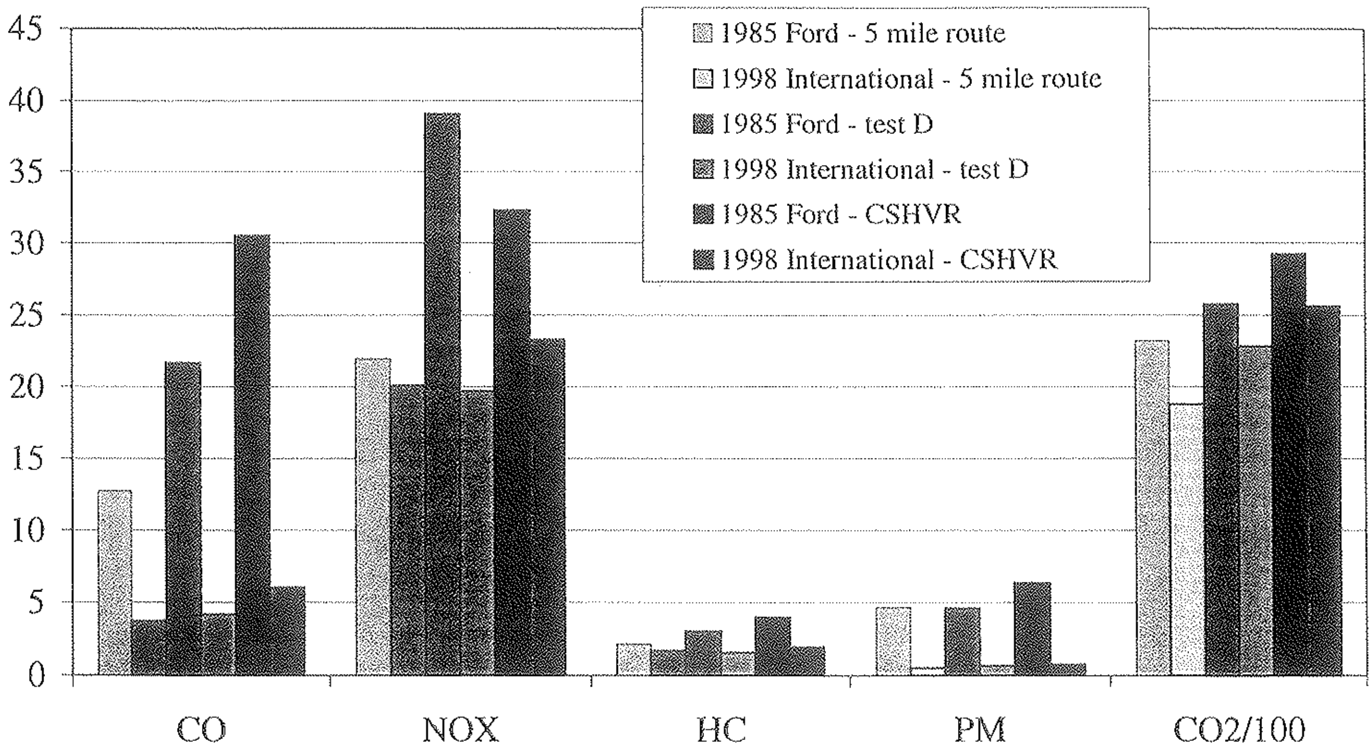
EXHAUST GAS RECIRCULATION
- Lower NO_X emissions
- Concern over engine life

NEW INJECTION SCHEMES
- Unit injectors
- Common rail injection
- HEUI injection system
- Split injection and rate shaping

ADVANCING DIESEL ENGINE TECHNOLOGY (cont.)

AFTERTREATMENT
- Oxidation Catalysts
- Lean NO_X Catalysts
- Continuously Regenerating Traps
- Urea Injection Systems
- NO_X Storage Catalysts
- Novel Trap Regeneration Schemes

LOWER EMISSIONS FUEL FORMULATIONS
- Lower sulfur
- Higher cetane
- Lower aromatic content
- Fischer-Tropsch fuels
- Biodiesel from soy



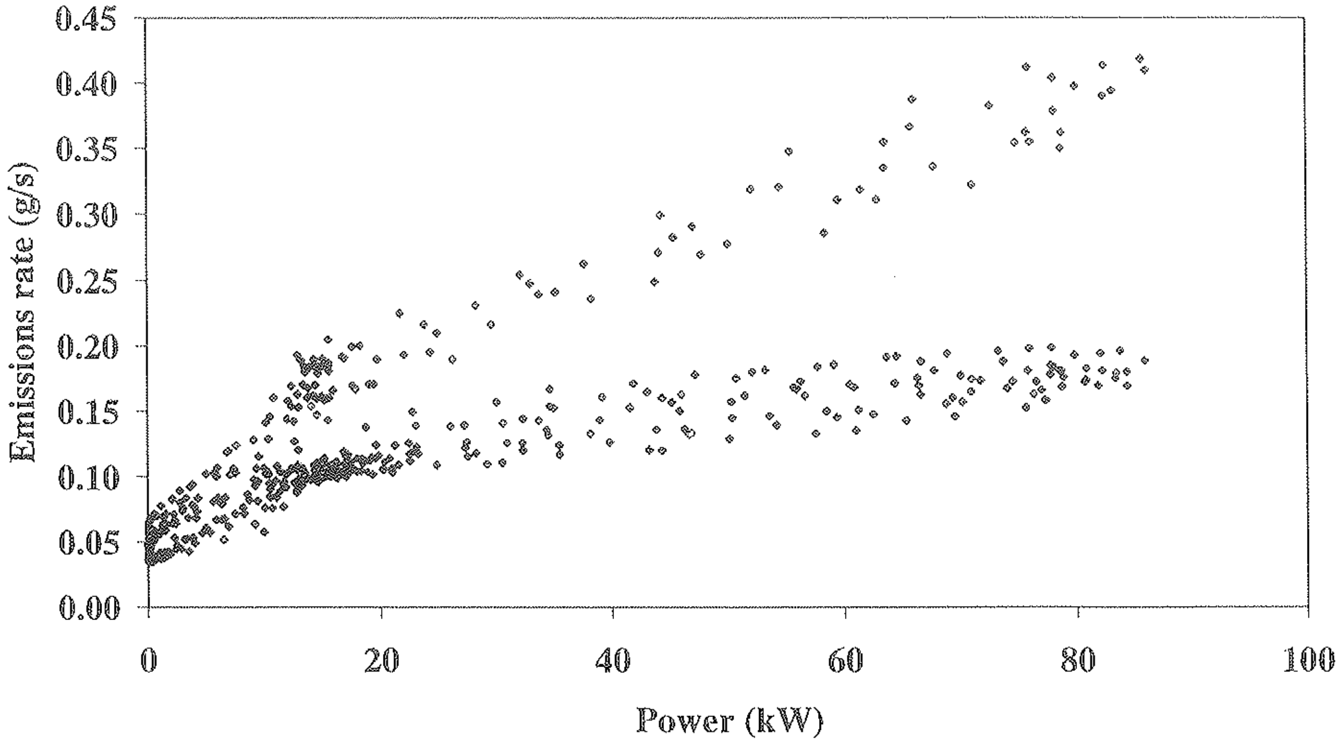
Effect of vehicle model year on emissions

REPEATABILITY OF EMISSIONS

Emissions Results (g/mile)						Fuel Economy		
Run Seq. No.	CO	NO_x	FIDHC	PM	CO_2	mile/gal	BTU/mile	Miles
1241-1	4.21	20.7	0.24	0.68	1979	5.12	25378	2.00
1241-2	3.24	20.7	0.24	0.63	1963	5.17	25153	2.00
1241-3	2.98	20.7	0.24	0.58	1917	5.29	24559	2.01
1241-4	2.64	21.4	0.24	0.59	1977	5.13	25326	2.00
1241-5	3.39	21.5	0.23	0.64	1952	5.20	25018	2.01
1241-6	4.00	21.2	0.25	0.65	1914	5.30	24537	1.98
1241 Average	3.41	21.0	0.24	0.63	1950	5.20	24995	2.00
Std. Dev.	0.60	0.4	0.01	0.04	29	0.08	369	0.01
CV%	17.5	1.8	2.2	6.0	1.5	1.5	1.5	0.6

Emissions and fuel economy data collected from a transit bus over a central business district (CBD) test series

EFFECT OF OFF-CYCLE CONTROL STRATEGIES



NO_X emissions from a transit bus tested using the Central Business District test schedule. Bifurcation of data is due to injection timing changes. The effect of timing changes on PM has not been well quantified. (see Ramamurthy and Clark, Environmental Science and Technology, 1999)

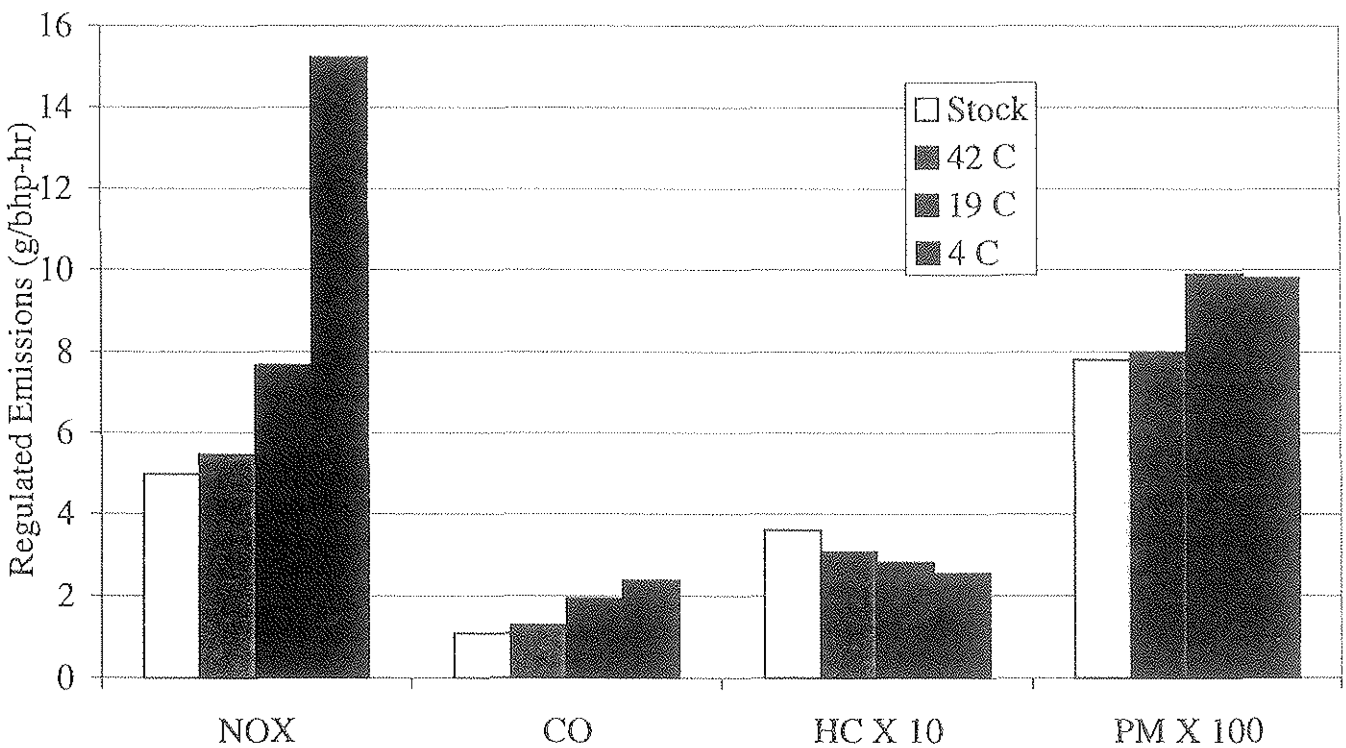
HEAVY DUTY VEHICLE EMISSIONS INVENTORY

Certification Data

- Engine emissions data (g/bhp-hr) are converted to vehicle data (g/mile) using a procedure described by Machiele (EPA, 1998)

$$\frac{g}{mile} = \frac{g}{bhp-hr} \bullet \frac{bhp-hr}{lb} \bullet \frac{lb}{gal} \bullet \frac{gal}{mile}$$

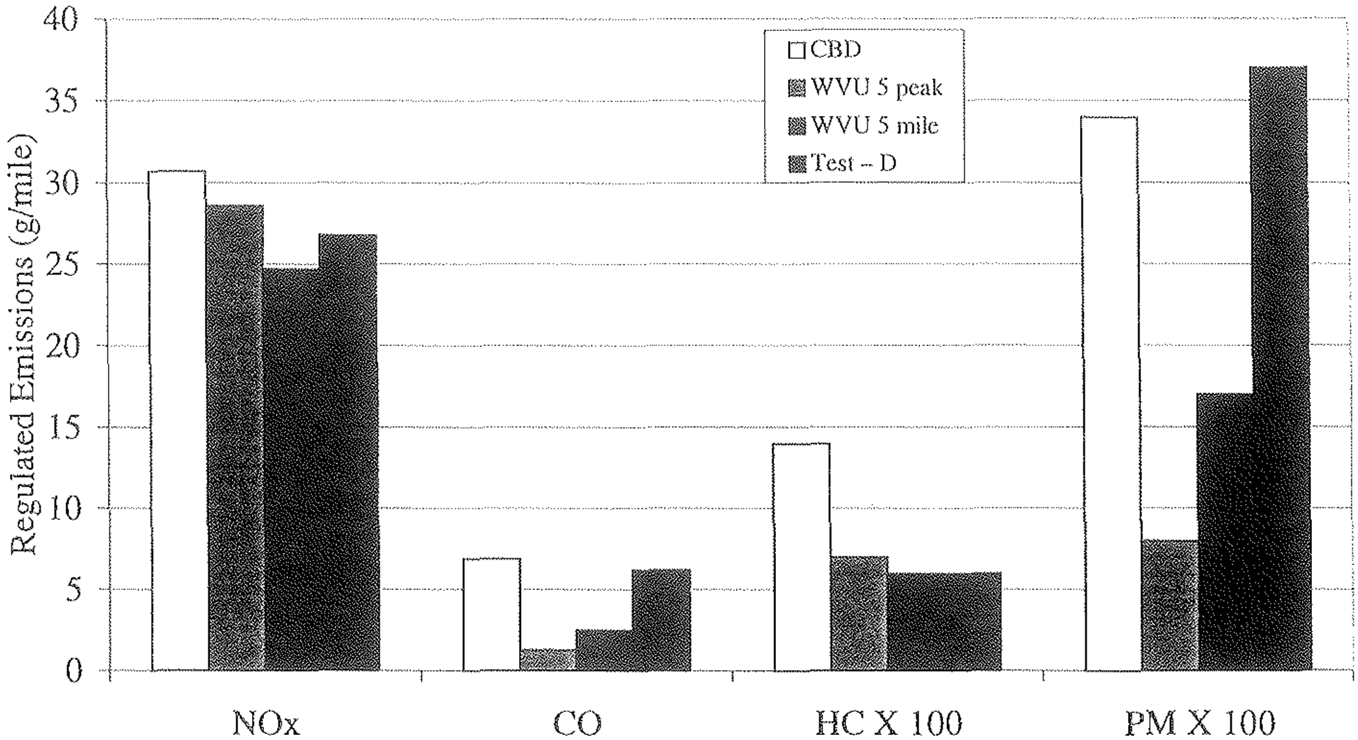
- Engine degradation, less than ideal operating conditions, tampering and malmaintenance are not considered in this approach



Emissions from a diesel engine in various "tampered" modes. The temperatures shown represent a false engine oil temperature signal (see SAE 980407)

Direct use of Chassis Dynamometer Data

- Emissions and fuel economy are measured from in service vehicles
- Less costly and time consuming than engine removal for testing
- Cycles used for testing do not necessarily represent real-world operating conditions
- Strong dependence of emissions on cycle



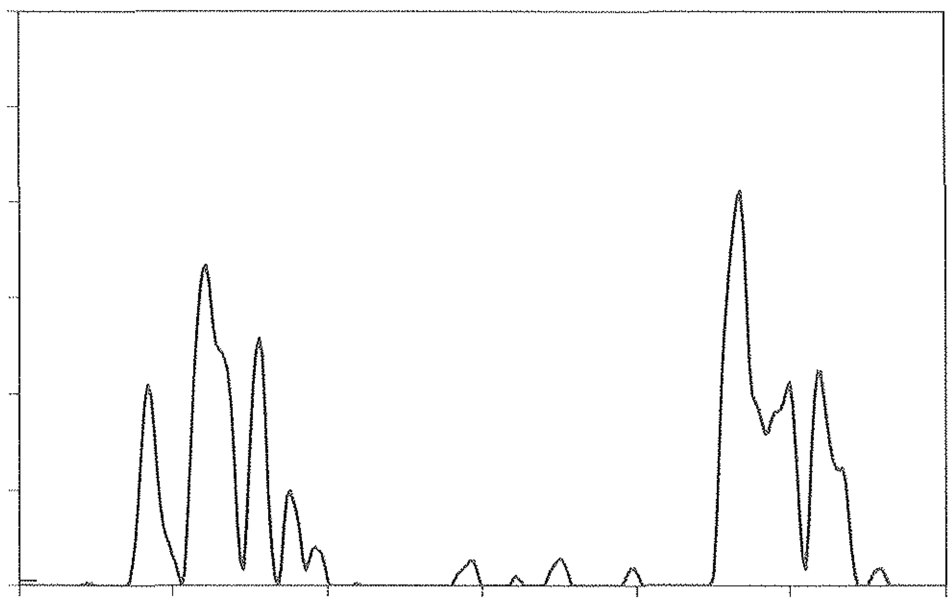
Exhaust emissions from a bus over several chassis cycles employed by the WVU Transportable Emissions Testing Laboratory (see SAE 973203)

g / mile ? **g / bhp-hr ?**

g / g CO_2 ? **g / gallon fuel ?**

Model Based Emissions Factors

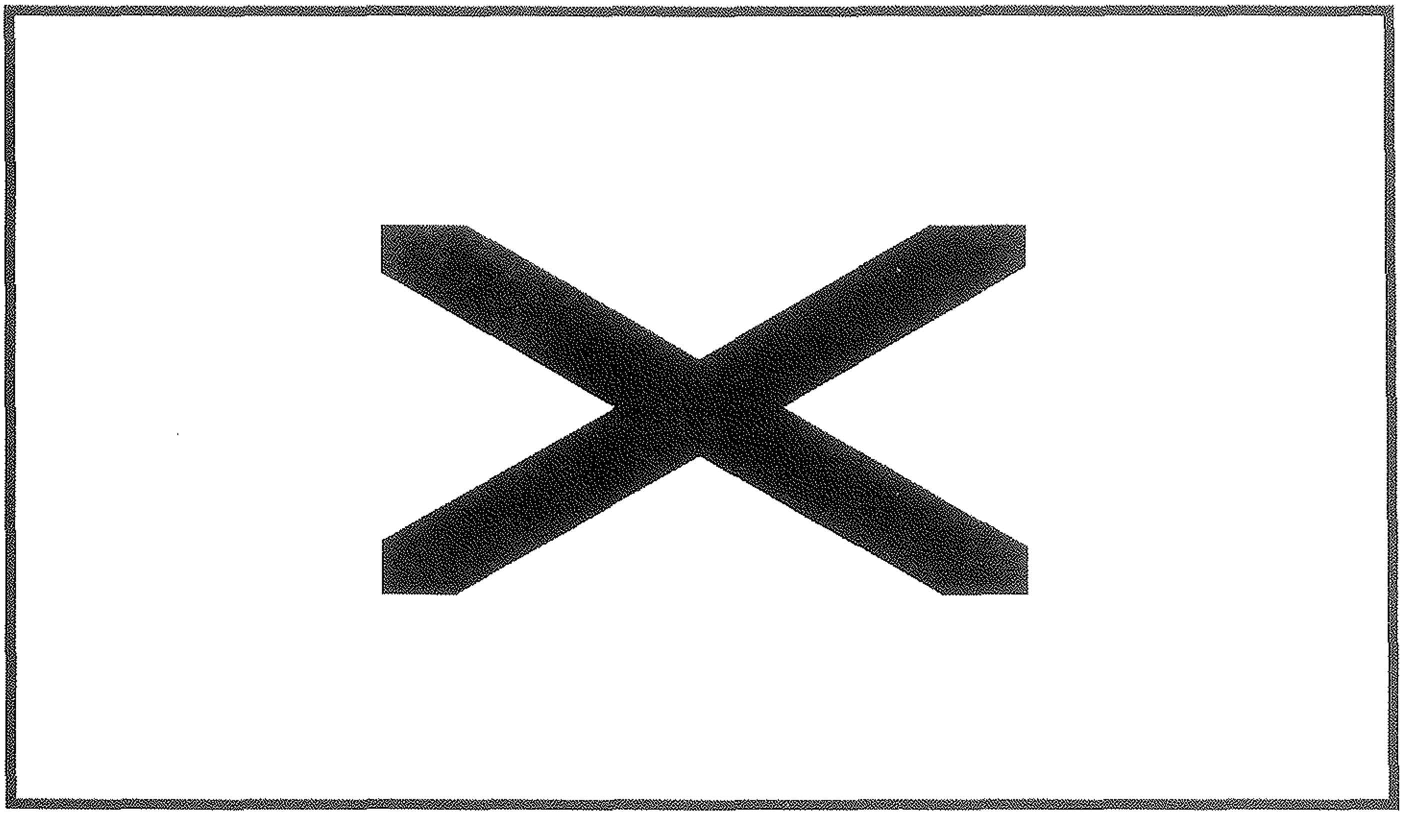
- Chassis dynamometer data used to develop a model to yield factors employed to predict emissions for operating cycles representative of real-world vehicle operation
- Details of continuous emissions data, especially transient dependent CO and HC, are lost due to data smearing as a result of sampling system residence time distribution and emissions analyzer response delays
- Continuous PM data are not readily available
- Models must be verified using chassis testing



Simple model developed to predict engine NO_X emissions from power. Time shifting and data smoothing performed in conjunction with a linear regression to obtain the mathematical model.

Modal Approaches

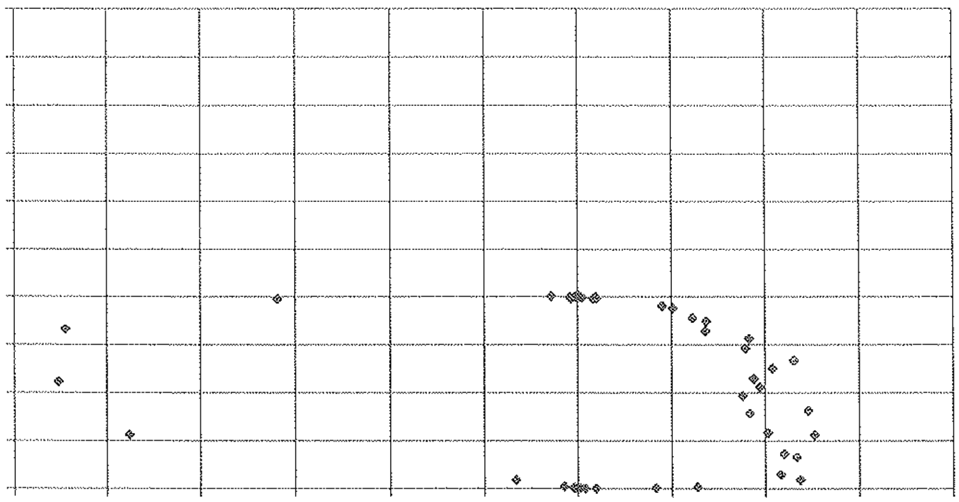
- Segments of test cycles are broken down and considered individually
- Vehicle behavior may be viewed as a collection of modes
- Simplification of model based emissions factors
- Modal approaches have difficulty with transient PM and CO



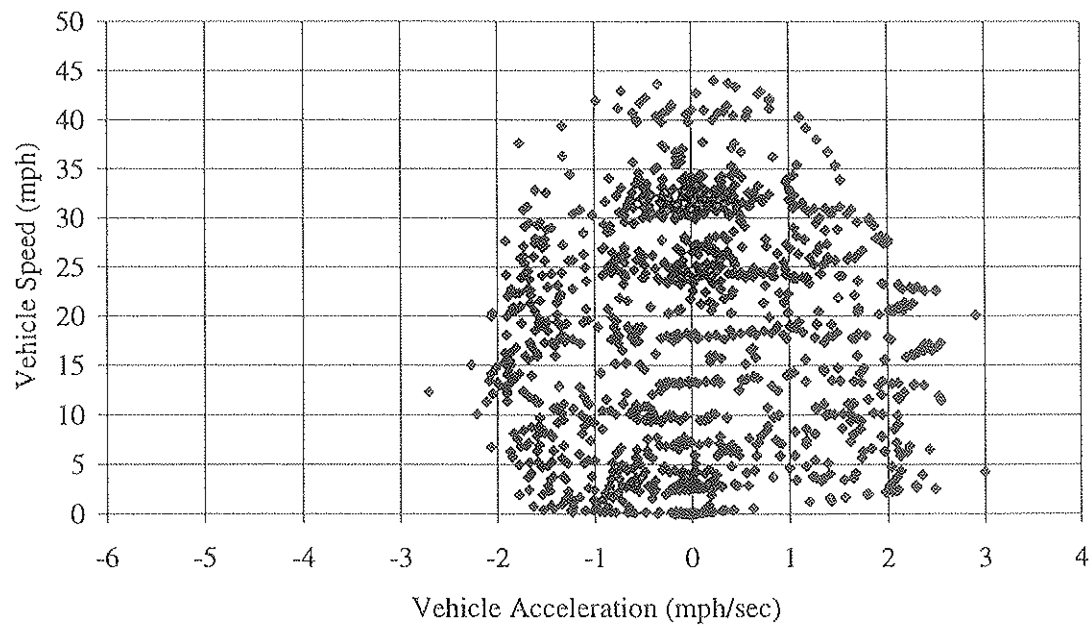
NO_X emissions from a vehicle over the WVU 5 peak driving cycle

Speed-Acceleration Data

- Vehicle speed and acceleration are viewed as independent variables used to predict emissions
- Central business district cycle does not cover a wide enough range to be useful although both the City-Suburban Heavy Vehicle Route and Test D cycle do



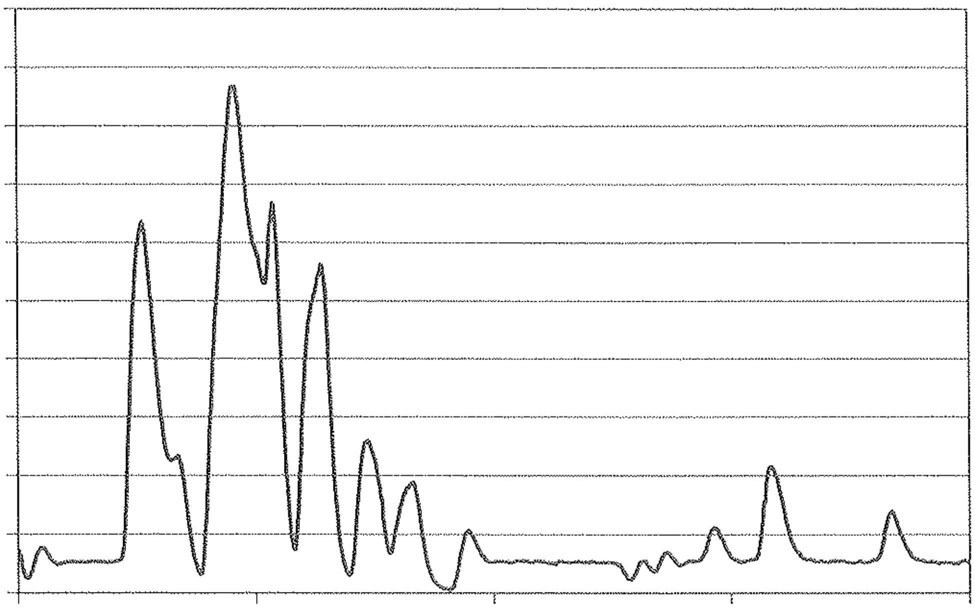
Central Business District Cycle (SAE Recommended Practice J 1376)



City-Suburban Heavy Vehicle Route
(see SAE 1999-01-1467)

Neural Networks

- Emissions data are related to vehicle operating parameters using neural networks
- Resulting emissions factors can be combined with vehicle miles traveled to yield inventory



Actual NO_X (normalized) and that predicted using a neural network (Atkinson, Thompson and Clark)

DIESEL EMISSIONS CORRECTION FACTORS

Existing but poor

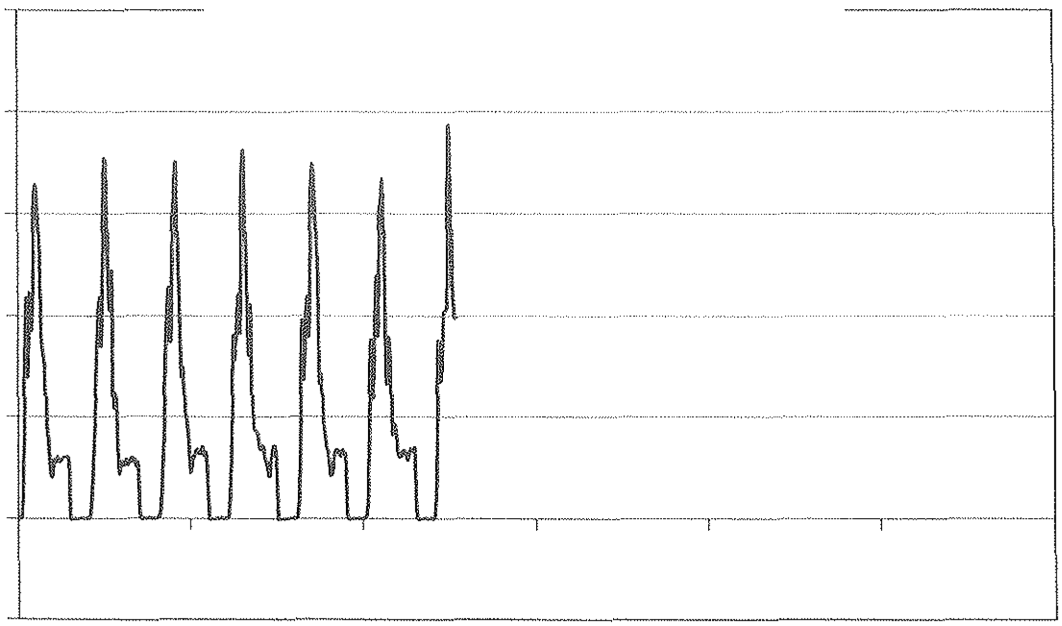
- Speed
- Engine degradation

Lacking

- Cold start
- Terrain
- Vocation
- Altitude
- Temperature
- Humidity
- Tampering/Malmaintenance

$$\mathbf{Power = Mg\ dV/dt + 0.5\rho A C_D V^3 + \mu MgV + MgV \sin(\theta)}$$

CONTINUOUS MASS PARTICULATE MEASUREMENT

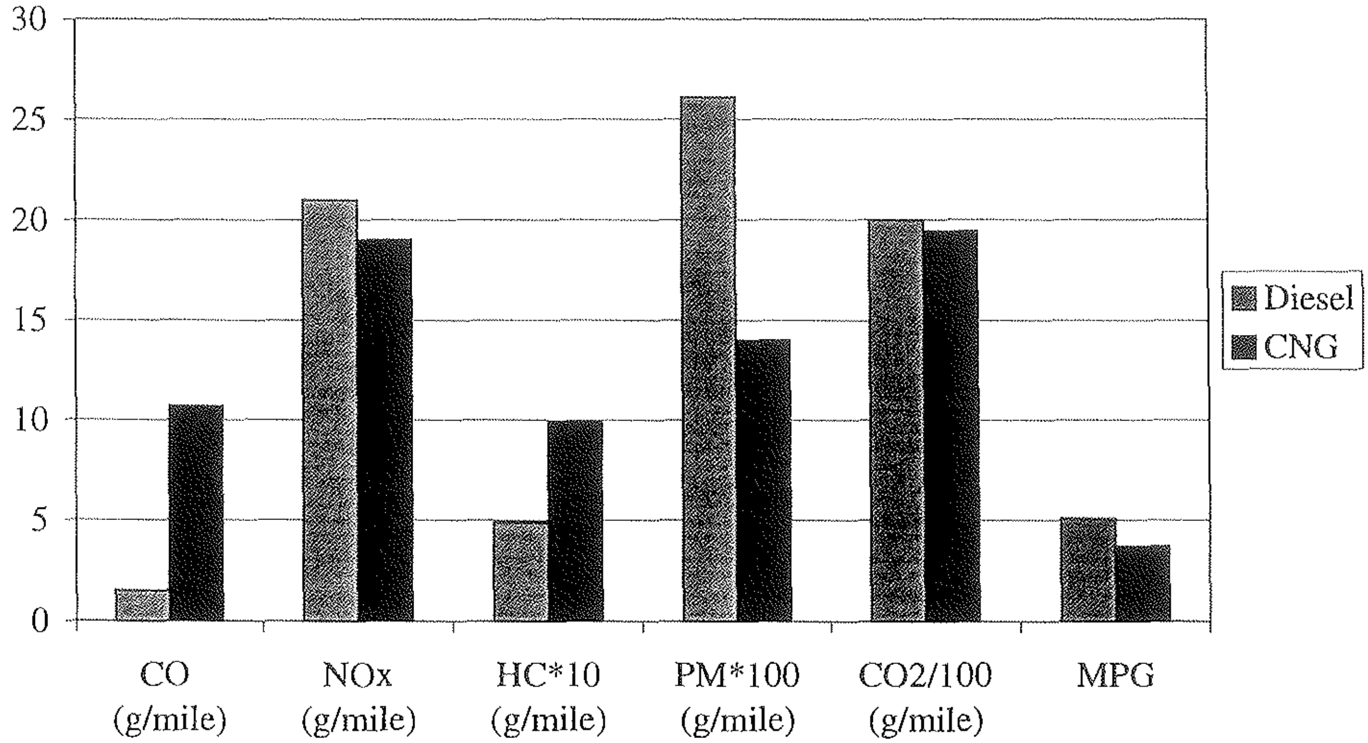


Particulate Matter data measured using a TEOM (Tapered Element Oscillating Microbalance) from a transit bus tested using the Central Business District test schedule. Availability of continuous data will permit superior inventory modeling.

ALTERNATIVE FUELS

NATURAL GAS FUELED HEAVY DUTY VEHICLES

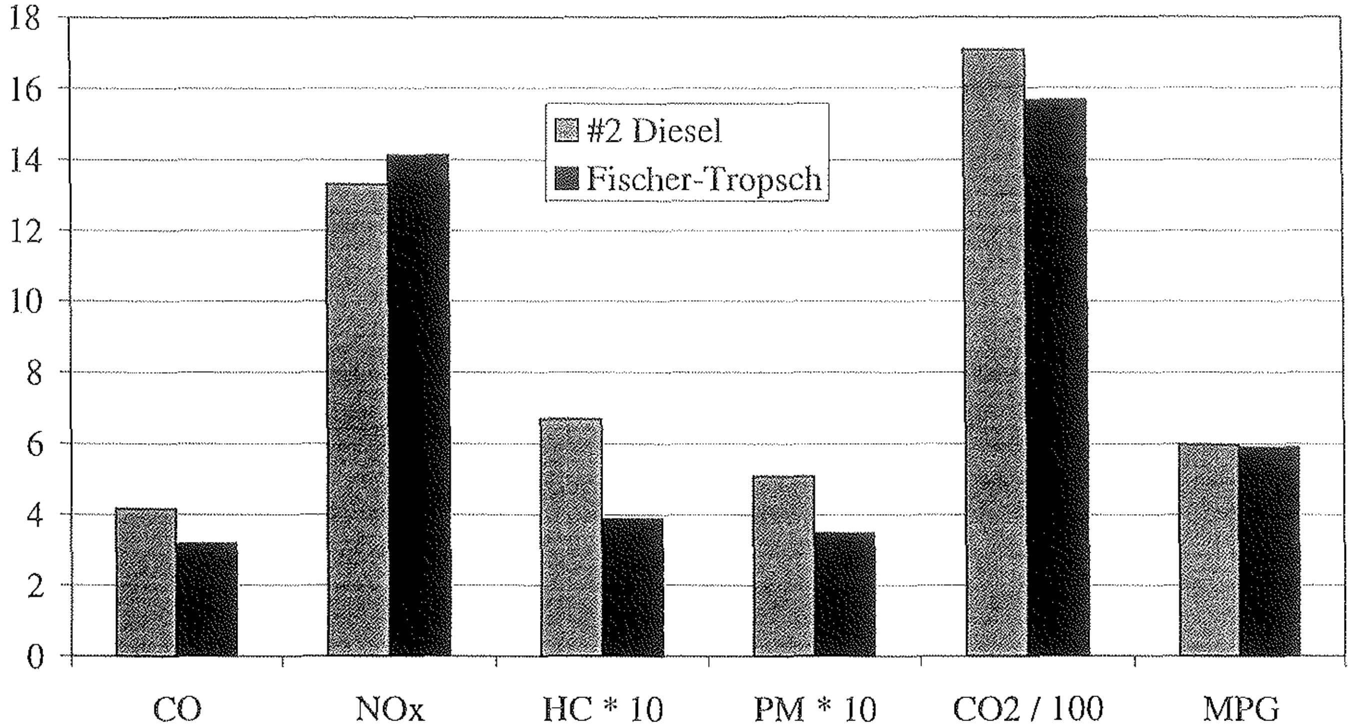
- Significant reduction in particulate emissions
- Reduction in toxic emissions associated with hydrocarbon and particulate emissions



Data from diesel and compressed natural gas (CNG) fueled school buses
(see ASME Spring ICE meeting)

FISCHER-TROPSCH DIESEL FUEL

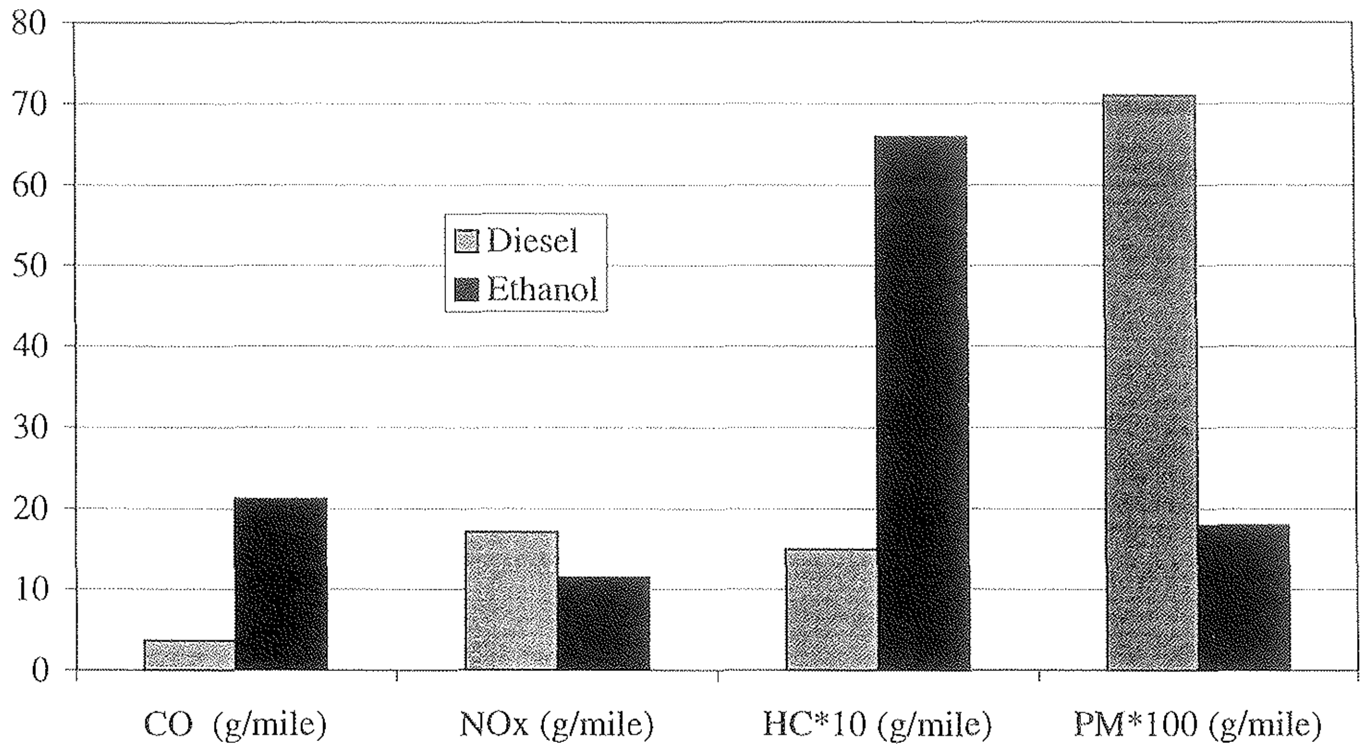
- Manufactured from natural gas or coal
- Much lower aromatic content than #2 diesel
- Near-zero sulfur content



Average data from several fleet vehicles tested on both #2 diesel and Fischer-Tropsch fuels (Recent data, WVU Transportable Laboratories, see also SAE 981393)

ALCOHOL FUELS

Ethanol offers the ability to reduce NO_X but emits unburned ethanol and aldehydes



Emissions data from tractors tested over the WVU 5 peak cycle using ethanol and diesel fuels

CONCLUSIONS

- Diesel engine technology is evolving rapidly to meet mass emissions standards
- In-use emissions do not always reflect certification emissions levels
- Heavy duty vehicle emissions inventory estimation is in disarray
- Alternative fuels offer advantages, but also carry burdens
- Risks cannot be assessed without quantitative, realistic projections of emissions

Characterization of Passenger Car Emissions

Peter Kohoutek (presented by Michael Spallek)

Exhaust Gas Components of Modern Diesel Passenger Cars

Peter Kohoutek, VOLKSWAGEN AG

In this paper the emissions of regulated and unregulated exhaust gas components of a fleet of diesel passenger cars measured at Volkswagen in the eighties are compared with the results of a new investigation on modern direct-injection diesel vehicles. The potential of improved diesel fuels to reduce emissions is also examined. It is presented as an emission comparison of modern TDI engines using a European reference fuel and a diesel fuel enjoying tax privileges in Sweden. This so-called Sweden diesel is almost completely free of sulfur and contains very low levels of aromatics.

As a result of the systematic further development of Volkswagen and Audi diesel engines, it has been possible to reduce the emissions of regulated exhaust gas components as well as fuel consumption and the related emission of carbon dioxide significantly over the last years. The emissions of carbon monoxide have been reduced by more than 90%, those of nitrogen oxides and carbon dioxide by 30% each, and the emissions of particulates and total hydrocarbons by 80% each.

As was to be expected, improved engine and exhaust gas after-treatment technology has also had a positive influence on the emissions of unregulated exhaust gas components. Besides the insignificant increase in methane, it has been possible to reduce the critical emissions of benzene by more than 80%, those of ethene by 70% and those of aldehydes and ketones by at least 50%. The emissions of 1,3-butadiene were virtually undetectable in the exhaust gas of modern diesel passenger cars. The polycyclic aromatic hydrocarbons and PAH nitro derivatives adsorbed to the diesel particulates have been reduced especially effectively by around 95% in the case of modern direct-injection diesel engines with oxidation-type catalytic converters.

The already known positive effects of Sweden diesel fuel on the emissions of regulated components were confirmed. With largely unchanged carbon dioxide and nitrogen oxide emissions, total hydrocarbons fell by more than 10%, carbon monoxide by more than 40% and particulate emissions by almost 30%.

The fuel also has a significant influence on the emissions of unregulated exhaust gas components.

Using Sweden diesel reduced the ozone forming potential by a third. The emissions of the PAH contained in the MAK List fell by 55% and the nitro-PAH by as much as 70%. The emission of benzene, which is already very low in modern diesel passenger cars, was reduced again by about 30%, of formaldehyde by around 40% and of acetaldehyde by about 35%. 1,3-butadiene could not be detected in the exhaust gas of Sweden diesel. The absolute number of particulates emitted by modern TDI engines is reduced by about 30% by the use of Sweden diesel.

It can be seen from the results of the investigation program that fuel quality which is currently found in the Sweden diesel should be promoted for environmental reasons.

A large scale improvement in fuel quality would have an immediate effect on reducing the emissions of all vehicles on the road and not, as in the case of technical vehicle improvements, only after a period of 10 to 15 years in the course of which these improvements became established into the vehicle stock.

HEI DIESEL WORKSHOP
March 7-9, 1999, Stone Mountain, Georgia

Exhaust Gas Components of Modern Diesel Passenger Cars

Dr. Peter Kohoutek

VOLKSWAGEN AG Group Research and Development

Unregulated Exhaust Gas Components
of Modern Diesel Passenger Cars

VOLKSWAGEN AG Group Research and Development

Exhaust Gas Components

Regulated	Unregulated
CO ΣHC NO_x $(NO+NO_2)$ Particulate Mass	All Other Components
Formaldehyde NMHC NMOG	

VOLKSWAGEN AG Group Research and Development

Unregulated Exhaust Gas Components of Modern Diesel Passenger Cars

1. Introduction
2. **Investigation Program Definition**
3. Measurement Results
   - Vehicle Comparison
   - Fuel Comparison
4. Summary

VOLKSWAGEN AG Group Research and Development

Task Definition
of the New Investigation Program

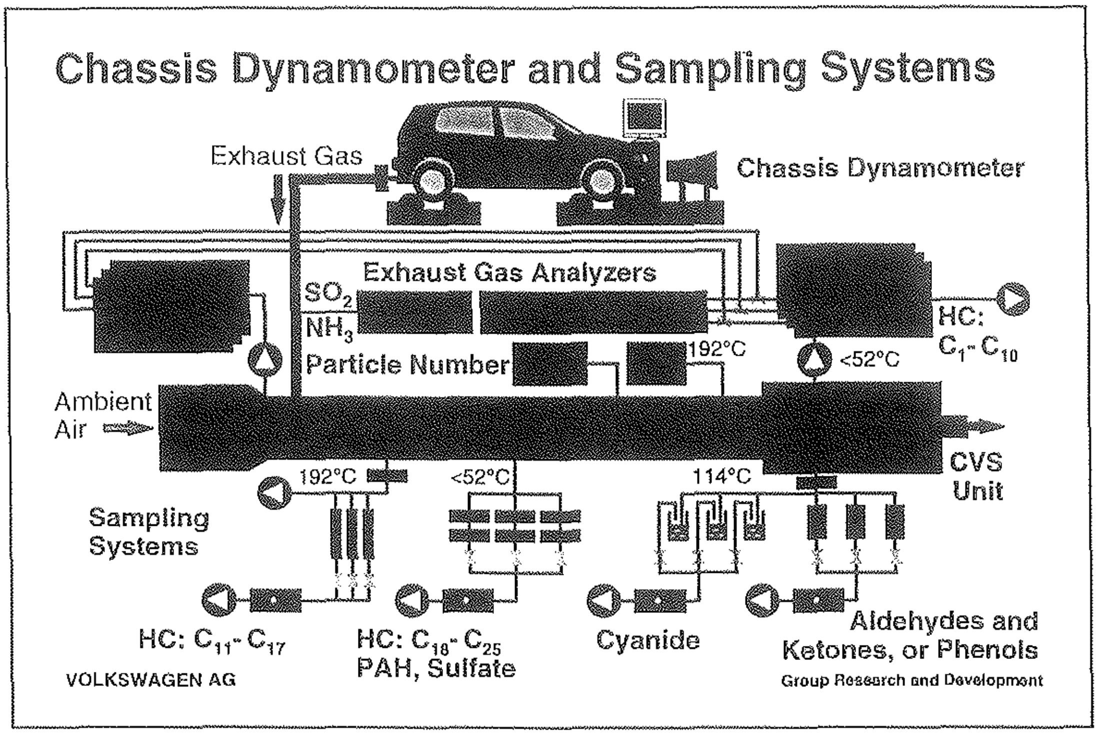
- How have the emissions of unregulated exhaust gas components of diesel production cars changed in the last years?
- How does improved diesel fuel influence the emissions of unregulated exhaust gas components?

VOLKSWAGEN AG Group Research and Development

Investigation Program Set-up

FTP 75	Vehicle Comparison: 7 Diesel Cars (US Version) Model Year 1978 - 1985	Vehicle Comparison: 2 Diesel Cars (EURO II) Model Year 1997	Fuel Comparison
ECE Reference Diesel	✔	✔	
Sweden Urban Diesel EC1	–	✔	

VOLKSWAGEN AG Group Research and Development



Unregulated Exhaust Gas Components
of Modern Diesel Passenger Cars

1. Introduction
2. Investigation Program Definition
3. Measurement Results
   - Vehicle Comparison
   - Fuel Comparison
4. Summary

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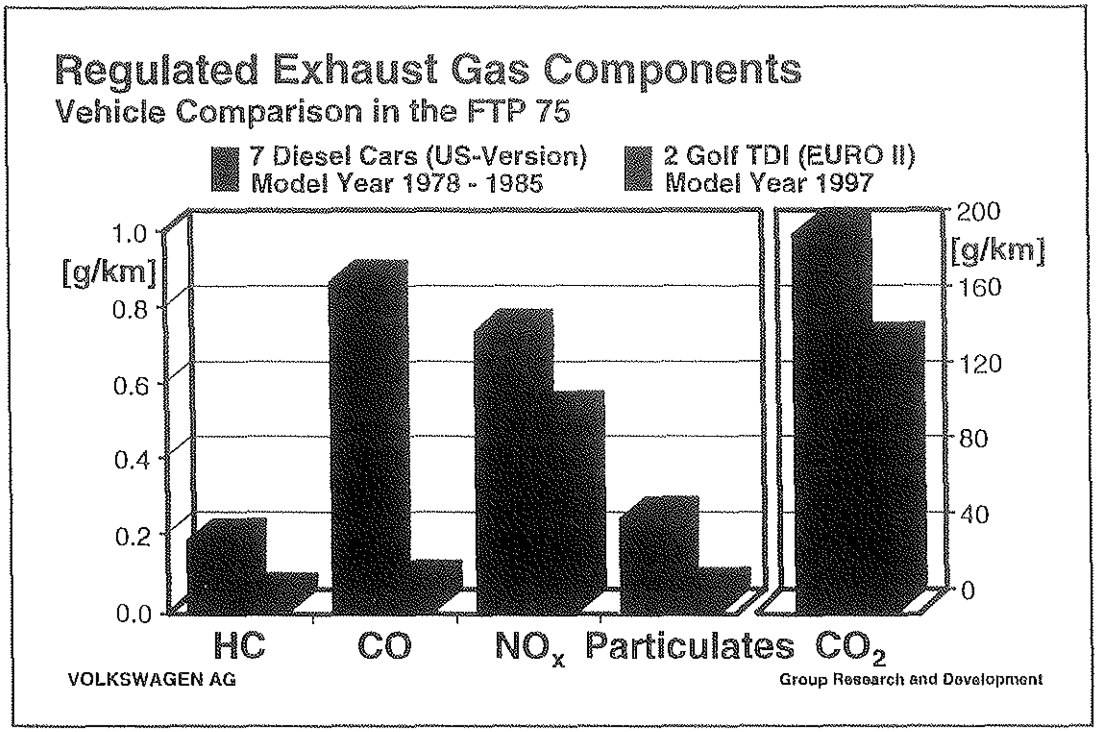
Regulated Exhaust Gas Components
Vehicle Comparison in the FTP 75
7 Diesel Cars (US-Version) Model Year 1978 - 1985
2 Golf TDI (EURO II) Model Year 1997
1.0
[g/km]
0.8
0.6
0.4
0.2
0.0
200
[g/km]
160
120
80
40
0
HC
CO
NOx
Particulates
CO2
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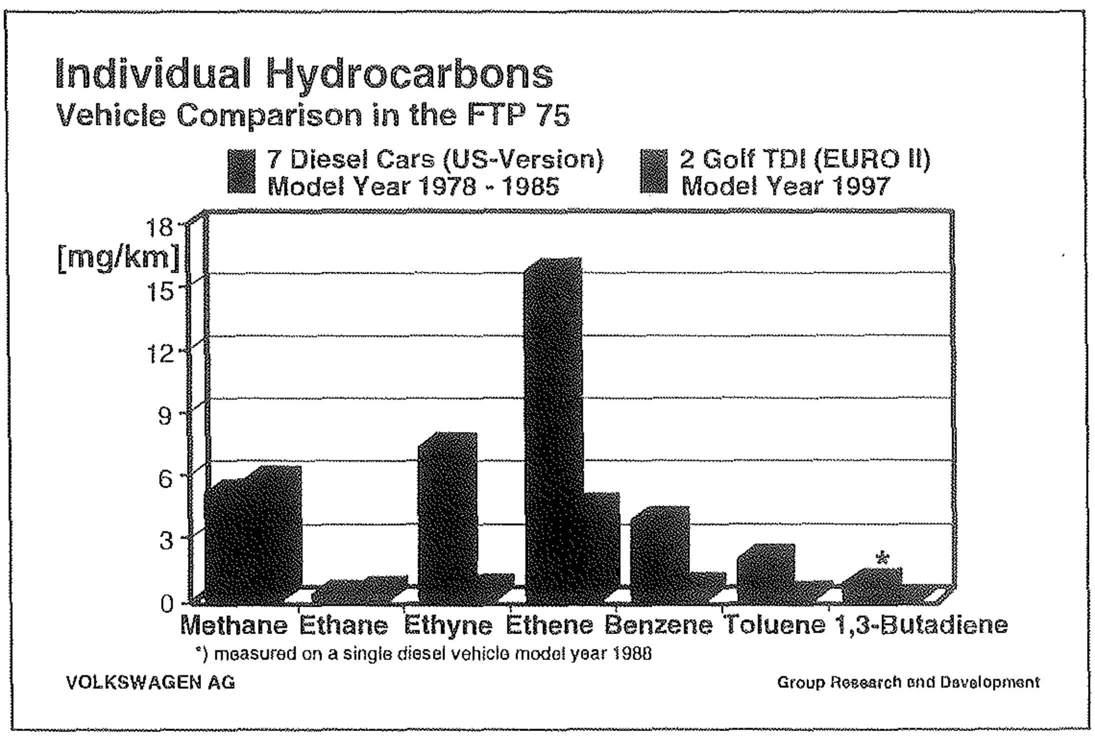
Individual Hydrocarbons
Vehicle Comparison in the FTP 75
7 Diesel Cars (US-Version) Model Year 1978 - 1985
2 Golf TDI (EURO II) Model Year 1997
18
[mg/km]
15
12
9
6
3
0
*
Methane
Ethane
Ethyne
Ethene
Benzene
Toluene
1,3-Butadiene
*) measured on a single diesel vehicle model year 1988
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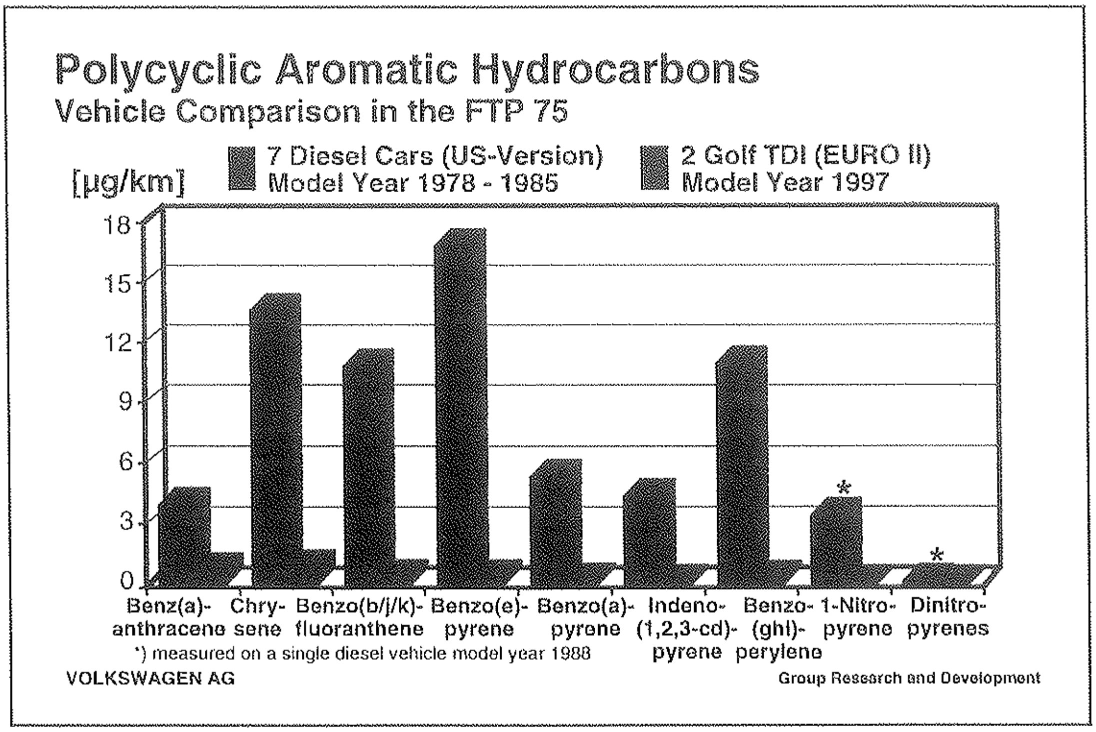
Polycyclic Aromatic Hydrocarbons
Vehicle Comparison in the FTP 75
7 Diesel Cars (US-Version) Model Year 1978 - 1985
2 Golf TDI (EURO II) Model Year 1997
[µg/km]
18
15
12
9
6
3
0
*
*
Benz(a)-anthracene
Chry-sene
Benzo(b/j/k)-fluoranthene
Benzo(e)-pyrene
Benzo(a)-pyrene
Indeno-(1,2,3-cd)-pyrene
Benzo-(ghi)-perylene
1-Nitro-pyrene
Dinitro-pyrenes
*) measured on a single diesel vehicle model year 1988
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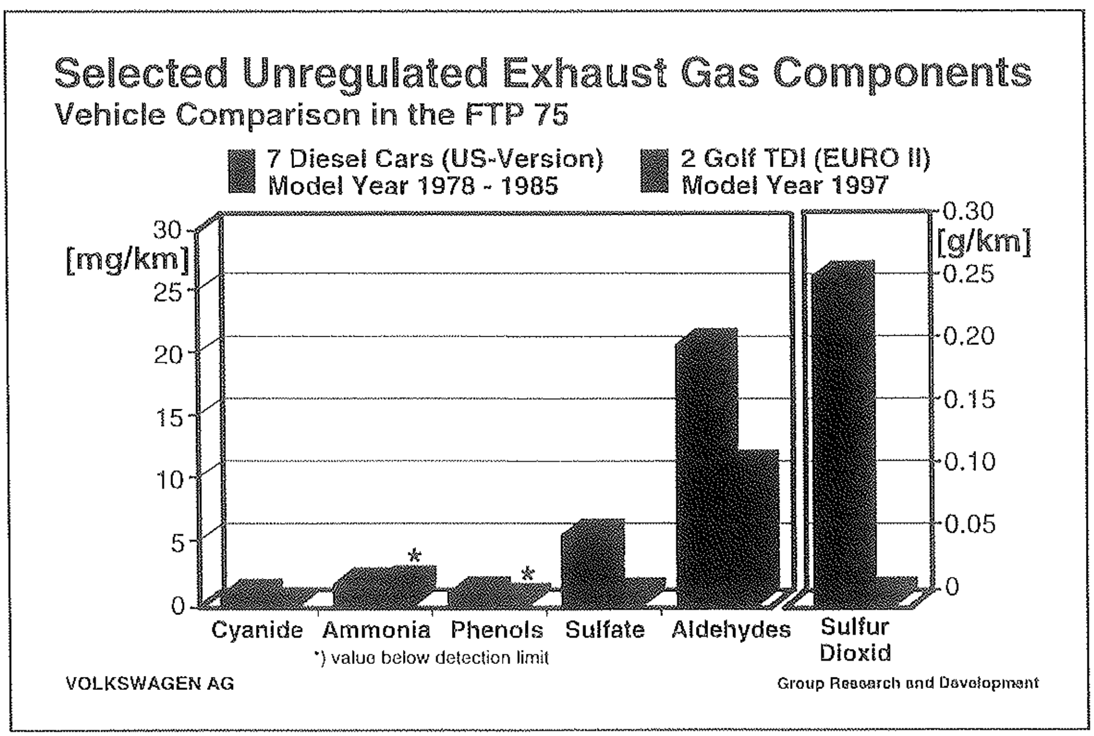
Selected Unregulated Exhaust Gas Components
Vehicle Comparison in the FTP 75
7 Diesel Cars (US-Version) Model Year 1978 - 1985
2 Golf TDI (EURO II) Model Year 1997
[mg/km]
30
25
20
15
10
5
0
0.30
[g/km]
0.25
0.20
0.15
0.10
0.05
0
*
*
Cyanide
Ammonia
Phenols
Sulfate
Aldehydes
Sulfur Dioxid
*) value below detection limit
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Unregulated Exhaust Gas Components
of Modern Diesel Passenger Cars

1. Introduction
2. Investigation Program Definition
3. **Measurement Results**
   - Vehicle Comparison
   - **Fuel Comparison**
4. Summary

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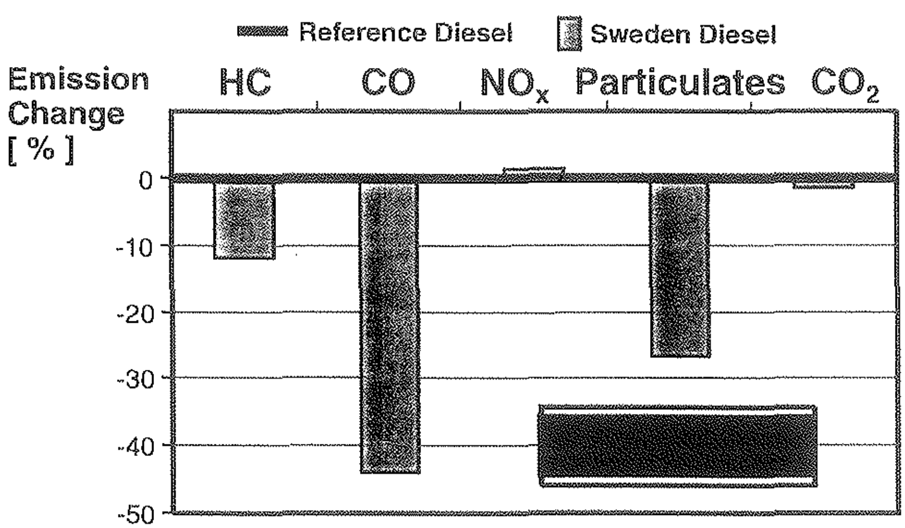
Regulated Exhaust Gas Components
Fuel Comparison in the FTP 75



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Assessment Criteria
for the Emissions Comparison of Different Diesel Fuels

- Maximum Ozone Forming Potential
- Polycyclic Aromatic Hydrocarbons (PAH)
- PAH Nitro Derivatives
- Air Toxic Components
- Particulate Number and Size Distribution

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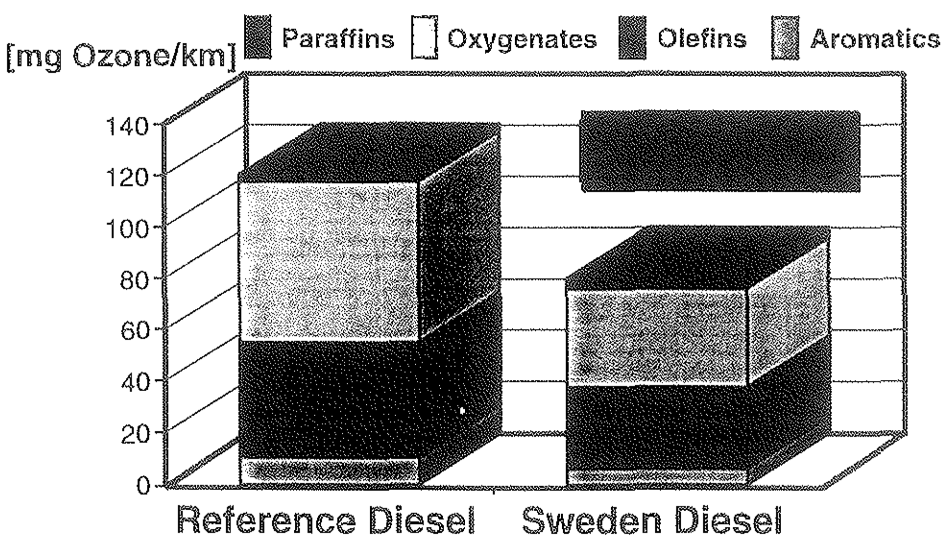
Maximum Ozone Forming Potential
Fuel Comparison in the FTP 75



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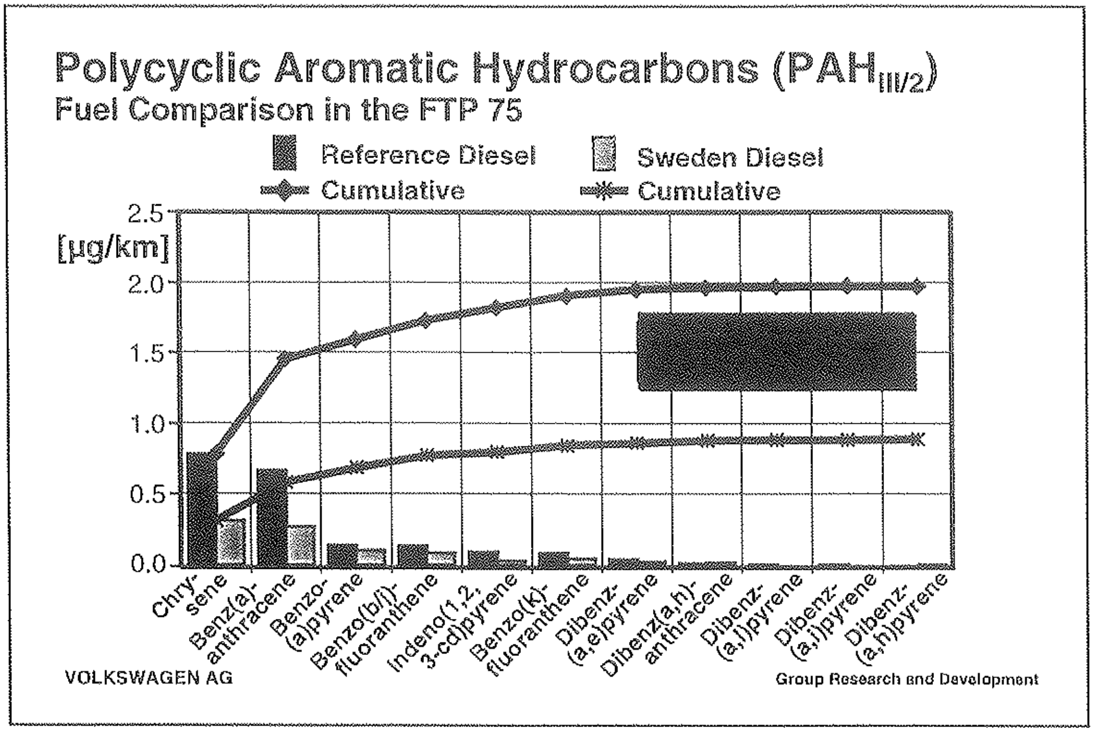
Polycyclic Aromatic Hydrocarbons (PAH$_{III/2}$)
Fuel Comparison in the FTP 75
Reference Diesel
Sweden Diesel
Cumulative
Cumulative
2.5
[µg/km]
2.0
1.5
1.0
0.5
0.0
[redacted]
Chry-sene
Benz(a)-anthracene
Benzo-(a)pyrene
Benzo(b/j)-fluoranthene
Indeno(1,2,3-cd)pyrene
Benzo(k)-fluoranthene
Dibenz-(a,e)pyrene
Dibenz(a,h)-anthracene
Dibenz-(a,i)pyrene
Dibenz-(a,l)pyrene
Dibenz-(a,h)pyrene
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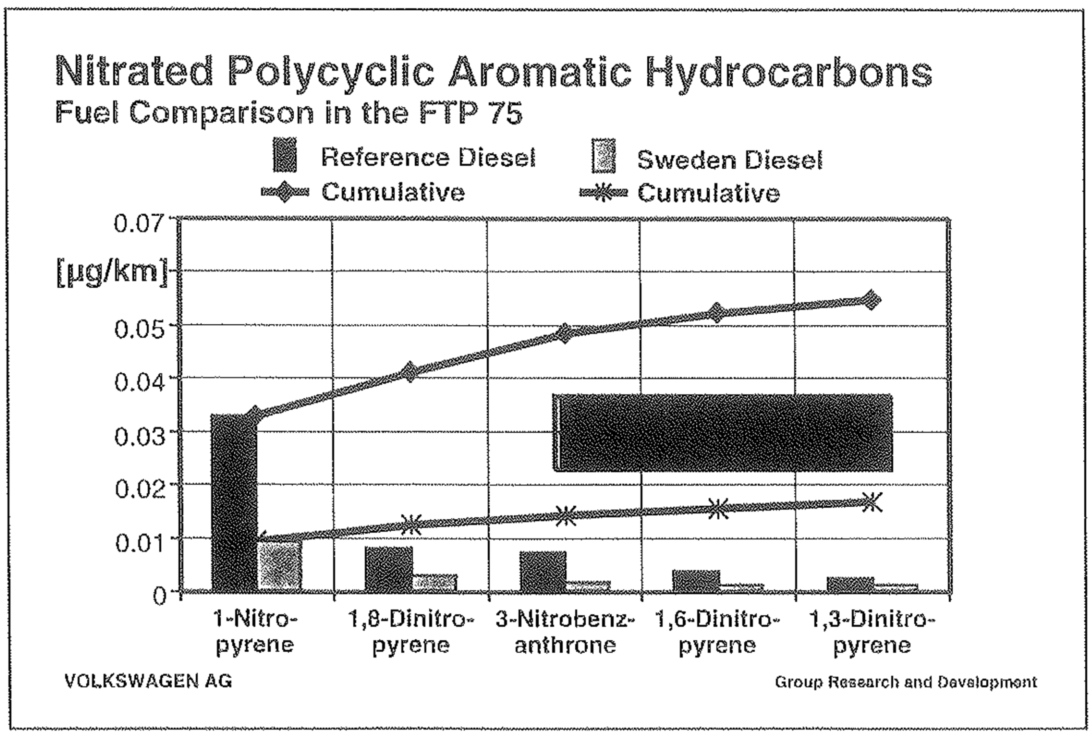
Nitrated Polycyclic Aromatic Hydrocarbons
Fuel Comparison in the FTP 75
Reference Diesel
Sweden Diesel
Cumulative
Cumulative
0.07
[µg/km]
0.05
0.04
0.03
0.02
0.01
0
[redacted]
1-Nitro-pyrene
1,8-Dinitro-pyrene
3-Nitrobenz-anthrone
1,6-Dinitro-pyrene
1,3-Dinitro-pyrene
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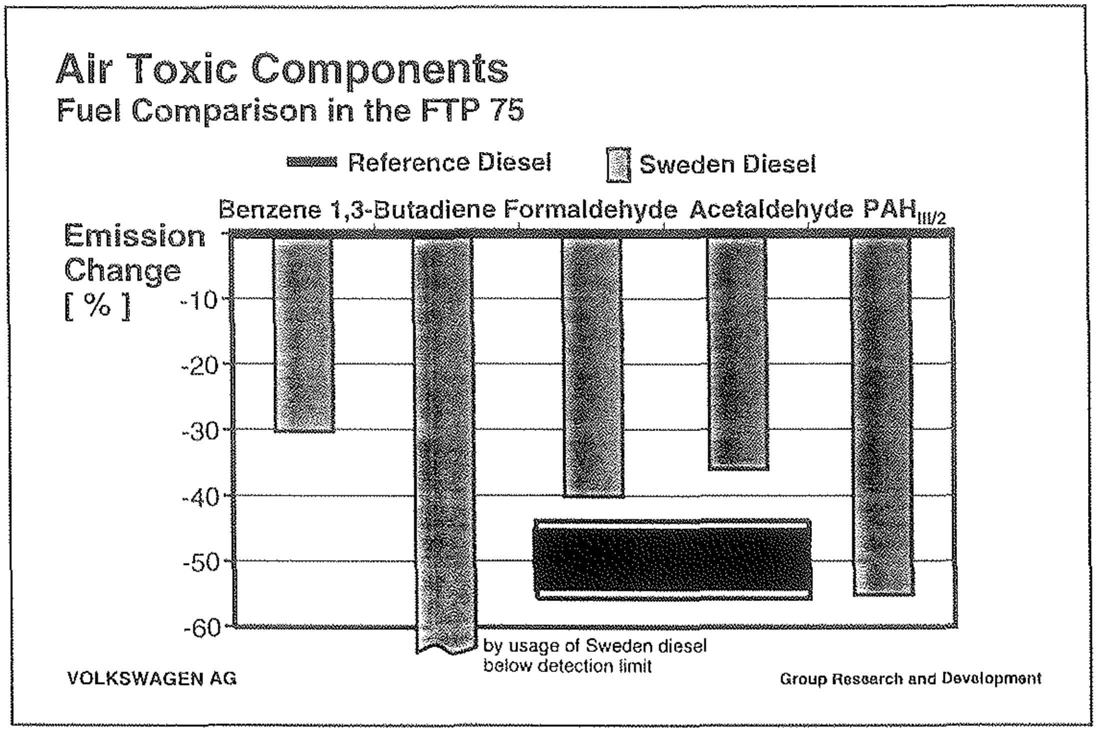
Air Toxic Components
Fuel Comparison in the FTP 75
Reference Diesel
Sweden Diesel
Benzene 1,3-Butadiene Formaldehyde Acetaldehyde PAH III/2
Emission Change [%]
-10
-20
-30
-40
-50
-60
[redacted]
by usage of Sweden diesel below detection limit
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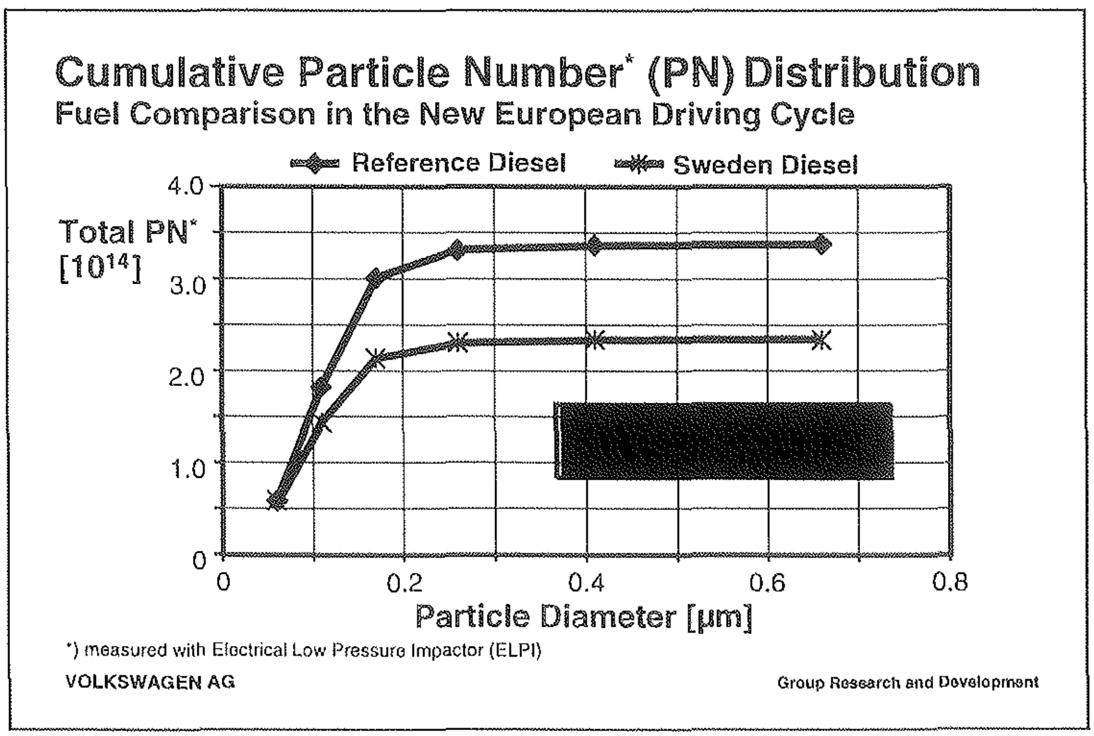
Cumulative Particle Number* (PN) Distribution
Fuel Comparison in the New European Driving Cycle
Reference Diesel
Sweden Diesel
Total PN* [10^14]
4.0
3.0
2.0
1.0
0
0
0.2
0.4
0.6
0.8
Particle Diameter [µm]
[redacted]
*) measured with Electrical Low Pressure Impactor (ELPI)
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Unregulated Exhaust Gas Components of Modern Diesel Passenger Cars

1. Introduction
2. Investigation Program Definition
3. Measurement Results
   - Vehicle Comparison
   - Fuel Comparison
4. Summary

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Summary - Vehicle Comparison* (1/3)

CO	– 90 %
NO_x	– 30 %
HC	– 80 %
Particulates	– 80 %
CO_2	– 30 %
Methane	+ 10 %
Benzene	– 80 %
Ethene	– 70 %
Aldehydes	– 50 %
1,3-Butadiene	≤ LOD
PAH	– 95 %

*) emission changes between 7 Model Year 1978 - 1986 and 2 Model Year 1997 diesel cars (ECE reference diesel, FTP 75)

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Summary - Fuel Comparison* (2/3)

CO	– 45 %
NOx	± 0 %
HC	– 10 %
Particulates	– 25 %
CO2	± 0 %
Ozone Forming Potential	– 35 %
Benzene	– 30 %
Formaldehyde	– 40 %
Acetaldehyde	– 35 %
1,3-Butadien	< LOD
$PAH_{III/2}$	– 55 %
PAH Nitro Derivatives	– 70 %
Particulate Number	– 30 %

*) emission changes by the use of Sweden diesel compared to ECE reference fuel (2 Golf TDI, FTP 75)

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Conclusion (3/3)

Fuel Quality
Which is Currently Found
in the Sweden Urban Diesel EC1
Should be Promoted
for Environmental Reasons

VOLKSWAGEN AG Group Research and Development

Changes in Diesel Engine Emissions

Barbara Zielinska

Changes in Diesel Engine Emissions over the Last Two Decades

Barbara Zielinska
Desert Research Institute, Reno, NV 89506

Abstract

Major research programs were carried out in the late 1970s and early 1980s to ascertain the physical and chemical characteristics of emissions from diesel engines and the biological effects of these emissions. New control technologies that have been introduced over the past 25 years have substantially reduced diesel emissions. The introduction of new, more stringent NO_x and particulate matter emission standards for heavy-duty diesel engines provides incentive for engine manufacturers and fuel producers to develop technology to meet emission reduction requirements. The varying chemical composition of diesel emissions (i.e., older engines vs. newer-technology ones, various emission controls, reformulated fuel, heavy-duty vs. light-duty engines, and engines operated under varied conditions) gives rise to questions regarding the applicability of recent risk assessment findings that are based on pre-1990s engines to present-day engine exhaust emissions.

This presentation examines the changes that have occurred over the last two decades in the chemical characteristic of heavy-duty diesel engine emissions. Changes in emission rates of particulate matter (PM), organic and elemental carbon, PAH and nitro-PAH are reviewed. Data obtained from heavy-duty diesel engine dynamometer tests (expressed in mass/work units) are compared with those data obtained from the chassis dynamometer tests (expressed in mass/travel distance units) in order to demonstrate the influence of real use factors on these emissions. While newer technology, various emission control measures, and fuel reformulation tend to reduce mutagenicity of diesel exhaust and mass emission rates of certain exhaust components, such as PM, sulfates, PAH, and nitro-PAH, these changes are less pronounced when expressed in mass/distance units than in mass/work units.

References

Chow J.C., J.G. Watson, L.W. Richards, D.L. Haase, C. McDade, D.L. Dietrich, D. Moon and C. Sloane (1991). "The 1989-90 Phoenix PM_{10} Study. Volume II: Source Apportionment." DRI Document No. 8931.6F1, prepared for Arizona Department of Environmental Quality, Phoenix, AZ, by Desert Research Institute, Reno, NV.

Copper J.A., D.C. Redline, J.R. Sherman, L.M. Valdovinos, W.L. Pollard, L.C. Scvone and C. Badgett-West (1987). "PM_{10} Source Composition Library for the South Coast Air Basin. Volume I: Source Profile Development." South Coast Air Quality Management District, El Monte, CA.

Dietzmann, H.E., M.A. Parness, and R.L. Bradow (1981). "Emissions from gasoline and diesel delivery trucks by chassis transient cycle." Paper No. 81-DGP-6, presented at ASME Energy-Sources Technology Conference and Exhibition, January 1981, Houston, TX. American Society of Mechanical Engineers, New York.

Graboski, M.S., R.L. McCormick, J. Yanowitz and L. Ryan (1998). "Heavy-duty diesel vehicle testing for the Northern Front Range Air Quality Study." Final Report prepared for Colorado State University by the Colorado Institute for Fuels and High-Altitude Engine Research, Colorado School of Mines, Golden, CO.

Gertler, A.W., E.M. Fujita, W.R. Pierson and D.N. Wittorff (1996). "Apportionment of NMHC tailpipe vs non-tailpipe emissions in the Fort McHenry and Tuscarora Tunnels." *Atmos. Environ.*, **30**, 2297-2305.

Houck J.E., J.C. Chow, J.G. Watson, C.A. Simons, L.C. Pritchett, L.C. Goulet and C.A. Frazier (1989). "Determination of particle size distribution and chemical composition of particulate matter from selected sources in California. Volume I", prepared for California Air Resource Board, Sacramento, CA, by OMNI Environmental Services, Beaverton, OR, and Desert Research Institute, Reno, NV.

Norbeck, J.M., T.J. Truex, M.R. Smith, J. Arey, N. Kado, B. Okamoto, K. Keifer, P. Kuzmicky and I. Holcomb (1998). "Evaluation of factors that affect diesel exhaust toxicity." Final Report, prepared under Contract No. 94-312, for California Air Resources Board, by Center for Environmental Research and Technology, College of Engineering, University of California, Riverside, CA.

Pierson, W.R., and W.W. Brachaczek (1983). "Particulate matter associated with vehicles on the road. II." *Aerosol Sci. Technol.*, **2**, 1-40.

Rogge, W., L.M. Hildemann, M.A. Mazurek, and G.R. Cass (1993). "Sources of fine organic aerosol. 2. Noncatalyst and catalyst-equipped automobiles and Heavy-duty diesel trucks." *Environ. Sci. Technol.*, **27**, 636-651.

Sawyer, R.F., and J.H. Johnson (1995). "Diesel emissions and control technology." In *Diesel Exhaust: A Critical Analysis of Emissions, Exposure, and Health Effects.* A Special Report of the Institute's Diesel Working Group. Health Effects Institute, Cambridge, MA, pp. 65-81.

Schuetzle, D., and J.M. Perez (1983). "Factors influencing the emissions of nitrated-polynuclear aromatic hydrocarbons (nitro-PAH) from diesel engines." *J. Air Pollut. Control Assoc.*, **33**, 751-755.

Schuetzle, D., and J.A. Frazier (1986). "Factors influencing the emission of vapor and particulate phase components from diesel engines." In *Carcinogenic and Mutagenic Effects of Diesel Engine Exhaust: Proceedings of the International Satellite Symposium on Toxicological Effects of Emissions from Diesel Engines,* July 1986, Tsukuba Science City, Japan, N. Ishinishi, A. Koizumi, R.O. McClellan, and W. Stöber, eds. (Developments in Toxicology and Environmental Science, Vol. 13). Elsevier Science Publishers B.V., Amsterdam, The Netherlands, pp. 41-63.

Steenland, K., J. Deddens, and L. Stayner (1998). "Diesel exhaust and lung cancer in the trucking industry: Exposure-response analyses and risk assessment." *Am. J. Ind. Med.*, **34**, 220-228.

Watson, J.G.; Chow, J.C.; Richards, L.W.; Anderson, S.R.; Houck, J.E.; Dietrich, D.L. "The 1987-88 Metro Denver Brown Cloud Air Pollution Study, Volume II: Measurements." DRI Document No. 8810.1F2, prepared for the Greater Denver Chamber of Commerce, Denver, CO, by Desert Research Institute, Reno, NV, 1988.

Watson, J., E. Fujita, J. Chow, B. Zielinska, L. Richards, W. Neff and D. Dietrich (1998). *Northern Front Range Air Quality Study Final Report.* Prepared for Colorado State University, Fort Collins, CO, April 1998.

Zielinska, B., D. Lowenthal and T. Hayes (1997). "The characterization of motor vehicle emissions." Final report prepared for U.S. Environmental Protection Agency, Office of Exploratory Research, Washington, DC, by Desert Research Institute, Reno, NV.

Changes in Diesel Engine Emissions Over the Last Two Decades

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Energy and Environmental Research Center
Reno, NV 89506

Comparison of heavy-duty diesel engine emissions is not straightforward:

- differences in testing procedures;
- differences in units emission rates are expressed

Testing procedures of heavy-duty diesel engine emissions:

- Engine dynamometers: the steady-state (13-mode) operating cycles (past), and the transient cycles (current);
- Chassis dynamometer (limited): EPA heavy-duty transient truck (HDT) cycle; central business district (CBD); West Virginia truck (WVT) cycle.

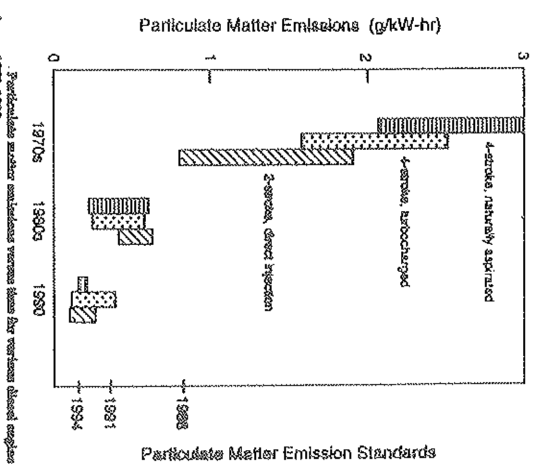
Units:

- Mass/work units: grams/brake-horsepower-hours (g/bhp-hr) or grams per kilowatt-hour (g/kW-hr)
  - 1kW = 1.341 hp
- Mass/travelled distance units (g/mile or g/km)
- Miscellaneous units: mass/PM or PM extract weight (mg/g), mass/volume of raw or diluted exhaust, etc.

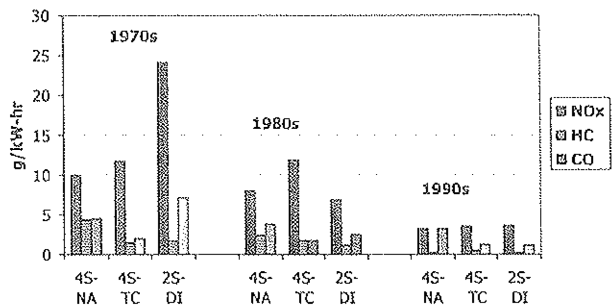
Federal Standards for Heavy-Duty Highway Diesel Engines and Urban Buses (g/bhp-hr)

Year [a]	CO	HC	NMHC + NO_x	NO_x	PM
1988-89	15.5	1.3		10.7	0.6
1990	15.5	1.3		6.0	0.6
1991-93	15.5	1.3		5.0	0.25; 0.1[b]
1994-97	15.5	1.3		5.0	0.1; 0.07[b]
1998+	15.5	1.3		4.0	0.1; 0.05[b]
2004+	15.5		2.4 or 2.5 with a limit of 0.5 on NMHC		0.1; 0.05[b]

a. Year refers to the date of diesel engine manufacture.
b. Standard for urban buses.



Gaseous Emissions as Functions of Time Period and Diesel Engine Type



Particulate Matter Emissions as Functions of Time Period and Diesel Engine Type

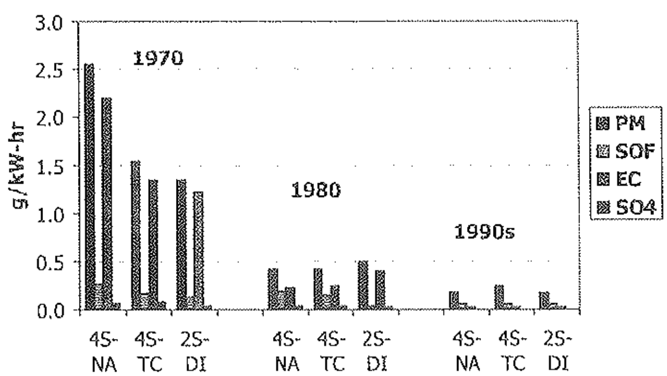


Table 1. Characteristic of Heavy-Duty Diesel Vehicles Tested on Chassis Dynamometer

veh_use	MYear	GVWR (lbs)	engine_mfg	odometer_mi	PM (mg/mile)	Source
City, 2 strokes	77	36,900	DDC	60,000	3.22	a
Van	79	27,500	Caterpillar	7,000	0.92	a
Tractor	79	80,000	Mack	69,000	1.95	a
Tractor	79	80,000	Cumin	26,000	1.65	a
RTD Bus	81	38,000	GMC	119,000	6.00	b
Water Truck	81	49,560	Cummins	17,867	7.43	b
Tel. Truck	83	80,000	Cummins	80,876	1.96	b
Dump Truck	84	80,000	Cummins	595,606	2.85	b
RTD Bus	86	38,000	DDC	66,780	0.65	b
Dump Truck	87	28,000	International	89,528	2.31	b
School Bus	88	28,000	GMC	89,054	1.80	b
Dump Truck	89	33,000	Navistar	101,925	2.53	b
RTD Truck	89	8,900	GMC	13,518	1.16	b
Food Delivery	90	33,000	International	142,242	1.49	b
Garbage Haule	90	50,000	Cummins	72,251	1.73	b
Food Delivery	91	80,000	Cummins	47,797	0.75	b
School Bus	91	30,000	Cummins	62,549	2.70	b
Lease	93	25,500	Navistar	122,406	1.26	b
RTD Bus	93	38,000	DDC	85,200	0.73	b
Lease	93	11,050	Isuzu	82,618	2.20	b
RTD Bus	93	38,000	DDC	85,200	0.61	b
Delivery	93	22,000	Isuzu	150,788	1.18	b
Concrete Mixe	93	60,000	Cummins	96,262	1.11	b
CDOT Truck	93	36,220	Navistar	37,009	0.72	b
Dump Truck	95	36,200	Navistar	5,320	0.80	b

a. From Dietzmann et al., 1981

b.Tests performed by Colorado School of Mines (Graboski et al., 1998) for NFRAQS

Steenland et al. (1998)

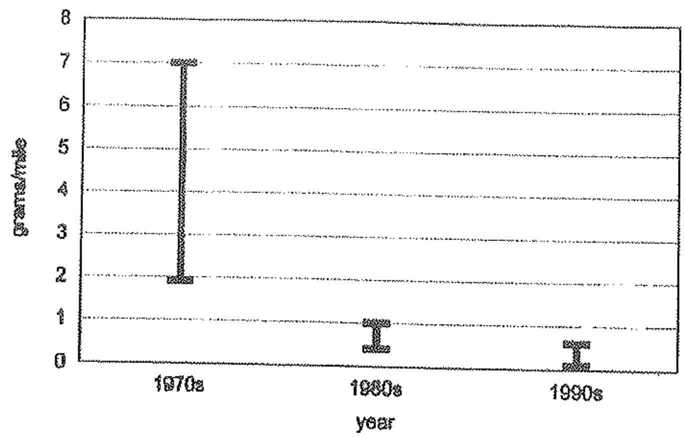
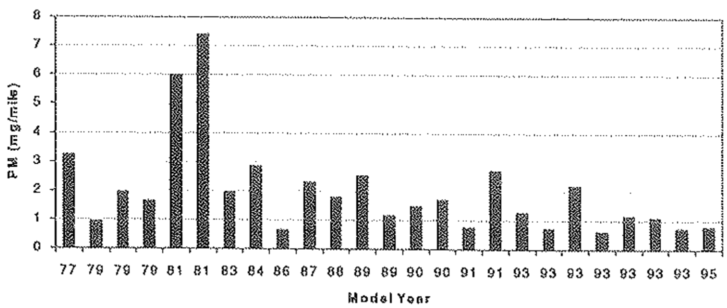


FIGURE 2. Estimated U.S. heavy duty engine particulate emissions (gm/mi) by decade (adapted from Sawyer and Johnson (1995)).

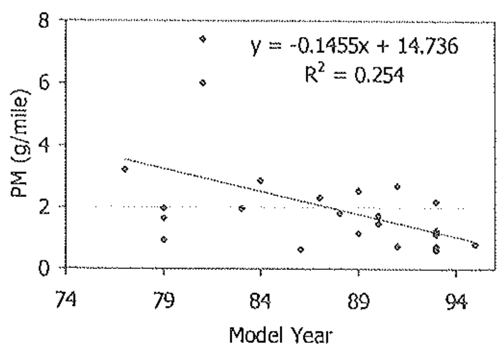
Eight factors that affect exhaust emissions from heavy duty vehicles:

- Vehicle weight;
- Driving activity;
- Vehicle vocation;
- Fuel composition;
- Engine exhaust aftertreatment;
- Vehicle age;
- Terrain traveled;
- Engine operation - injection timing

Heavy-Duty Diesel PM Emission Rates



PM Emissions as a Function of Model Year



Historical Trend in PM Emission Rates

	1970s	1980s	1990s
Dynamometer	1.93[a]	1.89[a]	1.27[a]
	3.53[b]	1.89[b]	1.02[c]
Tunnel	1.4[e]		0.67[d]

a. Data from Table 1, excluding 1981 vehicles
b. Data from Table 1, adding 1981 vehicles to 1970s group
c. Data from NFRAQS and from Gertler et al., 1995 (13 HDD, WVU cycle)
e. Pierson and Brachaczek, 1983: from Tuscarora, PA, tunnel.
d. Gertler et al., 1996: from Fort McHenry, Baltimore, tunnel.

Table 2. NFRAQS Heavy-Duty Diesel Test Summary[a]

Sample ID	Veh_use	MYear	GVWR	Engine_mfg	Odomet_mi[b]	Opacity(%)[c]	Cycle
HDD2	Food Delivery	90	33,000	International	142,242	27.6	WVT
HDD5	Lease	93	25,500	Navistar	122,406	16.3	CBD
HDD6	RTD Bus	93	38,000	DDC	85,200	10.7	CBD
HDD7	Replicate	93					HDTT
HDD8	Dump Truck	87	28,000	International	89,528	25	HDTT
HDD10	Lease	93	11,050	Isuzu	82,618	64.7	CBD
HDD12	School Bus	88	28,000	GMC	89,054	26.6	HDTT
HDD13	Replicate	88					CBD
HDD14	RTD Bus	93	38,000	DDC	85,200	5.8	HDTT
HDD15	Replicate	93					CBD
HDD16	Food Delivery	91	80,000	Cummins	47,797	46.8	CBD
HDD17	Dump Truck	84	80,000	Cummins	595,606	25.5	HDTT
HDD18	School Bus	91	30,000	Cummins	62,549	29.2	CBD
HDD24	Concrete Mixer	93	60,000	Cummins	96,262	6.9	WVT
HDD32	Garbage Hauler	90	50,000	Cummins	72,251	75.3	WVT

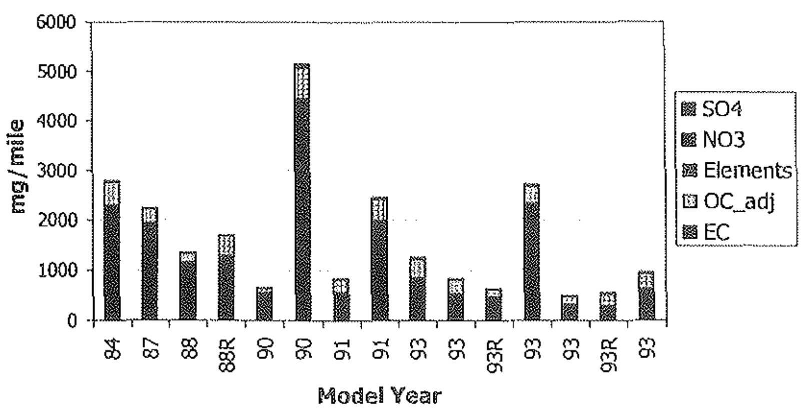
[a]Tests performed by Colorado School of Mines (CSM) Institute for Fuels and High Altitude Engine Research (CIFER), Graboski et al., 1998
[b]Since rebuilt [c]SAE J-1667 snap idle procedure

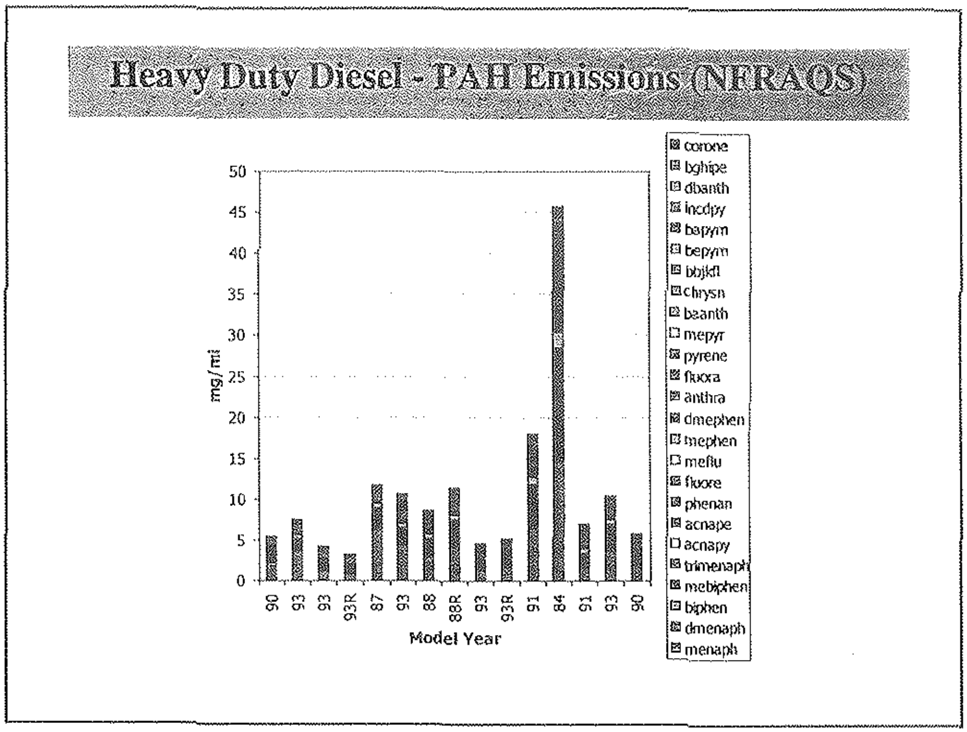
FRACTIONAL ORGANIC AND ELEMENTAL CARBON ABUNDANCES IN DIESEL EMISSIONS

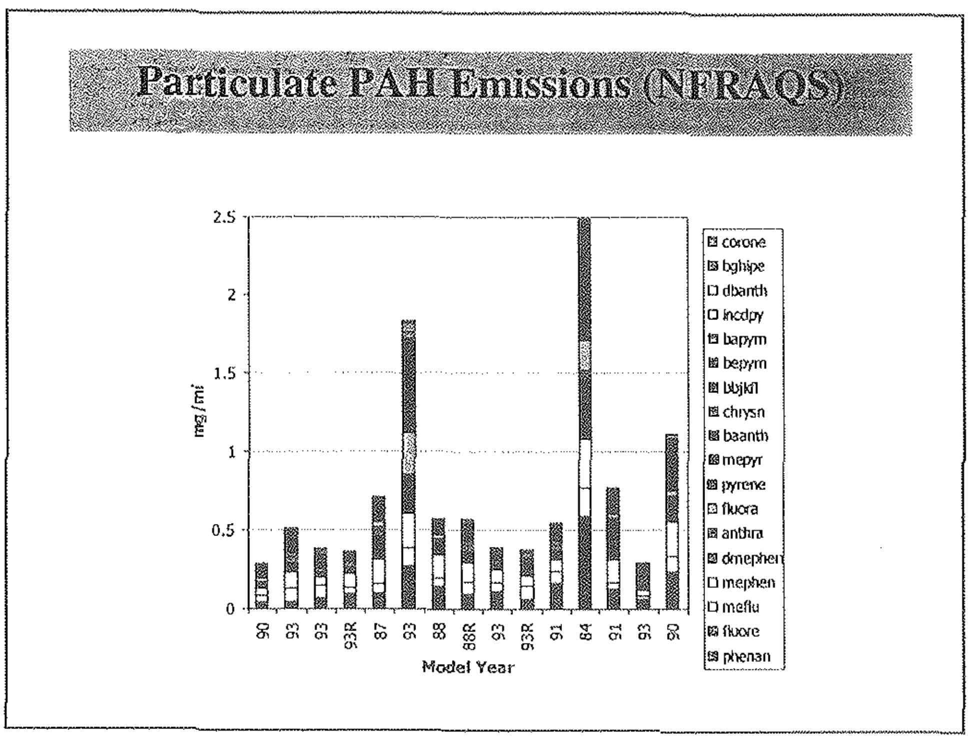
Location	Organic Carbon	Elemental Carbon	N[c]	Sources
Denver, CO[a]	19±8%	75±10%	15	NFRAQS, 1998
Denver, CO[a]	23±8%	74±21%	3	Watson et al., 1988
Los Angeles, CA[a]	36±3%	52±5%	2	Cooper et al., 1987
Los Angeles, CA[a]	33.6%	40.5%	2	Rogge et al., 1993
Bakersfield, CA[a]	49±13%	43±8%	3	Houck et al., 1989
Phoenix, AZ[b]	40±7%	22±8%	8	Chow et al., 1991
Phoenix, AZ[b]	64±11%	33±10%	7	Zielinska et al., 1997

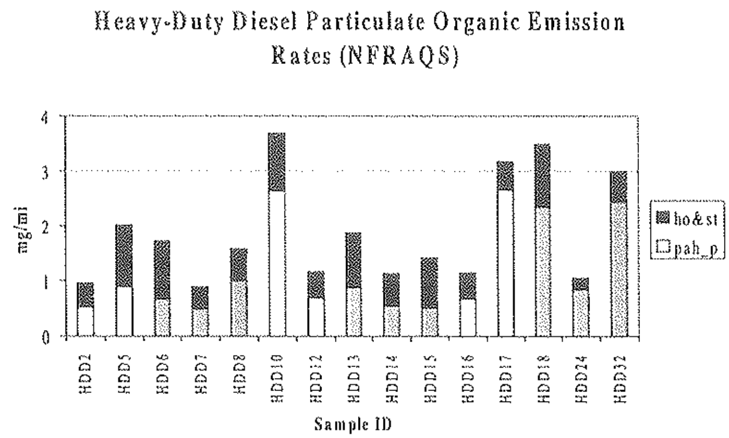
NOTES: (a) Modified Federal Test Procedures followed in dynamometer tests
(b) Roof monitoring at inspection station
(c) N = Number of samples

NFRAQS Heavy-Duty Diesel OC/EC Emissions

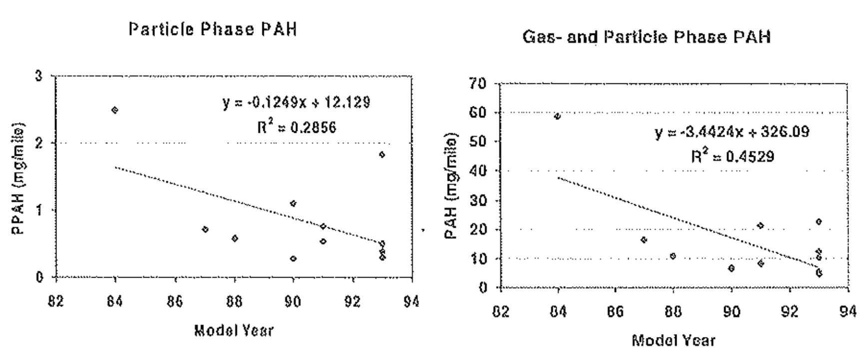



Heavy Duty Diesel - PAH Emissions (NFRAQS)
mg/mi
0
5
10
15
20
25
30
35
40
45
50
90
93
93
93R
87
93
88
88R
93
93R
91
84
91
93
90
Model Year
corone
bghipe
dbanth
incdpy
bapyrn
bepyrn
bbjkfl
chrysn
baanth
mepyr
pyrene
fluora
anthra
dmephen
mephen
meflu
fluore
phenan
acnape
acnapy
trimenaph
mebiphen
biphen
dmenaph
menaph


Particulate PAH Emissions (NFRAQS)
mg/mi
0
0.5
1
1.5
2
2.5
90
93
93
93R
87
93
88
88R
93
93R
91
84
91
93
90
Model Year
corone
bghipe
dbanth
incdpy
bapyrn
bepyrn
bbjkfl
chrysn
baanth
mepyr
pyrene
fluora
anthra
dmephen
mephen
meflu
fluore
phenan



PAH Emissions as a Function of Model Year



Emission Rates of Representative PAH from Heavy-Duty Diesel (in µg/mile)

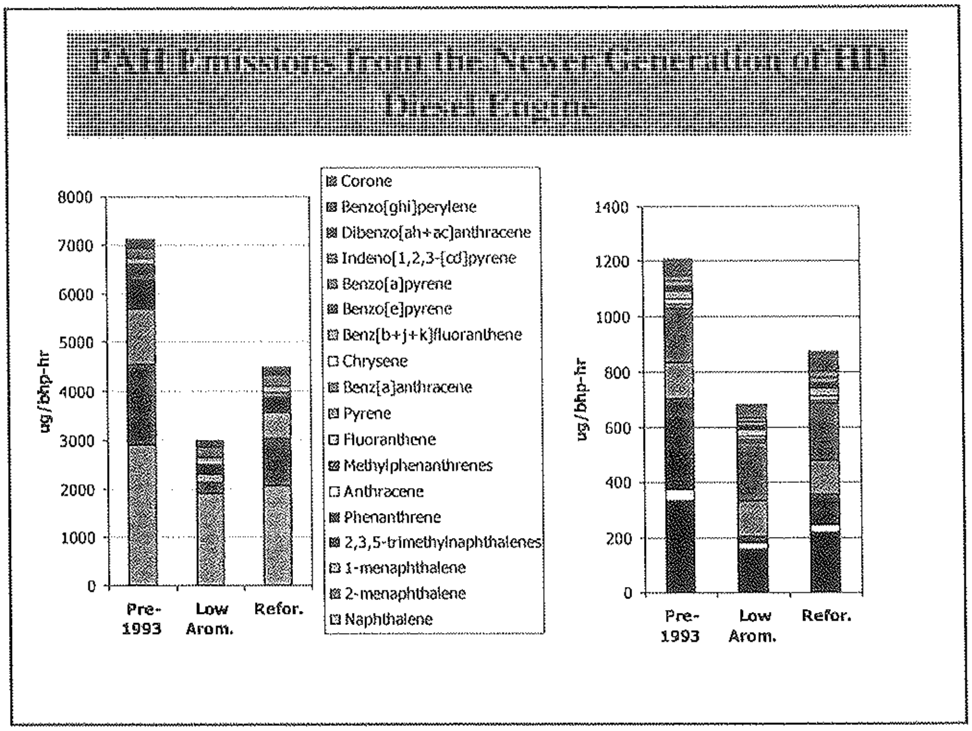
PAH	1970s	1980s	1990s
Pyrene		1762[b] 109.1[d]	72.1[d]
Fluoranthene		2173[b] 80.6[d]	49.1[d]
Benzo[a]pyrene	2.7[a]	54[b] 2.1[c] 12.2[d]	12.0[d]
Benzo[e]pyrene		64[b] 4.2[c] 12.3[d]	10.9[d]
1-Nitropyrene		45[b] 2.4[e]	

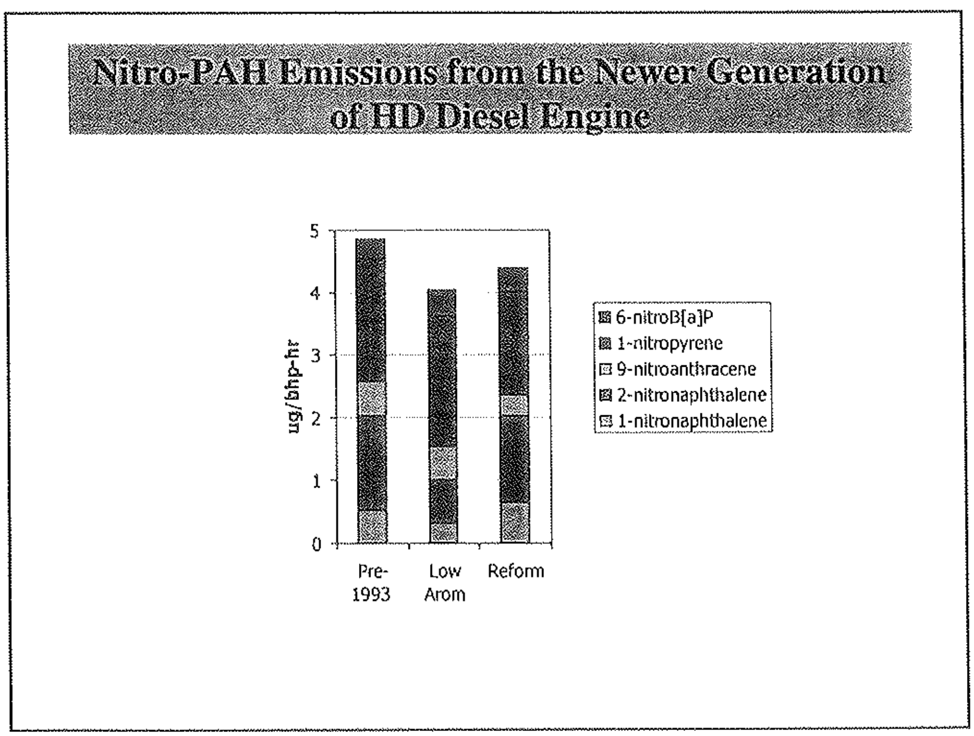
a. From Dietzman et al, 1981; and from Williams and Swarin, 1979
b. From Schuetzle and Frazier (1986); including gas phase
c. From Rogge et al., 1993
d. From NFRAQS (Watson et al., 1998); including gas phase
e. From Schuetzle and Perez (1983);

Norbeck et al. (1998)

Table 1. Diesel Fuel Specification Ranges

Fuel Type	Pre-1993	Low Aromatic	Reformulated
Aromatic HC-Vol. %	33	10 max.	20-25
Sulfur-ppm Wt.	<5000	500 max.	100-300
Cetane Number	>40	48 min.	50-55
PAH - Wt. %	8	1.4 max.	2-5
Nitrogen -- ppm Wt.	300-600	10 max.	40-500


PAH Emissions from the Newer Generation of HD Diesel Engine
8000
7000
6000
5000
4000
3000
2000
1000
0
ug/bhp-hr
Pre-1993
Low Arom.
Refor.
Corone
Benzo[ghi]perylene
Dibenzo[ah+ac]anthracene
Indeno[1,2,3-[cd]pyrene
Benzo[a]pyrene
Benzo[e]pyrene
Benz[b+j+k]fluoranthene
Chrysene
Benz[a]anthracene
Pyrene
Fluoranthene
Methylphenanthrenes
Anthracene
Phenanthrene
2,3,5-trimethylnaphthalenes
1-menaphthalene
2-menaphthalene
Naphthalene
1400
1200
1000
800
600
400
200
0
ug/bhp-hr
Pre-1993
Low Arom.
Refor.


Nitro-PAH Emissions from the Newer Generation of HD Diesel Engine
5
4
3
2
1
0
ug/bhp-hr
Pre-1993
Low Arom
Reform
6-nitroB[a]P
1-nitropyrene
9-nitroanthracene
2-nitronaphthalene
1-nitronaphthalene

Fuel type and brake-specific emissions of particle-associated mutagenic activity (rev/bhp-hr)

Fuel Type	TA98 (+S9) x 10^6	TA98 (-S9) x 10^6
Pre-1993	5.25 ± 0.43	5.79 ± 1.0
Low Aromatic	2.36 ± 0.72	2.53 ± 1.19
Reformulated	2.28	2.89

From Norbeck et al., 1998

Conclusions:

- Heavy-duty diesel engine gaseous emissions decline significantly during the last two decade.
- Particulate matter emissions, expressed in mass/distance traveled show a declining trend, however less pronounced that emissions expressed in mass/work units.
- PAH emissions show a declining trend

Conclusions:

- PAH and nitro-PAH composition in emissions have not change significantly during the last two decades
- Exhaust aftertratment methods and improved fuel formulation have significant impact on heavy-duty diesel emissions

SESSION III

Assessment of Exposure to Diesel Engine Emissions

Philip Lorang, Chair

Brian Leaderer

Alan Rogers

Glen Cass

Douglas Lawson

Alison Pollack

The second day started with a session considering issues in assessment of exposure to diesel emissions. In presenting these issues, speakers discussed differences in exposure assessment in occupational versus ambient settings, analytical approaches to measuring diesel constituents in mines, different approaches to determining contributions of diesel engine emissions to ambient PM, and the use of models to estimate general population exposures to DPM.

Occupational and Ambient Exposures

Brian Leaderer

HEI DIESEL WORKSHOP

ABSTRACT

CONSIDERATIONS IN ASSESSING EXPOSURES TO DIESEL ENGINE EMISSIONS IN OCCUPATIONAL AND AMBIENT SETTINGS. Brian P. Leaderer, Department of Epidemiology and Public Health, Yale University School of Medicine, 60 College Street, P.O. Box 208034, New Haven, Connecticut 06520.

Epidemiologic studies exploring an association between exposure to diesel exhaust emissions and adverse health effects must employ effective exposure assessment protocols that can characterize the nature and extent of the exposure in the population at risk while adjusting for confounding factors. Exposure protocols for assessing diesel exhaust exposure in occupational or ambient settings can employ either direct or indirect methods of assessment or some combination of both. Direct methods include personal exposure monitoring or use of biomarkers of exposure, while indirect methods include environmental monitoring, historical surrogates of exposure, modeling, questionnaires and daily diaries. The selection of one or more methods to be used depends on a number of issues including: the specificity of the study hypothesis, nature of the air contaminant(s) of interest, contaminant sources of interest, technical feasibility, study design, available resources (air sampling equipment, work force, financial resources, etc.), whether the study is retrospective or prospective, availability and quality of historical data, size of study population, subject participation, time frame of the study and acceptable level of uncertainty. In most applications, two or more of the methods are typically employed in a hybrid fashion.

Several considerations play into the exposures assessment approach employed in any study of diesel exhaust emissions. The following information is desirable in designing an exposure assessment protocol for diesel exhaust:

a) a detailed review of available data on diesel source characterization should be conducted. This involves specific information on the nature of the emissions (chemical and physical characterization of gas and particle phase emissions), historical trends of emissions, source use factors which impact the emissions, and a knowledge of the factors which control the transport and transformation of the emitted contaminants as they leave the source and are

distributed within occupational, indoor or outdoor environments.

b) since diesel exhaust emissions exhibit a complex pollutant emission profile, a proxy or tracer contaminant should be identified and evaluated relative to its representativeness to other pollutants emitted from the source and its specificity to the source in a background of air contaminants emitted from a variety of other sources. Elemental carbon (EC) has been proposed as a reasonable surrogate for diesel particle emissions. The uncertainty associated with using EC as a marker should be clearly specified.

c) other potential sources of the particulates, particularly background levels, and the environments in which they are found should be identified and assessed as possible confounders. In occupational settings such as mines radon, asbestos, or other air pollutant exposures should be assessed or controlled for. In studies where the diesel exposure is likely to be low (i.e., truckers or railroad workers) levels of air contaminants in ambient air or the residential setting may be important to consider. Active and passive smoke exposure could also be important.

d) all available air sampling data on diesel related air contaminants, which would indicate environmental concentrations of the contaminant(s) related to diesel emissions in all relevant indoor and outdoor environments, should be identified and evaluated so that the potential magnitude, frequency and duration of exposures in a target population to pollutants from the source can be assessed. This includes assessing any data that would reveal any spatial and temporal patterns in exposures. In retrospective studies of occupational exposures job histories need to be determined and verified for the population at risk and combined with historical and more recent pollutant measurements to construct exposure histories.

e) in designing and conducting air monitoring programs to assess exposures (i.e., personal monitoring) the selection of individuals, locations to be sampled and frequency of sampling should be done in such a way as to make the results representative of the population under study.

f) a clear definition of the health outcome of interest and establish the biologically relevant time for linking exposure to the effect. For example, exposures to diesel particle mass in a study of lung cancer, the primary focus of diesel health studies, needs to be assessed over a period of years rather than days or weeks.

g) identify and evaluate any potential biomarkers of diesel exhaust exposure for use.

Due to a host of limitations (information limitations, available resources, costs, practical issues, time limitations, etc.) few of the considerations identified above are incorporated into exposure assessment protocols for assess exposure to diesel exhaust. It is important for investigators to carefully identify limitations and sources of uncertainty in the assessed exposures and provide cautions as to how the assessments should or should not be used.

CONSIDERATIONS IN ASSESSING EXPOSURES TO DIESEL ENGINE EMISSIONS IN OCCUPATIONAL AND AMBIENT SETTINGS

CONSIDERATIONS

1) Source characterization
2) Marker compounds
3) Biological relevance
4) Biomarkers of exposure
5) Other Air Pollutants
6) Total exposure
7) Selection of representative samples
8) Space and time variability in exposure
9) Assessment methods
10) Specification of uncertainty
11) Reality check

DIESEL SOURCE CHARACTERIZATION

1) Sources
   a) occupational - transportation workers (i.e., truckers)
      diesel equipment workers
      mine workers.
   b) ambient - diesel powered trucks and cars.

2) Physical
   a) mass median diameter of 0.2 um.
   b) 90% to 95% of particles < 1 um.

3) Chemical
   a) complex chemical mix, both vapor and particle phase.
   b) vapor phase - NOx, CO, PAHs, NH3, acroleins, etc.
   c) particle phase - 40 - 70% elemental carbon.

4) Factors
   a) occupational -
      * engine & fuel characteristics (fuel quality, duty cycle, treatment devices, etc.).
      * production related issues (number of vehicles, fuel use, etc.).
      * workplace factors (ventilation, size, etc.).
      * variations in time and space.
   b) ambient -
      * vehicle mix and density
      * changes in engine design and emissions
      * miles traveled
      * other sources
      * meteorology
      * variations in time and space

MARKER OR PROXY COMPOUNDS

1) Criteria -
   * unique to the diesel source.
   * easily measured at low concentrations.
   * similar emission rates from different diesels.
   * in a consistent ratio to individual diesel contaminants that are of health or comfort importance.
   * easily and accurately measured.

2) Markers used or proposed -
   * elemental carbon
   * submicron aerosol mass
   * PAHs
   * extractable mass
   * NOx, CO, CO_2
   * particle phase nicotine (ETS correction)

BIOLOGICAL RELEVANCE

1) Clear specification of outcome under study (acute, chronic, etc.) with identification of suspected contaminants.

2) Exposure assessment time frame should be consistent with the biologically relevant time frame of the exposure/effect.

BIOMARKERS OF EXPOSURE

1) Some proposed markers -
   nitro-PAH metabolites in urine
   DNA adducts

2) Some things to consider -
   * a number of positives (integrated exposure, etc.)
   * biomarkers are indirect indicators of exposure
   * biological relevance - time frame
   * specificity to contaminant(s)/source(s)
   * sensitivity
   * validity
   * overall utility

OTHER AIR POLLUTANTS

1) Occupational -
   * active smoking
   * workplace/process related contaminants (asbestos, radon, other particulate pollutants, etc.)
   * passive smoke exposure

2) Ambient -
   * outdoor particulate air pollution.
   * indoor air pollution.
   * active and passive smoke.

TOTAL EXPOSURE

1) Consider -

* different micro environments - occupational, residential, transportation, etc.
* time and activity patterns - time spent in various micro environments and activities engaged in.
* other media and routes of exposure - food, water, ingestion and dermal.

SELECTION OF SAMPLE FOR MONITORING

1) If personal monitoring is conducted the sample of individuals should be representative of the range of activities, behaviors, and exposures of the study population. Some issues to consider:
   * random sample or stratified sample.
   * convenience sample.
   * repeat sampling of individuals.
   * variations in relation to space and time.
   * predictive value for use in constructing historical personal exposures in relation to work histories, residential.
   * specification of uncertainty.

2) If area monitoring is conducted the sampling should consider:
   * different work, ambient or indoor environments.
   * present and historical working conditions - source types and use patterns, ventilation rates, work shifts.
   * monitoring factors impacting concentrations.
   * activity patterns of occupants.
   * utility in construction historical concentrations.
   * specification of uncertainty.

VARIABILITY IN EXPOSURE

1) In conducting personal, area monitoring or constructing historical exposure models, consideration should be given to exploring all available data in order to capture:
   * magnitude of the concentrations.
   * duration of concentrations.
   * frequency of concentrations.
   * variations in space - areas of the country, different locations of workplaces, etc.
   * variations in time - seasons, meteorology, sources, etc.
   * variations over time in work or residential histories.

ASSESSMENT METHODS

1) Direct

* personal monitoring - needs additional information to assess source contribution and factors impacting exposure.
* biomarkers - measure of dose and requires models to relate to exposure and sources.

2) Indirect

* questionnaires - to construct current and past general exposure categories.
* historical surrogates of exposure - job category and length of time in specific jobs, type and number of equipment used, fuel use, vehicle miles traveled, use of protective equipment or source controls, etc.
* environmental monitoring - needs additional information - source use, questionnaires etc.
* time activity information - needs additional information.
* ***models*** - utilizes some or all of the above direct and indirect measures of exposure assessment.

SPECIFICATION OF UNCERTAINTY

1) Each step in the effort to assess exposure entails uncertainty. The uncertainty associated with each step or element as well as the overall uncertainty should, to the extent possible, be specified and quantified. For many assessments the estimated uncertainty will out of necessity be based upon the subjective judgement of the investigators.

REALITY CHECK

1) Due to a host of limitations (information limitations, available resources, costs, practical issues, time limitations, etc.) few of the considerations identified above are incorporated into exposure assessment efforts. It is important for investigators to carefully identify limitations and sources of uncertainty in the assessed exposures and provide cautions as to how the assessments should or should not be used.

Exposures in Australian Mines

Alan Rogers

Comparison of Different Analytical Approaches for Measuring Exposures to Diesel Emissions in Australian Mines

Alan Rogers, Alan Rogers OH&S Pty Ltd and Bill Whelan, University of Sydney

Research conducted in Australian coal mines since 1990 indicates that diesel particulate matter (soot) is composed of solid elemental carbon cores (EC) produced during combustion together with a range of adsorbed organic material (OC). More than 90% of these carbon particles are respirable with aerodynamic diameters of one micron or less and hence capable of penetrating into the deep regions of the lung.

Two stage impaction samplers have been developed to separate diesel particulates from the coal mine dust. Results are expressed in terms of diesel particulate sub micron mass (DPSMM). In general for the sample collected, there is less than 10% interference from very fine coal dust. In high or peak dust conditions the samplers are subject to overload and produce false high results. Since 1995, DPSSM measurements have been supplemented with a thermal/optical reflectance elemental carbon analysis method so as to distinguish between the elemental carbon core, the absorbed organic carbon and the very fine coal dust in the sub micron mass.

Research funded by BHP and the Joint Coal Board Health & Safety Trust has resulted in the examination of workforce exposures in NSW and Queensland underground coal mines. Levels of 0.01-0.37 mg/m3 EC and 0.01-0.64 mg/m3 DPSMM have been recorded and at this early stage of the investigation there appears to be little difference between NSW and Queensland mines even though they have a different scale of operation and experience different mining conditions. To date some 800+ personal miner exposure samples and 500+ personal test tunnel driver samples have been collected and entered into a data base to assist in determining the relative effects of parameters such as engine size, ventilation, operating practices etc. in actual mining situations. Summaries of the data is available on the net at *www.jcb.org.au*

Monitoring in NSW and Queensland underground coal mines indicates that DP and EC exposure levels vary considerably depending on working conditions and control strategies. Generally transport duties produce the lowest results with heavy workloads particularly long wall moves producing the highest results. The factors that tend to identify lower exposures are the use of low sulphur fuel, good engine maintenance, good roadway conditions and good attitudes to driving. High exposures are identified when bad road or boggy conditions exist, poor engine maintenance is experienced and hard working engines are encountered, such that around 80-90% of the high exposures are due to 10% of the machines. Subjective effects of eye irritation and discomfort from odours occur more readily at levels in excess of 0.1 mg/m3 EC or 0.2 mg/m3 DPSMM.

Heavy load activities such as long wall moves, power traming, or dozer work give higher exposure levels. The indications are that there will be a requirement for these types of machines to be fitted with a combination of control technologies to reduce irritation and discomfort of the workforce and for compliance with the proposed exposure standards. Based on the results from various studies, an industry Code of Practice has been distributed and a number of mines have implemented control strategies from the Code that have significantly reduced workforce exposures.

In operating mine atmospheres, the amount of elemental carbon in DP is generally in the range 25-85%, and total carbon constitutes 60-100% of the submicron mass. In addition there is considerable variability in the proportion of diesel particulate found in the sub micron fraction versus that found in the respirable fraction of the mine aerosol. There are indications that the individual compounds such as PAH's vary considerably in the organic fraction. The variability in concentratiou and the analytical errors associated with measuring these trace and ultratrace components is completely expected and is part and parcel of the statistics associated with measuring small quantities of pollutants in environmental samples. In addition to the analytical problems of measuring these trace compounds, the composition of the compounds in the raw diesel exhaust is also variable and dependent on engine condition and operating parameters. Raw emissions consist of a complex variable and reactive mixture which changes chemical composition and particle size as it ages. Given the complexities of determining exactly what specific chemical analysis is best for defining diesel particulates and or what is the relevant component/s that are likely cause potential health effects, the current findings from our research indicate that group categories such as EC, TC or DPSMM may be used as a surrogate of diesel soot exposure in the mining industry. It is however necessary to fully understand the limitations of such measurements and standardise on field sampling and laboratory analysis techniques.

The accuracy limit of the microbalance limits the reporting of 8 hour duration DPSMM samples to the nearest 0.01 mg/m^3; for elemental carbon, organic carbon and total carbon the reporting limit is to the nearest 0.001 mg/m^3 based ou NIOSH Method 5040. The indications are that DPSMM has limited application for monitoring low level occupational exposures and is not applicable to environmental sampling. EC and TC monitoring are suitable for almost all occupational situations and for urban environmental monitoring but they are limited in application for rural environmental monitoring.

The current monitoring techniques are research tools and require considerable technical skills to obtain suitable exposure results. A practical reproducible and accurate method for measurement of exposure will need to be developed for routine use in the mining industry.

Further Publications on this Australian Research:

Rogers A, Davies B, Conaty G., *Diesel Particulates - Health Effects, Measurement and Standard Setting*, Proceedings 12th Annual Conference, Australian Institute of Occupational Hygienists, 97-101, 1993

Rogers A and Whelan W., *Elemental Carbon as a means of Measuring Diesel Particulate Matter Emitted from Diesel Engines in Underground Mines*, Proceedings 15th Annual Conference, Australian Institute of Occupational Hygienists, 208-212, 1996.

Pratt S, et al, *Evaluation and Control of Employee Exposure to Diesel Particulate at Several Australian Coal Mines*, App Occup Environ Hyg, 12(12) 1032-1037, 1997.

COMPARISON OF DIFFERENT ANALYTICAL APPROACHES FOR MEASURING EXPOSURES TO DIESEL EMISSIONS IN AUSTRALIAN MINES

Alan Rogers, AROH&S Pty Ltd and

Bill Whelan, University of Sydney

DIESEL EXHAUST EMISSIONS

1. GASEOUS

   OXIDES OF CARBON CO, CO_2

   OXIDES OF NITROGEN NO, NO_2, NO_X

2. PARTICULATE (SOOT)

SUB MICRON DUST FRACTION

DP = EC + OC + MINERAL DUST

DP EXPOSURES - FULL SHIFT REPEAT DUTY CYCLE TUNNEL TESTS

Fuel type	1. Commercial low S high aliphatic		3. Experimental low S fuel		9. Commercial on road diesel		7. imported high S, high aromatics	
% S	0.01		0.03		0.19		0.41	
DP mg/m3	0.43	0.46	0.42	0.38	0.48	0.43	0.83	0.90
PAH as % organics	7.9	0.9	3.4	4.2	2.7	4.9	5.6	3.6

PAH EXPOSURES (UG/G ORGANICS) FULL SHIFT REPEAT DUTY CYCLE TUNNEL TESTS

Fuel type	1. Commercial low S, high aliphatic		3. Experimental low S fuel		9. Commercial on road diesel		7. imported high S high aromatics	
Naphthalene	3228	664	1312	1508	1697	1607	2418	1137
Fluorene	257	89	61	39	132	182	132	137
Phenanthrene	511	73	125	50	302	226	418	231
Pyrene	77	4	11	4	84	31	37	13
Benz(a)anthracene	10	<3	<3	<3	5	<3	18	10

METHOD 5040

ELEMENTAL CARBON (DIESEL EXHAUST)

NIOSH MANUAL OF ANALYTICAL METHODS 1995

SUMMARY OF ELEMENTAL CARBON SAMPLING CONDUCTED IN AUSTRALIAN MINING OPERATIONS

Mine and Operation		DPSMM mg/m³	EC mg/m³	TC mg/m³	EC/DP	EC/TC	% EC sub/resp
NSW Mine 1	general duties	0.04 - 0.32	0.02 - 0.59	0.04 - 0.33	(0.32)	(0.47)	-
	long wall move Oct 96	0.14 - 0.25	0.05 - 0.20	0.13 - 0.36	(0.44)	(0.46)	-
NSW Mine 2	long wall move Sep 96	0.12 - 0.69	0.07 - 0.47	0.12 - 0.57	0.58-0.86 (0.72)	0.57-0.83 (0.74)	-
	long wall move Oct 97	0.21 - 0.65	0.19 - 0.52	0.24 - 0.60	0.63-0.90 (0.77)	0.70-0.86 (0.79)	-
NSW Mine 3	general duties Oct 97	n d	0.011 - 0.096	0.046 - 0.214	nd	0.27-0.50 (0.39)	-
NSW Mine 4	place change Feb 98	0.04 - 0.08	0.013 - 0.047	0.061 - 0.104	0.28-0.58 (0.46)	0.22-0.45 (0.34)	18-89 (52)
Qld Mine 5	long wall move Dec 97	0.06 - 0.50	0.012 - 0.270	0.056 - 0.750	0.02-0.56 (0.39)	0.05-0.78 (0.55)	25-86 (73)
	long wall move June 98	0.12 - 0.46	0.050 - 0.299	0.110 - 0.402	0.29-0.92 (0.56)	0.46-0.74 (0.56)	80-92 (84)
Qld Mine 6	long wall move March 98	0.08 - 0.42	0.051 - 0.217	0.060 - 0.487	0.08-1.00 (0.44)	0.10-0.68 (0.49)	57-86 (71)
	general duties July 98	0.04 - 0.40	0.022 - 0.244	0.056 - 0.360	0.10-0.66 (0.51)	0.30-0.67 (0.53)	27-86 (65)
Qld Mine 7	long wall dev. May 98	0.13 - 0.31	0.020 - 0.130	0.070 - 0.300	0.14-0.57 (0.37)	0.31-0.68 (0.53)	25-85 (57)
	long wall dev. Sept 98	0.14 - 0.32	0.084 - 0.169	0.190 - 0.289	0.31-0.56 (0.44)	0.34-0.58 (0.47)	40-78 (65)
Qld Mine 8	Punch long wall inst Aug 98	0.11 - 0.64	0.046 - 0.339	0.108 - 0.575	0.35-0.58 (0.53)	0.31-0.67 (0.54)	73-87 (82)
Non-coal 9	Cu open stope May 98	0.11 - 1.30	0.042 - 0.372	0.112 - 0.572	0.34	0.47	-
Non-coal 10	Pb/Zn Development	0.12 - 0.38	0.090 - 0.173	0.115 - 0.293	0.46	0.54	-
Non-coal 11	Al_2O_3 Precip tank July 98	n d	0.015 - 0.077	0.067 - 0.353	n d	0.23	-
Sydney Air Mass	various	*	0.001 - 0.006	0.006 - 0.036	*	0.03	-

* awaiting PM10 results
DPSMM, EC and TC results are for submicron fraction of the mine aerosol
Results are presented as range and (mean values)
% EC sub/resp is the % of EC found in the submicron fraction versus that found in the respirable fraction of the mine aerosol

PROBLEMS FACING DP EXPOSURE MEASUREMENT

1. OVERLOAD OF SAMPLING DEVICE
   EFFECTS CUT OFF

2. FOR DPSMM THERE ARE ERRORS IN WEIGHING
   SUCH SMALL MASSES ON FILTERS WHICH ARE
   SUBJECT TO MOISTURE PICK UP,
   ELECTROSTATIC CHARGE, FILTER DAMAGE

3. VARIABLE ANALYTICAL SENSITIVITY AND
   RATIOS OF DP EC AND OC

REQUIREMENTS

A PRACTICAL REPRODUCIBLE AND ACCURATE
MEASUREMENT OF EXPOSURE SUITABLE FOR
ROUTINE USE IN THE MINING INDUSTRY

EXPOSURE ASSESSMENTS ?

+/- VE EPIDEMIOLOGICAL STUDIES INDICATING RISK OF LUNG CANCER
(no actual exposure measurements)

DRAFT ACGIH EXPOSURE STANDARD $0.05\ mg/m^3$ FOR GENERAL INDUSTRY
(<1um , particulate & adsorbed component)

JULY 1998 MSHA PROPOSED RULE FOR UNDERGROUND COAL MINES
(equipment to be fitted with control devices to achieve 95% reduction)

OCT 1998 MSHA PROPOSED RULE FOR UNDERGROUND METAL & NONMETAL MINES
(exposure standard 0.2 mg/m3 total carbon)

AUSTRALIAN ACTIVITIES

NSW JOINT COAL BOARD
TRIPARTITE DIESEL PARTICULATE
SUBCOMMITTEE

- REVIEW HEALTH EFFECTS

- CONDUCT EPIDEMIOLOGICAL STUDY ON NSW COAL MINERS

- RECOMMEND EXPOSURE STANDARDS
  - HEALTH
  - BEST PRACTICE

- CONTROL STRATEGIES

Diesel Engine Contribution to Atmospheric Particles

Glen Cass

ABSTRACT

Diesel Engine Contributions to Atmospheric Particles: Measurement Methods and Atmospheric Modeling Approaches

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In this presentation, methods for the characterization of fine particle emissions from diesel-powered vehicles first will be described. A two-stage dilution source sampler will be discussed that has been configured to permit measurement of the emissions of individual gas-phase organic compounds, fine particle mass, fine particle chemical composition, fine particle size distributions, as well as the size distribution of the chemical composition of the particles emitted from diesel engines. Then air quality modeling methods that can be used to determine how the various emissions sources present in a city combine to produce the observed airborne particle mixture will be discussed. In the source-oriented modeling approach, an atmospheric transport model is used to track diesel exhaust particulate matter emissions through a simulation of atmospheric fluid motion and chemical reaction as the pollutants are transported from their sources to community air monitoring sites. In the second modeling approach, use of organic compounds that act as tracers for the presence of the effluent from the particular sources that contribute to an atmospheric sample will be described. From these models, the contributions that diesel engine particulate matter emissions make to ambient fine particle concentrations can be determined in a way that assists the formulation of regional plans for the control of atmospheric particulate matter concentrations. Methods developed will be illustrated through analysis of the causes of the Los Angeles fine particle air pollution problem.

References:

J. J. Schauer, M. J. Kleeman, G. R. Cass and B. R. T. Simoneit. Measurement of Emissions from Air Pollution Sources. 2. C1 through C30 Organic Compounds from Medium Duty Diesel Trucks, Environmental Science and Technology, in press, 1999.

A. Eldering and G. R. Cass. A Source-Oriented Model for Air Pollutant Effects on Visibility, Journal of Geophysical Research-Atmospheres, 101 (D14), 19343-19369,1996.

M. J. Kleeman, A. Eldering, and G. R. Cass. Modeling the Airborne Particle Complex As a Source-Oriented External Mixture, Journal Of Geophysical Research - Atmospheres 102, 21355-21372, 1997.

M. J. Kleeman and G. R. Cass. Source Contributions to the Size and Composition Distribution of Urban Particulate Air Pollution, Atmospheric Environment, 32, 2803-2816, 1998.

J.J. Schauer, W.F. Rogge, L.M. Hildemann, M.A. Mazurek, G. R. Cass and B.R.T. Simoneit. Source Apportionment of Airborne Particulate Matter Using Organic Compounds as Tracers, Atmospheric Environment, 30 (22), 3837-3855, 1996.

DIESEL ENGINE CONTRIBUTIONS TO ATMOSPHERIC PARTICLES:

MEASUREMENT METHODS AND ATMOSPHERIC MODELING APPROACHES

GLEN CASS

CALIFORNIA INSTITUTE OF TECHNOLOGY

ACKNOWLEDGEMENTS

JAMIE SCHAUER, UNIVERSITY OF WISCONSIN

MICHAEL KLEEMAN, UC DAVIS

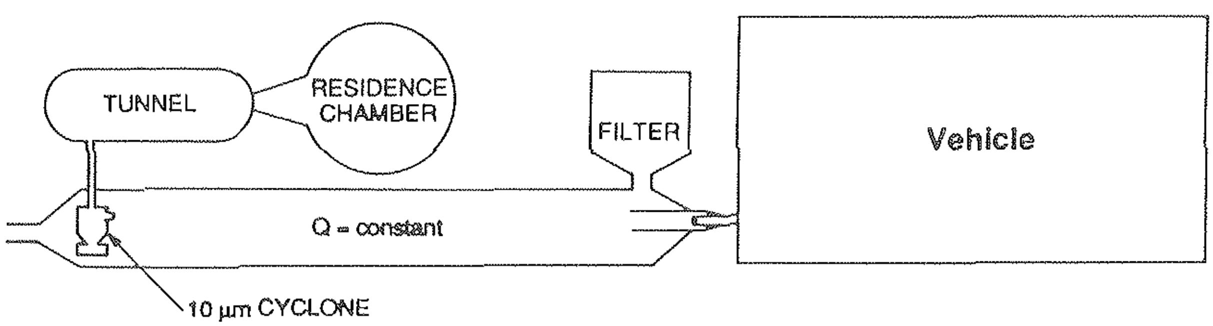
BERND SIMONEIT, OREGON STATE UNIVERSITY

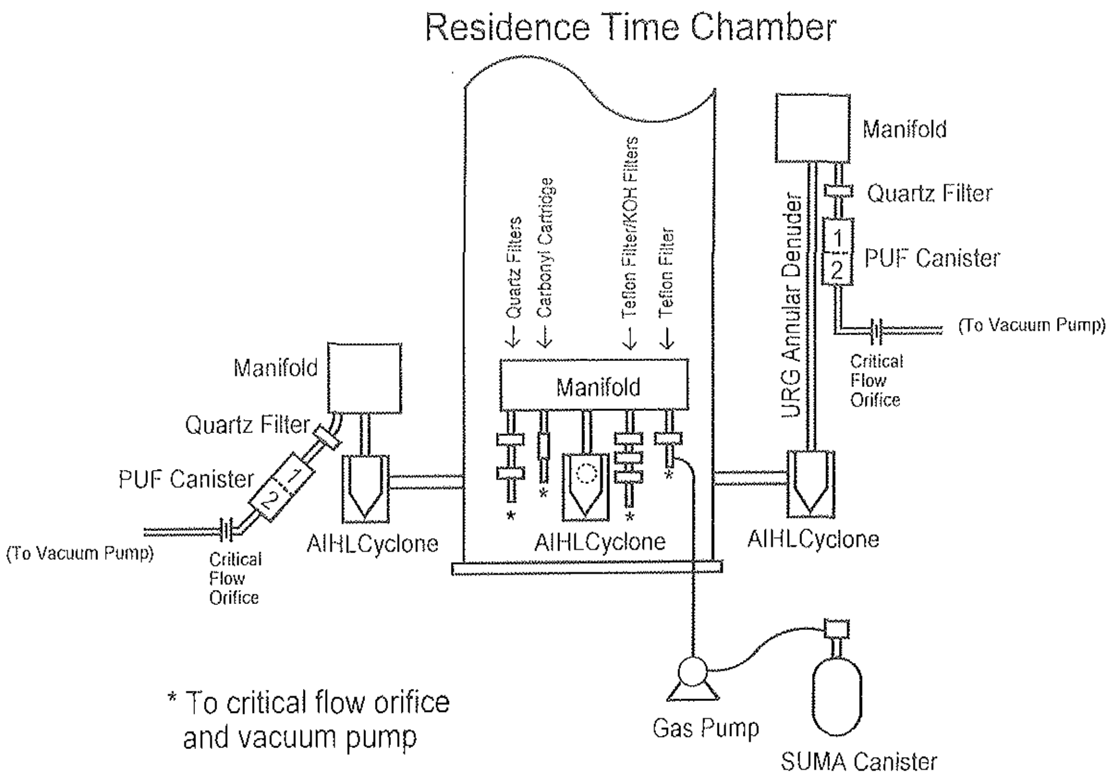
DIRECT PARTICLE EMISSIONS FROM SOURCES

- MEASUREMENT IS DIFFICULT BECAUSE ORGANIC VAPORS THAT WILL FORM PARTICLES AS PLUME COOLS IN THE ATMOSPHERE ARE STILL IN THE GAS PHASE AT STACK TEMPERATURES

- NEED TO SIMULATE THIS PROCESS OF DILUTION AND COOLING WHILE TAKING SOURCE SAMPLES

Sampler Configuration for Vehicle Experiments (Top View)





Fine Particulate Matter

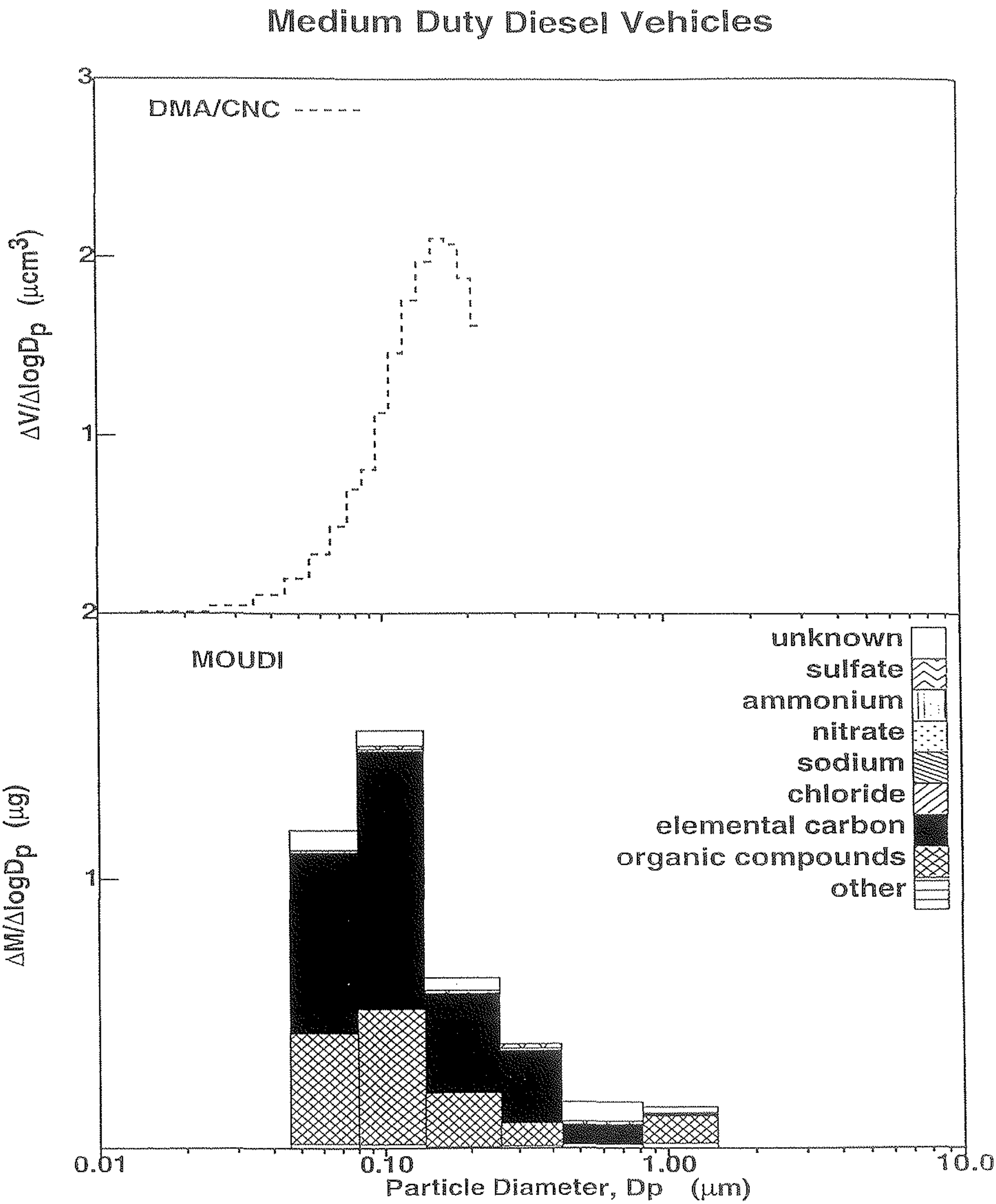
- Fine Particle Mass Emissions
- Elemental and Organic Carbon
- Trace Metals by XRF and AA
- Inorganic Ions by IC and Colorimetry

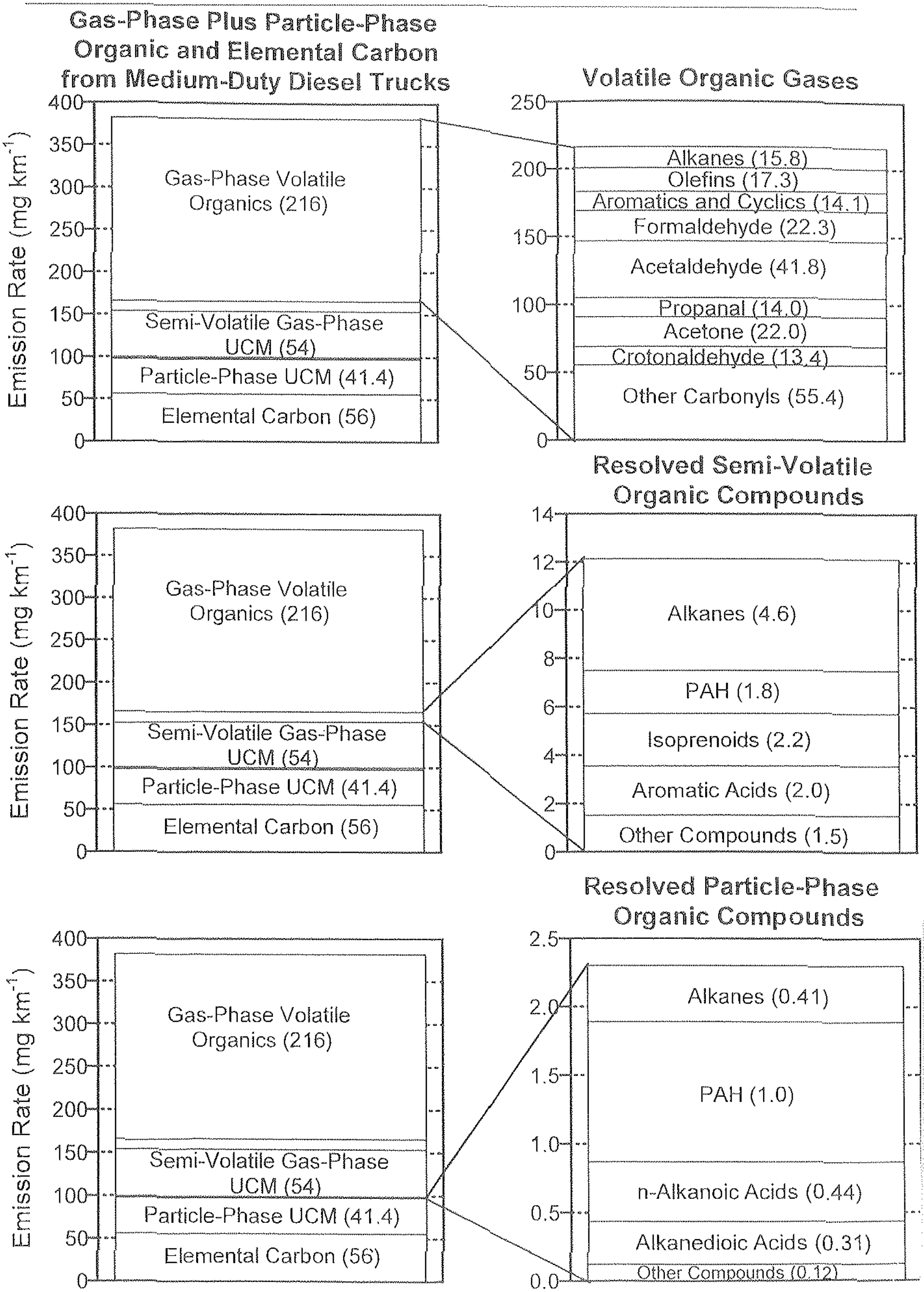
Particle Size Distributions

- Scanning DMA/CNC Combination
- Laser Optical Particle Counter
- Pair of Moudi Impactors
  - Mass as a Function of Size
  - Elemental and Organic Carbon
  - Trace Metals by Neutron Activation
  - Inorganic Ions by IC and Colorimetry

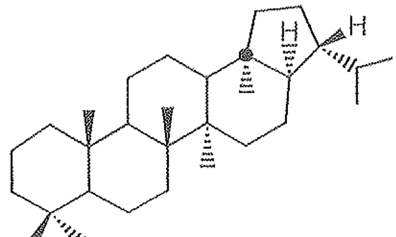
Organic Compound Measurements

- Gas Phase Hydrocarbons:
  - Polished Stainless Steel SUMA Canisters
  - GC/FID Analysis
- Semi-Volatile and Particle Phase Organics:
  - XAD Coated Denuder - Quartz Filter - PUF
  - Quartz Fiber Filter - PUF
  - GC/MS Analysis
- Carbonyls:
  - DNPH Cartridges -HPLC Analysis
- Organic Acids:
  - KOH Impregnated Quartz Filters
  - IC or GC/MS Analysis

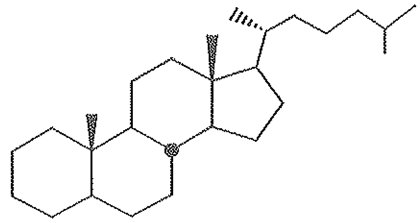

Medium Duty Diesel Vehicles
DMA/CNC
MOUDI
ΔV/ΔlogDp (μcm3)
ΔM/ΔlogDp (μg)
unknown
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
organic compounds
other
0.01
0.10
1.00
10.0
Particle Diameter, Dp (μm)
3
2
1
2
1


Gas-Phase Plus Particle-Phase Organic and Elemental Carbon from Medium-Duty Diesel Trucks
Emission Rate (mg km-1)
Gas-Phase Volatile Organics (216)
Semi-Volatile Gas-Phase UCM (54)
Particle-Phase UCM (41.4)
Elemental Carbon (56)
Volatile Organic Gases
Alkanes (15.8)
Olefins (17.3)
Aromatics and Cyclics (14.1)
Formaldehyde (22.3)
Acetaldehyde (41.8)
Propanal (14.0)
Acetone (22.0)
Crotonaldehyde (13.4)
Other Carbonyls (55.4)
Resolved Semi-Volatile Organic Compounds
Alkanes (4.6)
PAH (1.8)
Isoprenoids (2.2)
Aromatic Acids (2.0)
Other Compounds (1.5)
Resolved Particle-Phase Organic Compounds
Alkanes (0.41)
PAH (1.0)
n-Alkanoic Acids (0.44)
Alkanedioic Acids (0.31)
Other Compounds (0.12)

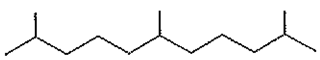
Motor Vehicles



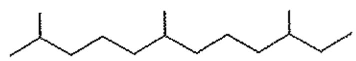
17α(H),21β(H)-Hopane



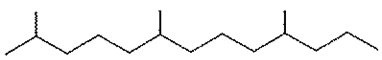
5α(H),14α(H),17α(H)-Cholestane



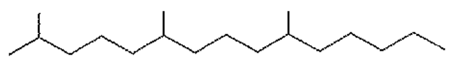
Norfarnesane
2,6,10-Trimethylundecane



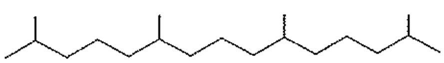
Farnesane
2,6,10-Trimethyldodecane



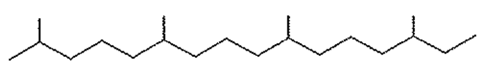
2,6,10-Trimethyltridecane



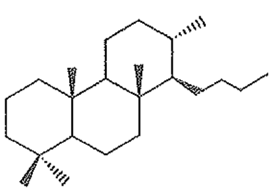
Norpristane
2,6,10-Trimethylpentadecane



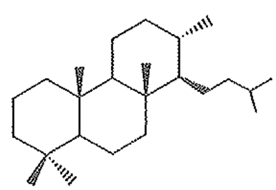
Pristane
2,6,10,14-Tetramethylpentadecane



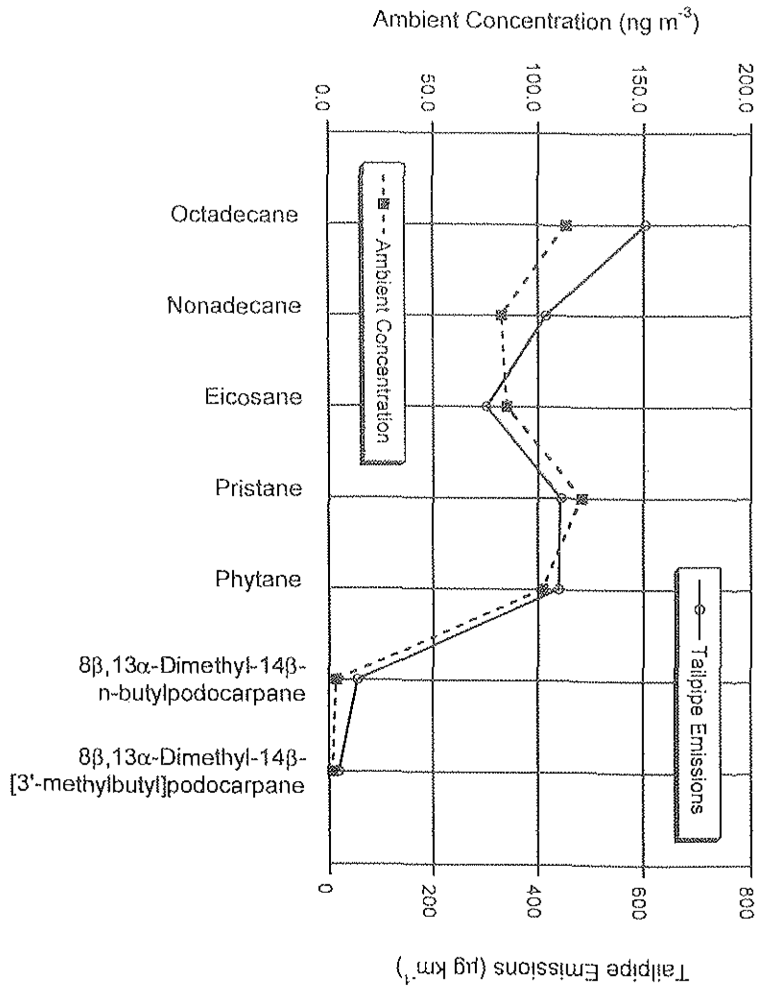
Phytane
2,6,10,14-Tetramethylhexadecane



8β,13α-Dimethyl-14β-n-butylpodocarpane



8β,13α-Dimethyl-14β-[3'methylbutyl]podocarpane



RELATING EMISSIONS TO AIR QUALITY: 2 APPROACHES

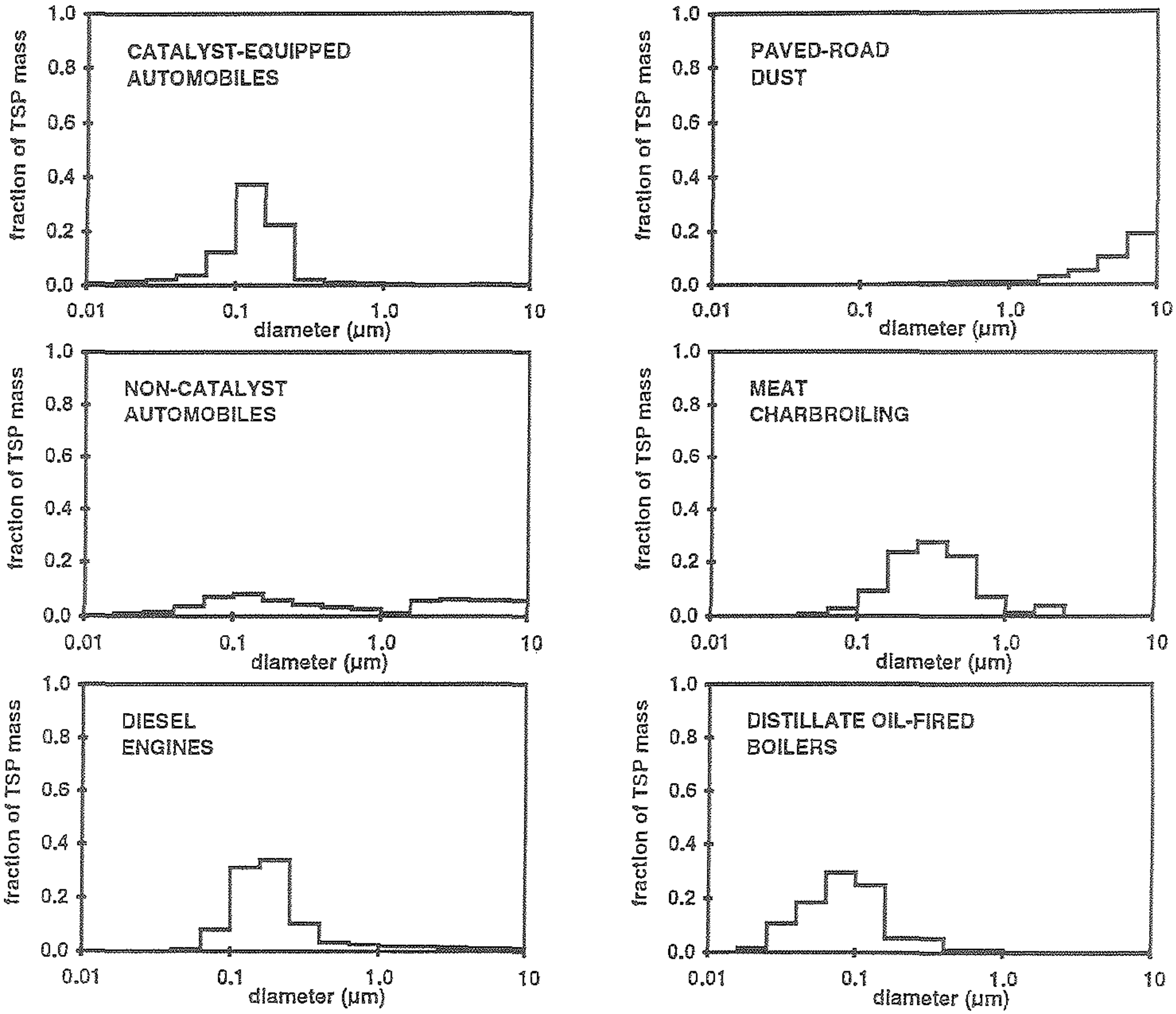
- SIMULATION OF ATMOSPHERIC TRANSPORT AND CHEMICAL REACTION
- ORGANIC CHEMICAL TRACER TECHNIQUES

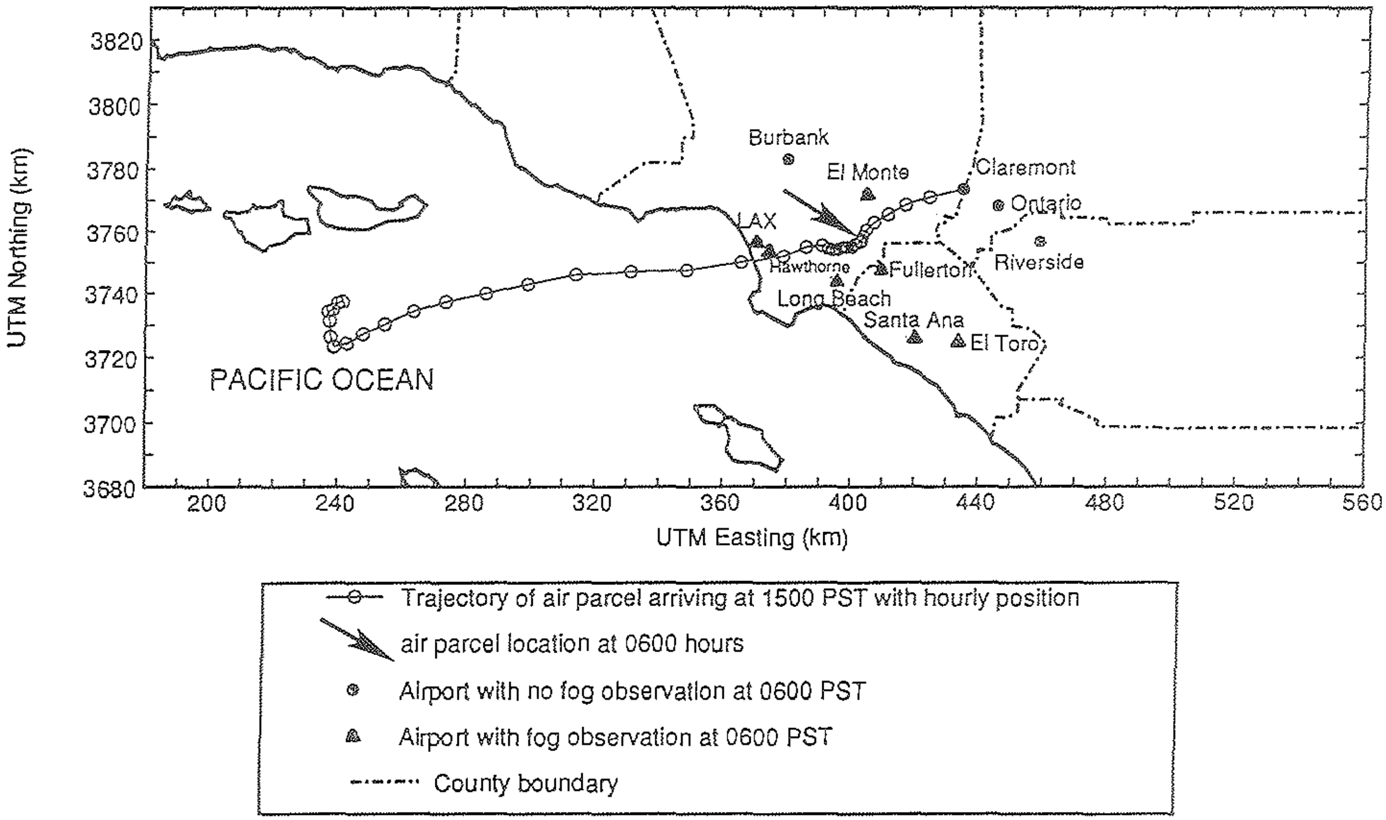
Aerosol Formation and Transport

- photochemical trajectory model - Russell *et al.*
- 15 discrete particle sizes that grow by condensation or incorporation into fog
- gas phase chemistry - Carter, 1990
- aerosol thermodynamics and diffusion of condensable secondary species to and from particles - Wexler *et al.*
- fog module
- model tracks reactive gases, primary aerosol, sea salt, secondary sulfates, nitrates, and organics

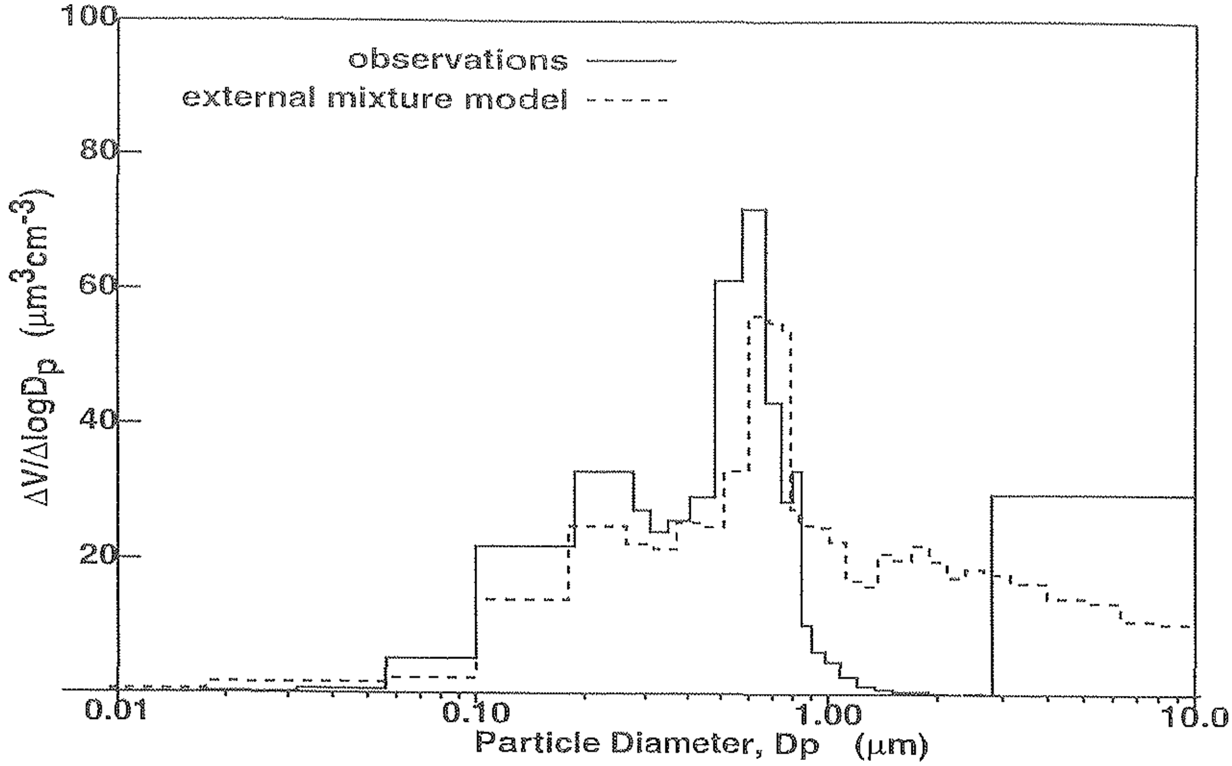
Sources Tested

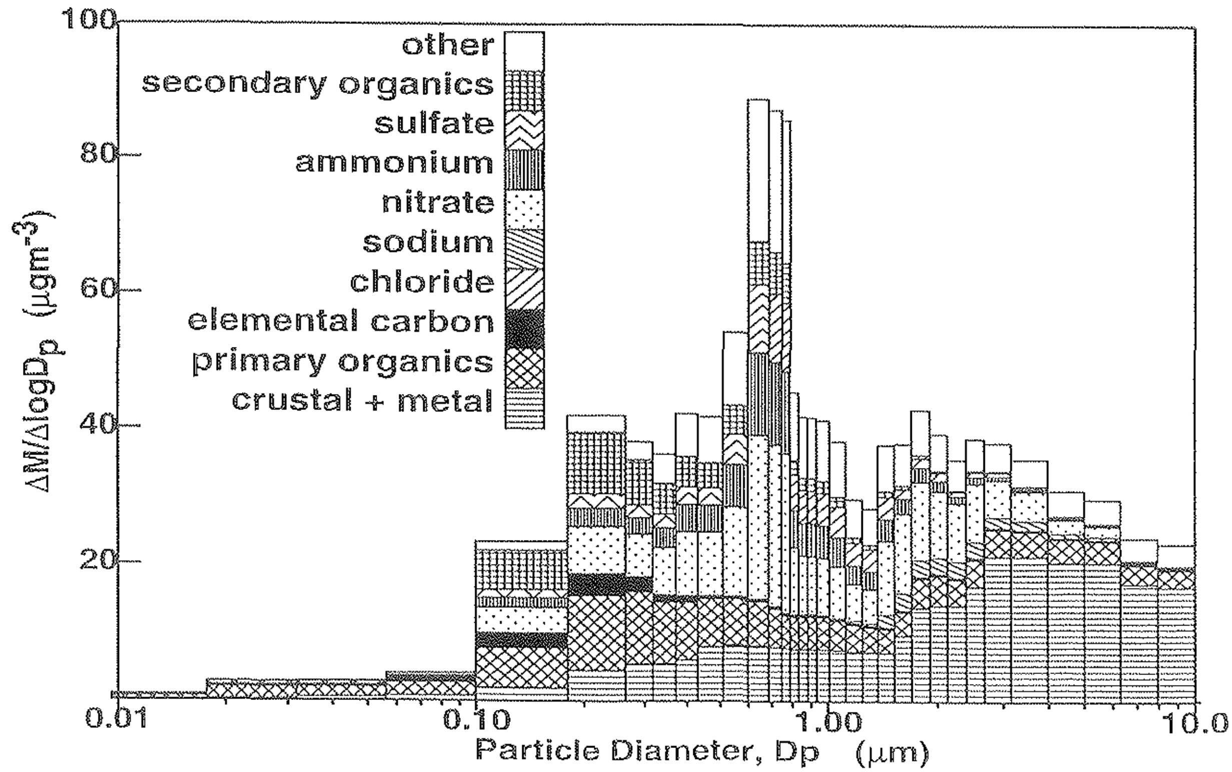
Catalyst-Equipped Autos
Non-catalyst Autos
Diesel Trucks
Fireplace Combustion of Wood
Oil-Fired Boilers
Meat Charbroilers
Natural Gas Home Appliances
Roofing Tar Pots
Cigarette Smoke
Tire Dust
Brake Dust
Paved Road Dust
Vegetative Detritus

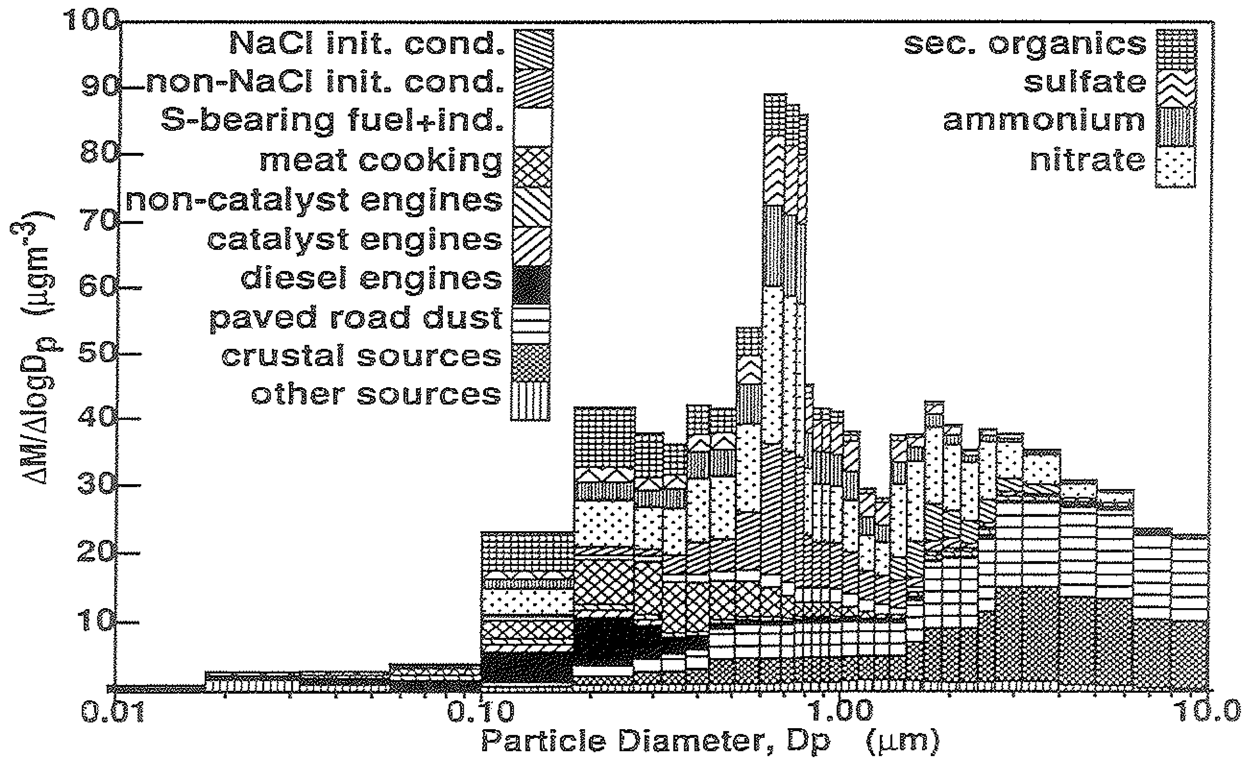

CATALYST-EQUIPPED
AUTOMOBILES
PAVED-ROAD
DUST
NON-CATALYST
AUTOMOBILES
MEAT
CHARBROILING
DIESEL
ENGINES
DISTILLATE OIL-FIRED
BOILERS
fraction of TSP mass
diameter (µm)
0.0
0.2
0.4
0.6
0.8
1.0
0.01
0.1
1.0
10

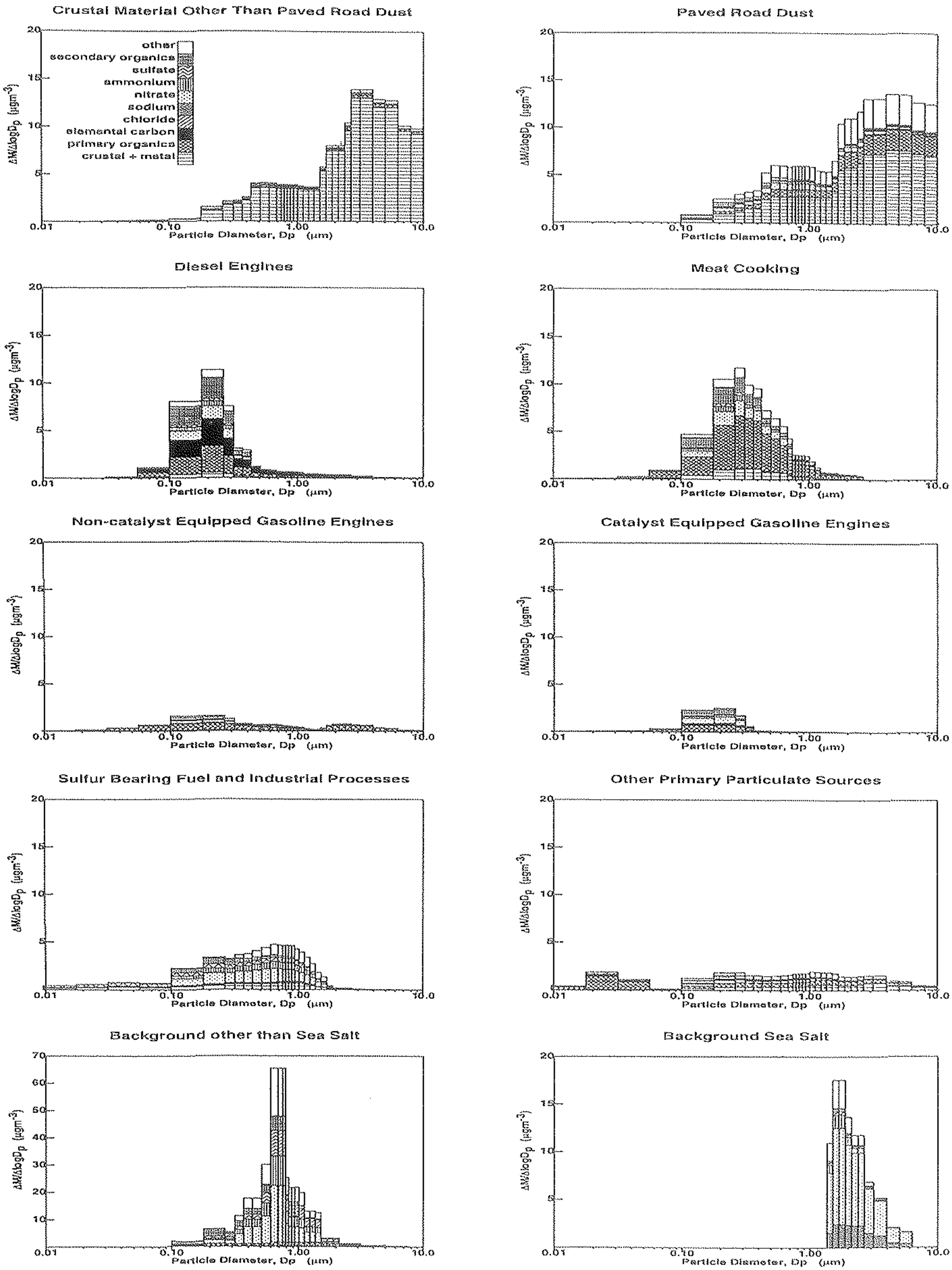


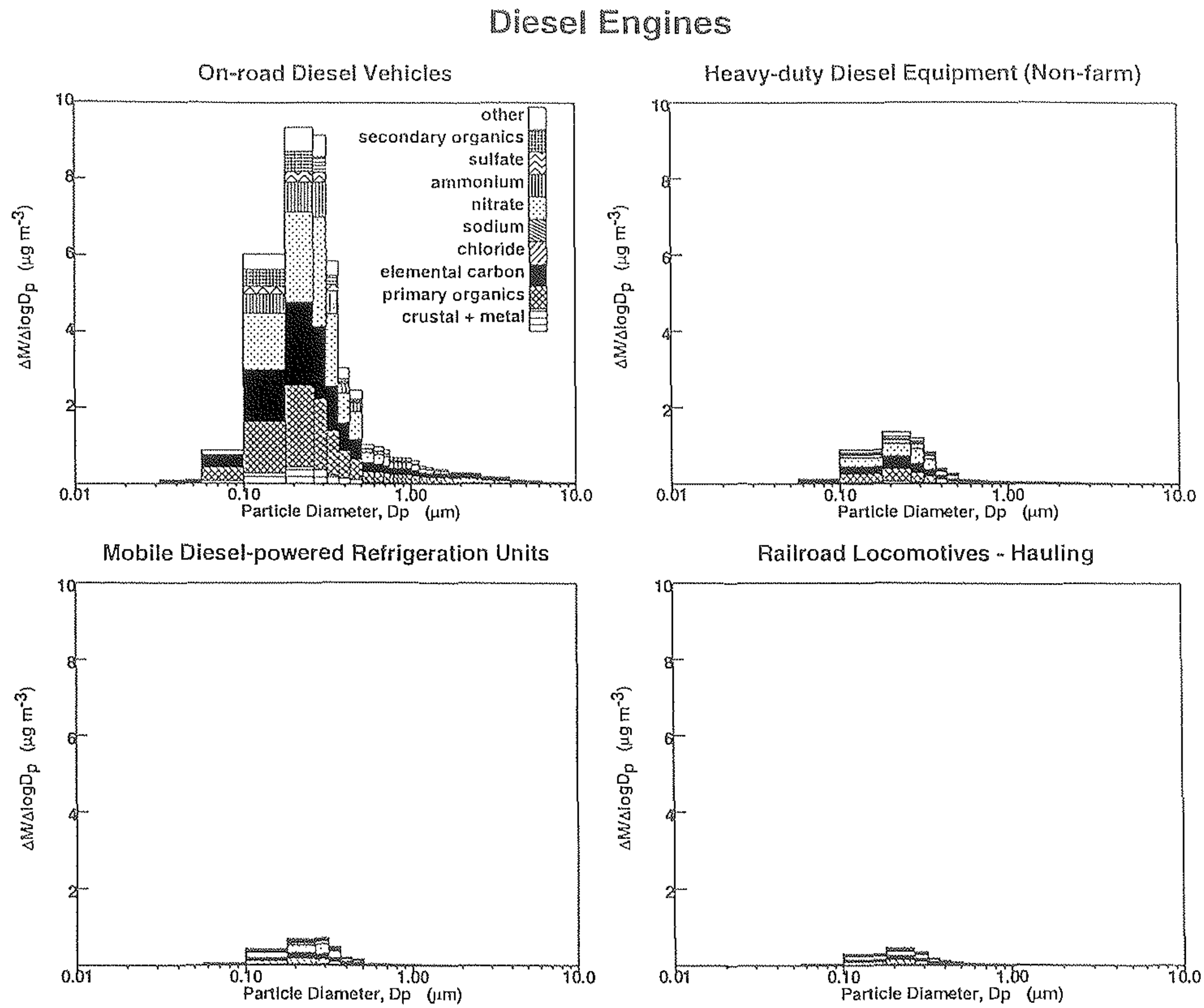
UTM Northing (km)
3820
3800
3780
3760
3740
3720
3700
3680
200
240
280
320
360
400
440
480
520
560
UTM Easting (km)
Burbank
El Monte
Claremont
Ontario
Riverside
LAX
Hawthorne
Fullerton
Long Beach
Santa Ana
El Toro
PACIFIC OCEAN
Trajectory of air parcel arriving at 1500 PST with hourly position
air parcel location at 0600 hours
Airport with no fog observation at 0600 PST
Airport with fog observation at 0600 PST
County boundary


100
80
60
40
20
0.01
0.10
1.00
10.0
observations
external mixture model
ΔV/ΔlogDp (μm³cm⁻³)
Particle Diameter, Dp (μm)


100
80
60
40
20
0.01
0.10
1.00
10.0
other
secondary organics
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
primary organics
crustal + metal
ΔM/ΔlogDp (μgm⁻³)
Particle Diameter, Dp (μm)


NaCl init. cond.
non-NaCl init. cond.
S-bearing fuel+ind.
meat cooking
non-catalyst engines
catalyst engines
diesel engines
paved road dust
crustal sources
other sources
sec. organics
sulfate
ammonium
nitrate
ΔM/ΔlogD$_p$ (μgm^{-3})
Particle Diameter, Dp (μm)
0.01
0.10
1.00
10.0
10
20
30
40
50
60
70
80
90
100


Crustal Material Other Than Paved Road Dust
other
secondary organics
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
primary organics
crustal + metal
Paved Road Dust
Diesel Engines
Meat Cooking
Non-catalyst Equipped Gasoline Engines
Catalyst Equipped Gasoline Engines
Sulfur Bearing Fuel and Industrial Processes
Other Primary Particulate Sources
Background other than Sea Salt
Background Sea Salt
$\Delta M/\Delta \log D_p$ ($\mu g m^{-3}$)
Particle Diameter, Dp (μm)
0.01
0.10
1.00
10.0


Diesel Engines
On-road Diesel Vehicles
Heavy-duty Diesel Equipment (Non-farm)
Mobile Diesel-powered Refrigeration Units
Railroad Locomotives - Hauling
other
secondary organics
sulfate
ammonium
nitrate
sodium
chloride
elemental carbon
primary organics
crustal + metal
ΔM/ΔlogDp (μg m-3)
Particle Diameter, Dp (μm)
0.01
0.10
1.00
10.0

MOLECULAR TRACER TECHNIQUES

- SEEK SINGLE COMPOUNDS OR COMPOUND GROUPS THAT ARE
  - CHARACTERISTIC OF A SOURCE
  - RELATIVELY STABLE

PROCEDURE:

- Characterize ambient pollutant concentrations
  - Air monitoring network
  - Bulk elemental analysis
  - Organic chemical composition by GC/MS

- Characterize major emissions sources
  - Use dilution source sampler
  - Measure 15 most important particle source types
  - Organic chemical analysis by GC/MS

- Compute linear combination of source effluents needed to reproduce distribution of particulate organic compounds in the atmosphere

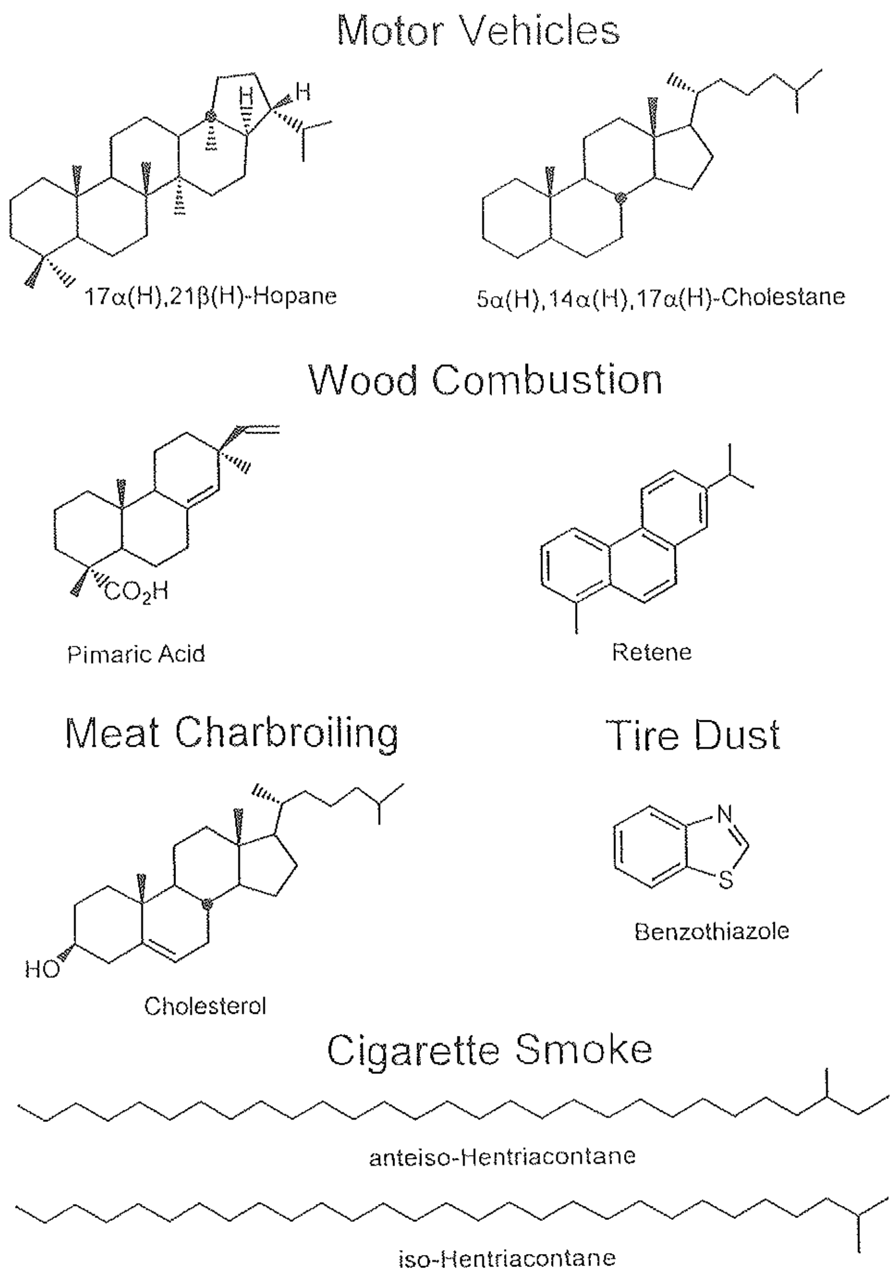
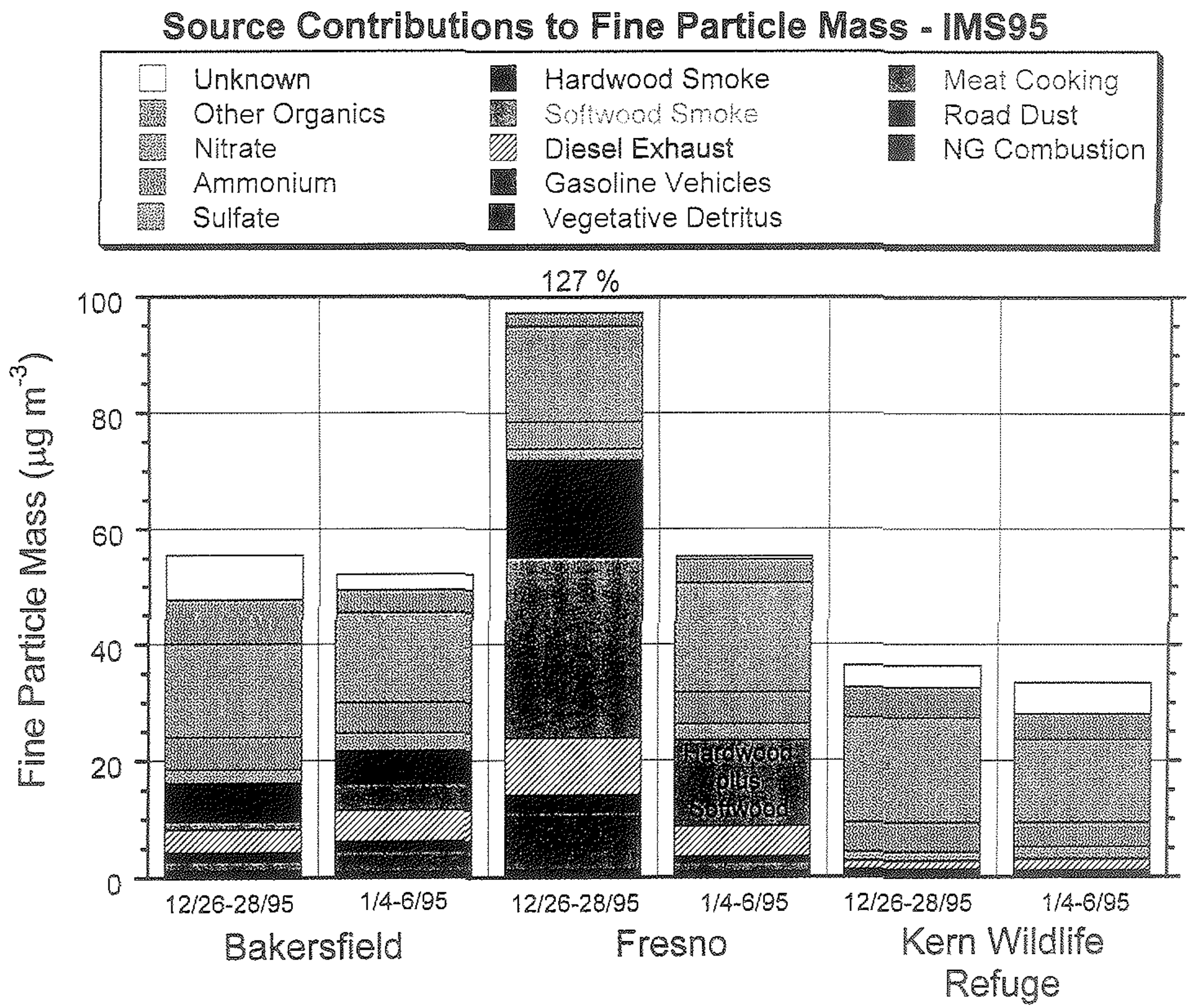


Fig. 1. The chemical structures of some useful organic molecular tracers for particulate air pollution sources.



Source Contributions to Fine Particle Mass - IMS95
Unknown
Other Organics
Nitrate
Ammonium
Sulfate
Hardwood Smoke
Softwood Smoke
Diesel Exhaust
Gasoline Vehicles
Vegetative Detritus
Meat Cooking
Road Dust
NG Combustion
127 %
Hardwood plus Softwood
Fine Particle Mass (µg m-3)
0
20
40
60
80
100
12/26-28/95
1/4-6/95
12/26-28/95
1/4-6/95
12/26-28/95
1/4-6/95
Bakersfield
Fresno
Kern Wildlife Refuge

Northern Front Range Air Quality Study

Douglas Lawson

The Northern Front Range Air Quality Study

Douglas R. Lawson
National Renewable Energy Laboratory
Golden, Colorado 80401

Air pollution along Colorado's Front Range is manifested as visible haze that can range in color from grayish-white to brown. This "brown cloud," caused mainly by airborne particles, is observed most frequently during the winter, when low wind speeds and stagnant conditions accumulate pollutants from diverse sources. During the winter, the brown cloud accumulates in a shallow layer of stagnant air near the South Platte River. To understand the contribution of different pollution sources to the brown cloud, the Colorado General Assembly approved House Bill 1345 in 1995. This legislation established the Northern Front Range Air Quality Study (NFRAQS) to identify sources of air pollution along Colorado's Front Range. The study objectives were reaffirmed in the next session of the General Assembly with passage of HB 96-1179, which expanded the scope of the Study. Nearly 40 government, industry, and research organizations provided funding for the 15 research groups that participated in the program.

The NFRAQS Technical Advisory Panel (TAP) established three policy-relevant objectives for the Study:

- Identify the sources or contributors to $PM_{2.5}$ (airborne particles less than 2.5 micrometers in diameter)
- Determine the role of gas-phase nitrogen oxides, sulfur dioxide, and ammonia in forming ammonium nitrate and ammonium sulfate constituents of $PM_{2.5}$
- Identify the sources responsible for forming ammonium nitrate and ammonium sulfate $PM_{2.5}$.

As the House Bill specified, Colorado State University managed the NFRAQS subject to concurrence on plans, selection of research groups and expenditures by the TAP. Fifteen research groups from throughout the United States participated in the three-year study. The NFRAQS program measured $PM_{2.5}$, which causes Denver's brown cloud. Scientists measured ambient meteorology, visibility, and air quality at several locations in the metro Denver area, north to Fort Collins, and along the South Platte River basin northeast to Fort Morgan during three separate periods – Winter 1996, Summer 1996, and Winter 1997.

Key Findings

The NFRAQS was designed to provide information to policy makers in Colorado who are responsible for managing air quality. The following key findings, based mainly on episodic observations made in Winter 1997, are organized by the Study's policy-relevant objectives.

OBJECTIVE 1 – Identify the sources or contributors to $PM_{2.5}$ in the NFRAQS region

During the winter episodes of increased $PM_{2.5}$ concentrations in the metro Denver area, receptor modeling estimated that the most important sources or contributors to $PM_{2.5}$ were:

- Gasoline vehicle and engine $PM_{2.5}$ exhaust, 28%
- Diesel vehicle and engine $PM_{2.5}$ exhaust, 10%
- Dust and debris, 16%
- Wood smoke, 5%

- Meat cooking, 4%
- Directly-emitted $PM_{2.5}$ from coal-fired power stations, 2%
- Particulate ammonium nitrate (formed in the atmosphere from a variety of sources), 25%
- Particulate ammonium sulfate (formed in the atmosphere from a variety of sources), 10%

During the episodes studied, 75% of the directly-emitted $PM_{2.5}$ from mobile sources was produced by gasoline-powered vehicles and engines and 25% of the directly-emitted $PM_{2.5}$ was produced by diesel-powered vehicles and engines. In contrast, in current emission estimates diesel vehicles are projected to produce more $PM_{2.5}$ emissions than gasoline-powered vehicles. High-emitting or smoking gasoline-powered vehicles, which comprise a small fraction of the in-use vehicle fleet, produced nearly one-half of the gasoline exhaust particles. The diesel exhaust particles come from trucks, locomotives, construction equipment and other sources. $PM_{2.5}$ directly emitted from diesel vehicles and engines was one-third of that from gasoline vehicles and engines, even though diesel-powered vehicles comprise only five percent of the regional vehicle miles traveled. Fine particles from road debris and dust, construction activities, and wind-blown sand contributed 16% of the total $PM_{2.5}$, an amount much lower than current emission estimates.

Particulate ammonium nitrate and ammonium sulfate are formed in the atmosphere from gas-phase emissions of ammonia, nitrogen oxides, and sulfur dioxide. These are called secondary particles because they are not emitted directly by pollution sources. Their sources are discussed in Objectives 2 and 3.

OBJECTIVE 2 – Determine the role of gas-phase nitrogen oxides, sulfur dioxide, and ammonia in the formation of ammonium nitrate and ammonium sulfate $PM_{2.5}$ particles

The NFRAQS region is ammonia-rich. Agricultural operations produced most of the ammonia in the Northern Front Range. Current ammonia emissions would have to be reduced 50% to achieve a 15% reduction in particulate ammonium nitrate levels. Further reductions in ammonia emissions would provide proportional decreases in ammonium nitrate concentrations.

OBJECTIVE 3 – Identify the sources responsible for the formation of ammonium nitrate and ammonium sulfate $PM_{2.5}$ particles

Because of limitations in funding, NFRAQS scientists were unable to completely apportion the contributing sources to ammonium nitrate and ammonium sulfate $PM_{2.5}$ particles. Atmospheric models also have not been adequately developed to model the atmospheric formation of particles from their sources. However, the Study found that the majority of nitrogen oxides, and therefore, particulate ammonium nitrate, are produced by mobile sources. The formation of $PM_{2.5}$ nitrate particles is not a linear process. Reductions of nitrogen oxide emissions, the precursor to particulate nitrate, would result in less-than-proportional reductions in $PM_{2.5}$ ammonium nitrate particles. Three-fourths of the sulfur dioxide emissions are produced by coal-fired power plants. Sulfur dioxide is a precursor to particulate ammonium sulfate.

Northern Front Range Air Quality Study

Final Report

Douglas R. Lawson

HEI Diesel Workshop

Stone Mountain, Georgia

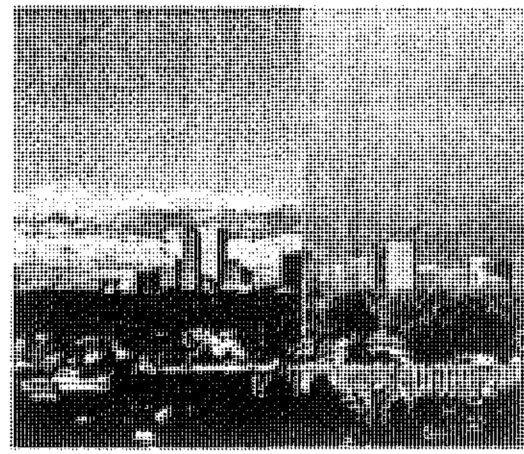
March 8, 1999

Northern Front Range Air Quality Study Sponsors -- $4 Million

- ARCO Coal Company
- Center for Energy and Economic Development
- City and County of Denver
- Colorado Office of Energy Conservation
- Colorado Interstate Gas Company
- Conoco, Inc.
- Coordinating Research Council
- Coors Brewing Company
- Cyprus Amax Mineral Company
- Denver Nuggets
- Eastman Kodak
- Englewood/Littleton Wastewater
- EPRI
- Fort Collins Consortium
    - Anheuser Busch
    - City of Fort Collins
    - Colorado State University/ CIRA
    - Hewlett Packard
    - Larimer County
- Kennecott Energy Company
- KN Energy
- Lockheed Martin
- Metro Denver Wastewater
- Pacific Power Corporation
- Phillips Petroleum
- Platte River Power Authority
- Public Service Company
- Regional Air Quality Council
- Rocky Mountain Hearth Products Association
- Seneca Coal Company
- State of Colorado
- Total Petroleum
- Trigen Colorado Energy Company
- Ultramar/Diamond Shamrock
- U.S. Environmental Protection Agency
- U.S. Department of Energy

Northern Front Range Air Quality Study (NFRAQS) Participants

- Aerosol Dynamics, Inc.
- Air Resource Specialists, Inc.
- Colorado Department of Public Health and Environment
- Colorado State University
- Colorado School of Mines
- Desert Research Institute
- ENSR
- EPRI
- General Motors R&D
- Midwest Research Institute
- National Institute of Standards and Technology
- National Oceanic and Atmospheric Administration



- National Renewable Energy Laboratory
- Regional Air Quality Council
- Sonoma Technology, Inc.
- U.S. Environmental Protection Agency

NFRAQS

Northern Front Range Air Quality Study
Goals and Objectives

Goals

- Attribute sources of existing air pollution in the Denver urban region.
- Collect data necessary to support informed decisions leading to attainment of federal air quality standards.

Policy-Relevant Objectives

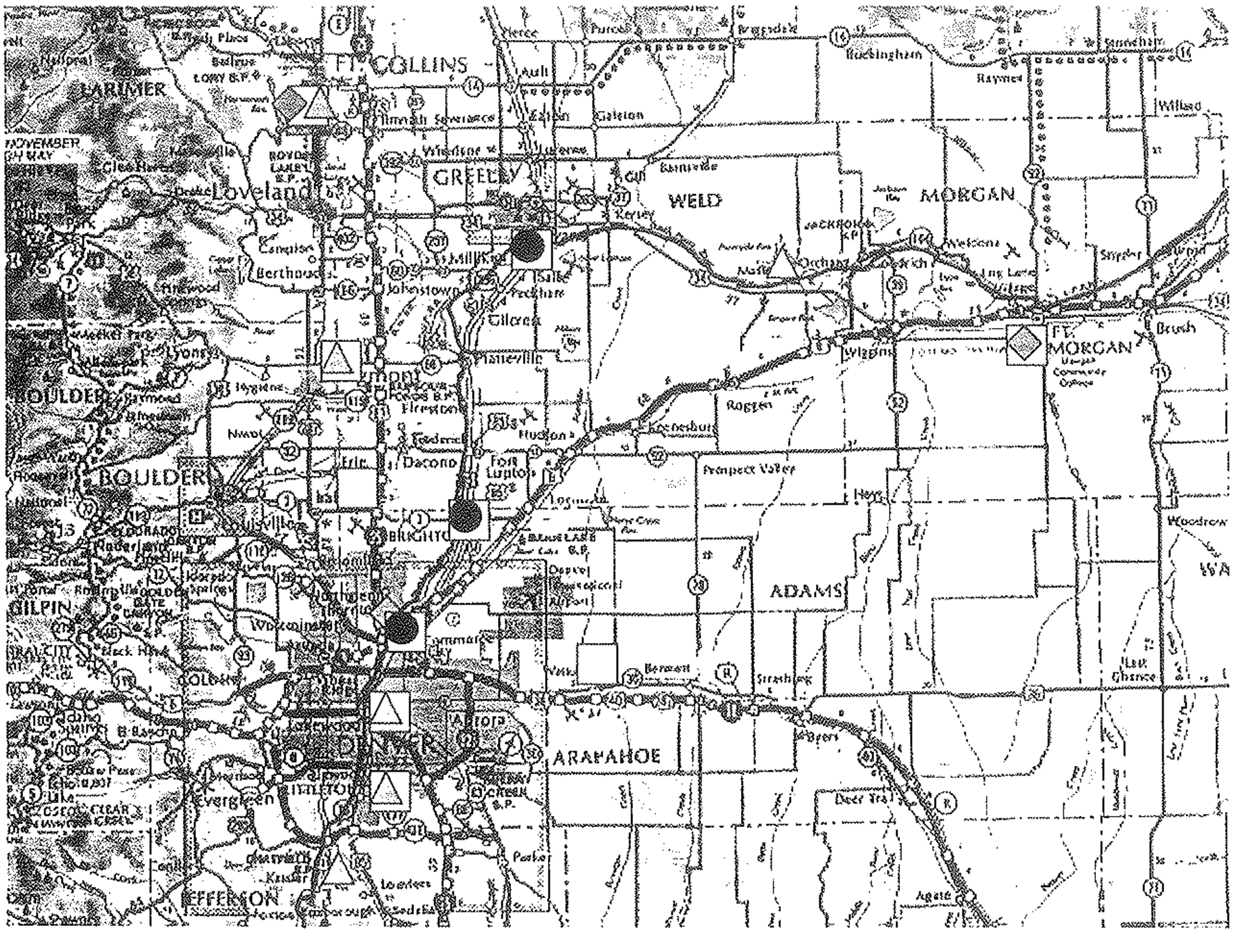
- Perform source apportionment of carbonaceous materials in $PM_{2.5}$ in the NFRAQS region.
- Determine which species (NO_x, SO_2, or NH_3) is limiting with regard to formation of secondary ammonium nitrate and ammonium sulfate.
- Apportion sources leading to the formation of the non-carbonaceous portion of $PM_{2.5}$ with emphasis on nitrate and sulfate.

NFRAQS Timeline

- **Phase 1 -- Winter 1996 Study at Welby**
  January 16-February 29, 1996 (45 days)
- **Phase 2 -- Summer 1996 Study at Welby**
  Satellite sites at Golden (NREL), Longmont, and Fort Collins
  July 17-August 31, 1996 (45 days)
- **Phase 3 -- Winter 1996/97 Study at Welby, Brighton, and Evans**
  Satellite sites at Chatfield Reservoir, Highlands Ranch,
  CAMP (downtown Denver), Longmont, Fort Collins, and Masters
  December 9, 1996-February 7, 1997 (60 days)
- **Draft final report -- December 31, 1997**
- **PUBLIC REVIEW PERIOD: MAY 4--MAY 28, 1998**
- **Final report -- July 1, 1998**
- **Summary report to Governor and Legislature -- December 1998**
- **Updates on study progress: http://nfraqs.cira.colostate.edu**

NFRAQS

Northern Front Range Air Quality Study
Winter 1997 Sampling Sites



Site Type	Location	Symbol
Core	Welby, Brighton, Evans	○
Satellite	CAMP, Fort Collins, Highlands, Longmont, Masters, Chatfield Reservoir	△
Video/35mm Slides	Thornton, Pawnee, Fort Collins	◇
Meteorological Measurements	Various	□

NFRAQS

Northern Front Range Air Quality Study

Policy-relevant Objective 1

Source Apportionment of Carbonaceous Particles

Approach -- Collect samples from most likely sources for chemical analysis and construction of source profiles for receptor modeling

- Light-duty motor vehicles (gasoline and diesel) tested under standard conditions and ambient conditions
- Heavy-duty diesel vehicles
- Wood burning samples -- hardwoods, softwoods, and synthetic logs in fireplace and wood stove
- Charbroiling samples from meat cooking -- beef and chicken
- Road dust samples

NFRAQS

NFRAQS CRC Light-Duty Vehicle Study

Indoor/Outdoor Comparison by Age and Type

83 Vehicles Tested in Denver, Winter 1997

FTP PM Mass Emission Rates, mg/mi

Category	n	Indoors	Outdoors
1991-96	9	3.7	25[a]
1986-90	14	12	28
1981-85	16	36	48
1971-80	15	57	83
"Smoker"	15	395	434
Diesel	12	460	503

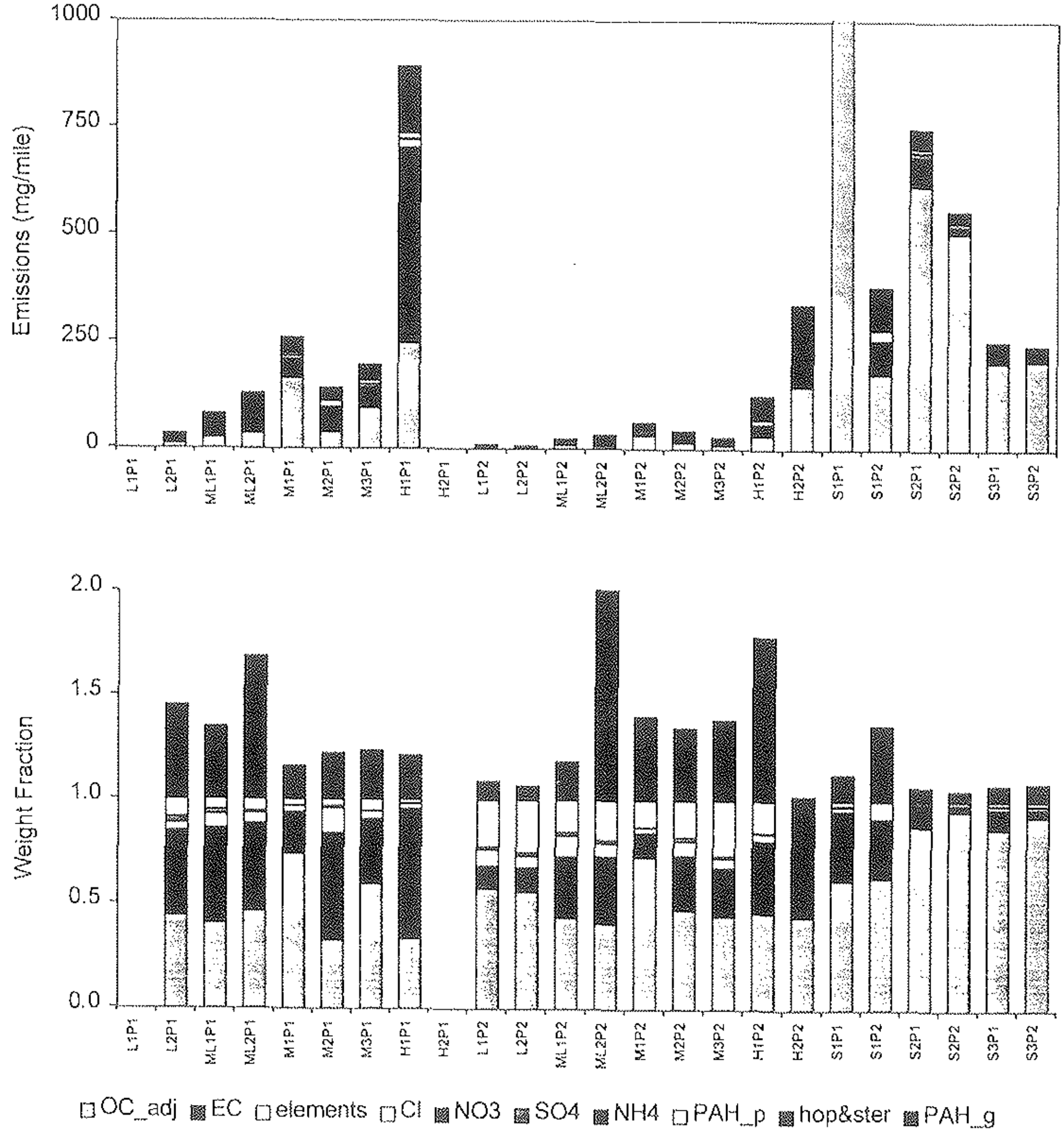
[a]13 without high emitter

NFRAQS

Light-Duty Gasoline Vehicles

Speciated $PM_{2.5}$ Emissions

Emission Rates (mg/mi) and Weight Fractions



NFRAQS

Heavy-Duty Vehicle Testing Cycles

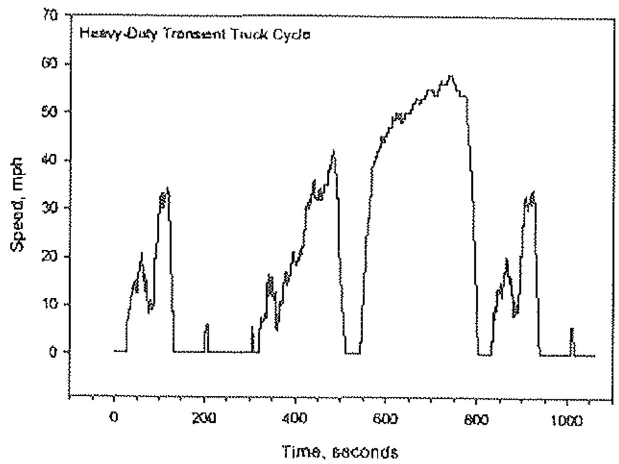


Figure 1. Heavy-duty transient truck cycle.

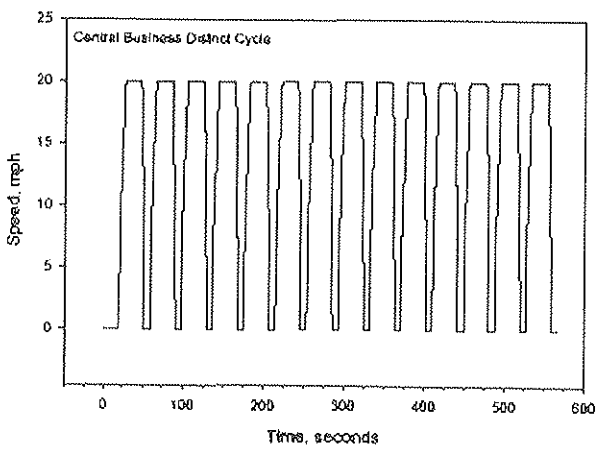


Figure 2. Central business district cycle.

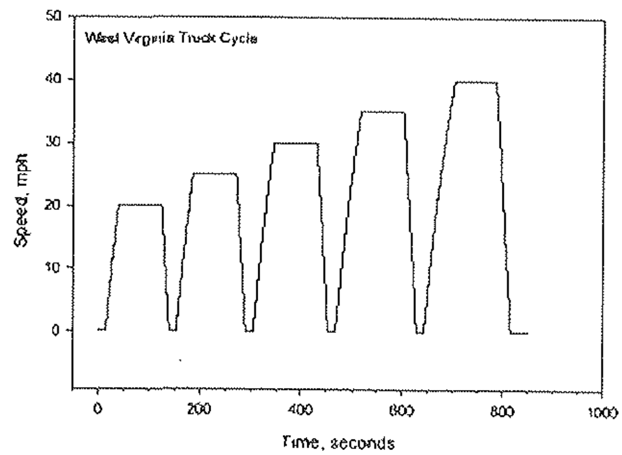


Figure 3. West Virginia truck cycle.

NFRAQS

Colorado School of Mines Study

Heavy-Duty Vehicle Emissions Results
21 Vehicles Tested

Average Mass Emission Rates

Test Cycle	PM_{10}, g/mi	CO, g/mi	HC, g/mi	NO_x, g/mi
Central Business District (CBD)	2.85	30.4	1.98	30.4
Heavy-Duty Transient (HDT)	1.68	16.8	1.31	21.0
West Virginia Truck (WVT)	1.24	9.75	1.90	17.8

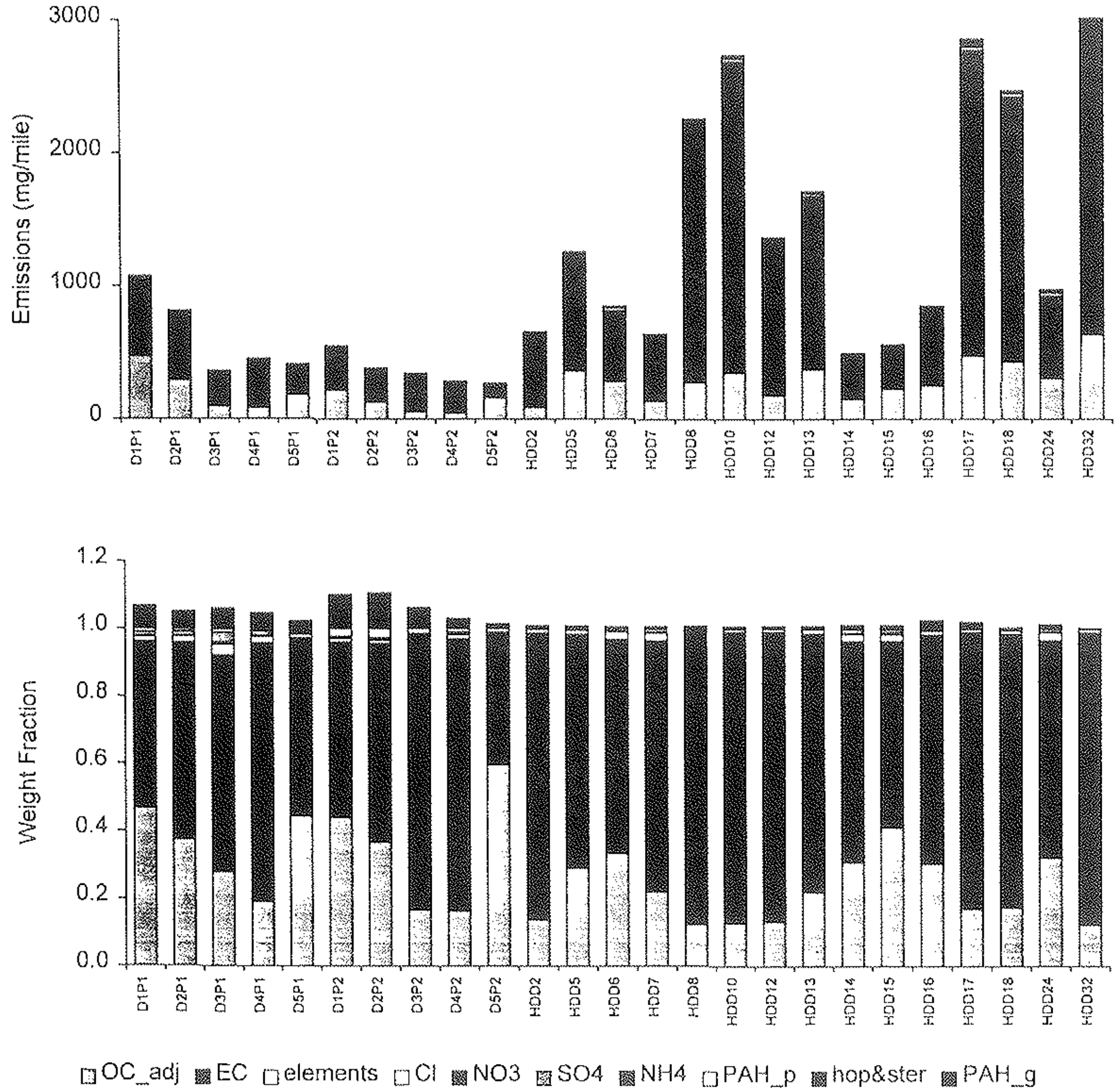
Overall Average PM_{10} Mass Emission Rate = 1.96 g/mi; min = 0.30 g/mi; max = 7.43 g/mi

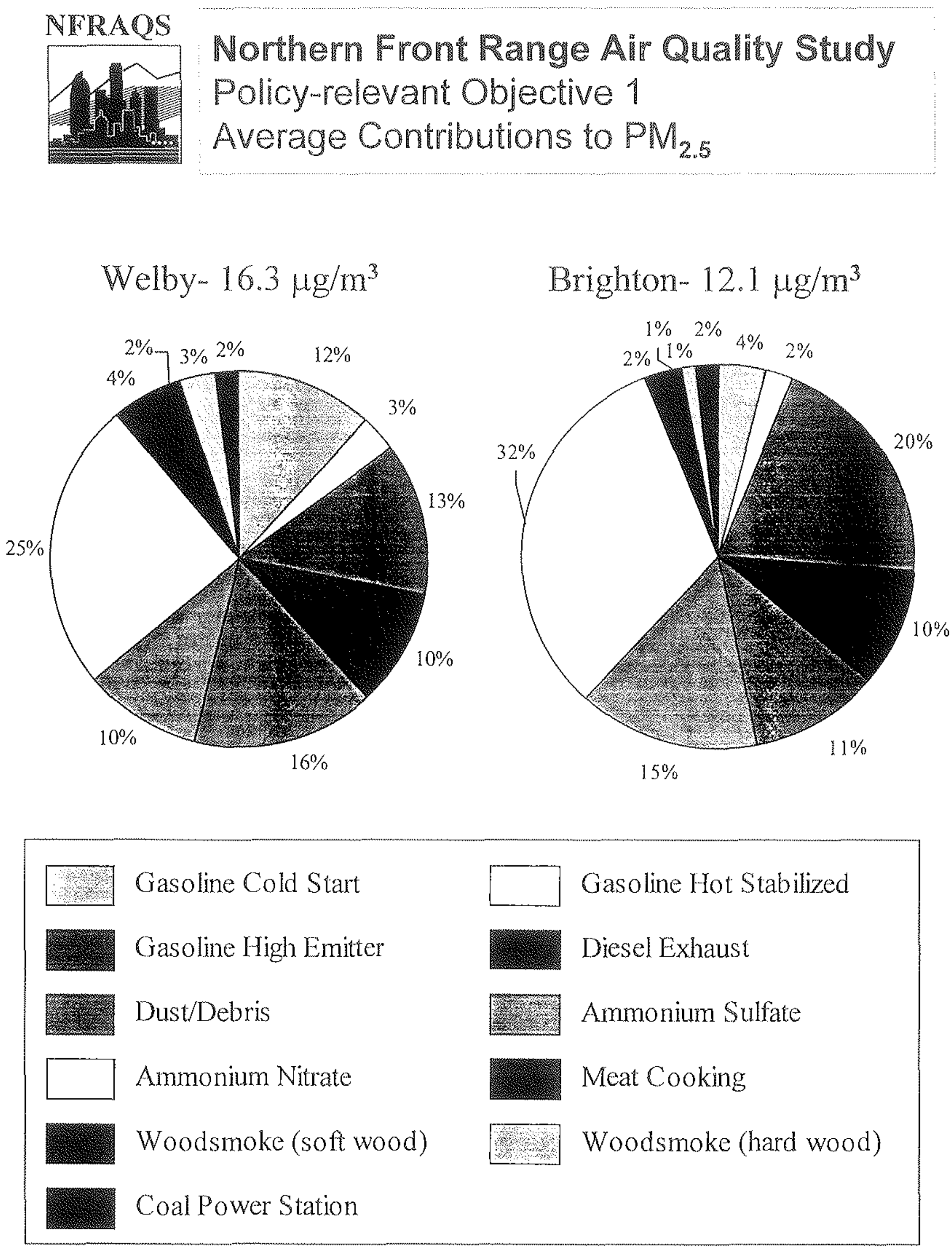
NFRAQS

Light- and Heavy-Duty Diesel Vehicles

Speciated $PM_{2.5}$ Emissions

Emission Rates (mg/mi) and Weight Fractions




NFRAQS
Northern Front Range Air Quality Study
Policy-relevant Objective 1
Average Contributions to PM2.5
Welby- 16.3 µg/m3
2%
3%
2%
12%
4%
3%
13%
25%
10%
10%
16%
Brighton- 12.1 µg/m3
1%
2%
2%
1%
4%
2%
32%
20%
10%
11%
15%
Gasoline Cold Start
Gasoline Hot Stabilized
Gasoline High Emitter
Diesel Exhaust
Dust/Debris
Ammonium Sulfate
Ammonium Nitrate
Meat Cooking
Woodsmoke (soft wood)
Woodsmoke (hard wood)
Coal Power Station

US Department of Energy
Office of Heavy Vehicle Technologies

The Environmental Safety and Health Program at the National Renewable Energy Laboratory

NREL

CENTER FOR TRANSPORTATION TECHNOLOGIES AND SYSTEMS

Diesel Aerosol Sampling Methodology CRC Project E-43 – FY 99 Funds

- Program started September 1998
- $1.5MM project funded by DOE through NREL ($850K), CRC, AAMA, API, EMA, SCAQMD
- In-kind contributions ($435K estimate) from Caterpillar and Cummins
- Investigators are University of Minnesota, West Virginia University, Carnegie Mellon University, and Paul Scherrer Institute (Switzerland)

CENTER FOR TRANSPORTATION TECHNOLOGIES AND SYSTEMS

Diesel Aerosol Sampling Methodology
CRC Project E-43

Program Objectives

- Determine actual particle size distributions and particle number concentrations in exhaust plume from on-road heavy-duty vehicles
- Compare on-road data with data generated in dynamometer testing facilities
- Determine the zone of influence of ultrafine particle emissions from a roadway
- Characterize chemical composition and surface properties of bulk PM emissions

Center for Transportation Technologies and Systems

Diesel Aerosol Sampling Methodology
CRC Project E-43

Augmentations to Univ. MN contract

1. Gasoline engine exhaust (PM and semi-volatiles) emissions testing
2. Chemical analysis of diesel and gasoline exhaust (possible collaboration with CARB)

Center for Transportation Technologies and Systems

Comparative Toxicity Study

- NREL issued RFP on Exhaust Emissions Collection for Comparative Toxicity Testing in August 1998
- Completed peer review on three bids received in response to RFP
- NREL has negotiated contracts with DRI and SwRI team
- 2 Phases for collection of gas and diesel exhaust
  - Dynamometer testing to acquire 2 gr PM and associated semi-volatile compounds
  - Tunnel study testing to acquire 2 gr PM and associated semi-volatile compounds
- 14 samples will be selected by NREL and sent "blind" to Lovelace for comparative toxicity testing between gas and diesel emissions

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Exhaust Samples for Comparative Toxicity Testing

- **Dynamometer Samples at 72 °F (PM and SVOC):**

  1) Ave. gasoline emitters
  2) High PM gasoline emitters
  3) Smoker gasoline emitter(s)
  4) Current technology diesel emitters
  5) High PM diesel emitters

- **Dynamometer Samples at 72 °F (SVOC only):**

  6) Ave. gasoline emitters (same as 1)
  7) Current technology diesel emitters (same as 4)

- **Dynamometer Samples at 30 °F (PM and SVOC):**

  8) Ave. gasoline emitters
  9) Current technology diesel emitters

- **Samples from Fort McHenry Tunnel (PM and SVOC):**

  1) "Diesel" fleet sample plus "background" sample
  2) "Gasoline" fleet sample plus "background" sample

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Exhaust Samples for Comparative Toxicity Testing

14 samples (9-10 from dynamometer and 4-5 from tunnel):

- will be analyzed at DRI for mass, TC, EC, OC, elements by XRF, sulfate, nitrate, and ammonium ions plus organic speciation for PAH, hopanes, steranes, nitro-PAH, and oxy-PAH
- will be sent to Lovelace Respiratory Research Institute for comparative toxicity testing

Composited fuel and lube oil samples from vehicles

- Composited fuel and lube oil from vehicles tested on the dynamometer will be chemically analyzed using standard methods and methods used for ambient measurements

Center for Transportation Technologies and Systems

Models for Population Exposure Estimates

Alison Pollack

Use of Models to Estimate General Population Exposures
To Diesel Particulate Matter

Alison K. Pollack
ENVIRON International Corporation
Novato, CA

Presented at
HEI Diesel Workshop
Stone Mountain, Georgia
March 7-9, 1999

This talk addresses two types of models used to estimate general population exposures to diesel particulate matter: emission inventory models, and exposure models. As the author's area of expertise is in emissions models, that is the focus of the paper, followed by a brief discussion of some exposure modeling studies performed to date. Also, because heavy duty diesel vehicles (HDDV) are the largest source of diesel PM, the focus is further on modeling emissions from this vehicle class.

HDDV diesel PM emissions are estimated by multiplying gram per mile emission factors by total miles traveled by HDDVs. The current EPA and California Air Resources Board (CARB) emission factor models (EMFAC7G and MOBILE5/PART5) for estimating HDDV emission factors are based on the same very limited test data and generally use similar modeling approaches. The only data source available at the time these models were developed is a 1984 joint study between EPA and the Engine Manufacturers Association (EMA). In this test program, only 22 HDDVs were tested; all are around 1980 vintage. For later model years, the models use manufacturer certification data. EPA's model estimates a flat emission factor for the lifetime of the vehicle. CARB's model adds emission factor deterioration over time, estimated from a roadside survey of HDDV tampering and malmaintenance.

To estimate fleet average HDDV emissions, the models first convert the work-specific emissions (in g/bhp-hr) to g/mi emission factors using average fuel economy, fuel density, and brake-specific fuel consumption; this calculation is done by weight class and by model year. These emission factors are aggregated across weight classes using sales estimates, and then composited across model years using model year age and mileage accumulation distributions.

Both EPA and CARB are currently working on major updates to their mobile source emission factor models, to be released later this year. The updates may incorporate newly available chassis dynamometer data, and will also incorporate new manufacturer certification data. In addition, conversion factors will be updated (using revised fuel economy estimates), weight class sales fractions will be updated, age and mileage accumulation distributions will be updated, and off-cycle driving effects will be added.

The HDDV emission factors are multiplied by estimates of HDDV vehicle miles traveled (VMT) to estimate total HDDV emissions. VMT estimates are typically derived from transportation models. While a few local air quality planning agencies have travel models specifically for heavy-duty vehicles, the most common approach is to estimate overall VMT by roadway, and to apportion that to the various vehicle classes. This means that heavy-duty vehicles are assumed to incur the same fraction of VMT on highways as on local roadways, clearly a problematic assumption.

As an example of exposure modeling for diesel toxics, the CARB 1998 results are briefly discussed. CARB used the emission inventory developed with EMFAC7G as described above. Chemical mass balance (CMB) was used to estimate primary PM_{10} from motor vehicles; some spatial interpolation/extrapolation from the existing air quality monitoring data was required to cover all counties in the state. Indoor exposures were estimated using the California Population Indoor Exposure Model (CPIEM), which relies upon estimates of population location and indoor/outdoor activity patterns as well as building ventilation rates. Statewide, the average general population ambient exposure in 1990 was estimated to be 3.0 $\mu g/m^3$, and total exposure (i.e., indoor and outdoor) was estimated to be 2.1 $\mu g/m^3$.

As many exposure estimates rely upon emission inventory estimates, the shortcomings in emission inventory models must be understood. The current emissions estimates of on-road diesel PM are based on very little real-world test data and thus are highly uncertain. The activity estimates also have large uncertainties. Off-road diesel PM emissions are even less well characterized; while there are new models to estimate off-road emissions, they are based on extremely limited data. Currently there are several large-scale studies underway, of both emission factors and activity data, that will improve our estimates of heavy-duty diesel emissions. The uncertainties in the existing emissions models need to be estimated, and these uncertainties need to be included in exposure modeling so that total uncertainty of predicted exposures can be estimated.

ENVIRON

Use of Models to Estimate Population Exposures To Diesel Particulate Matter

Alison K. Pollack
ENVIRON International Corporation
Novato, CA

Presented at
HEI Diesel Workshop
Stone Mountain, Georgia
March 7-9, 1999

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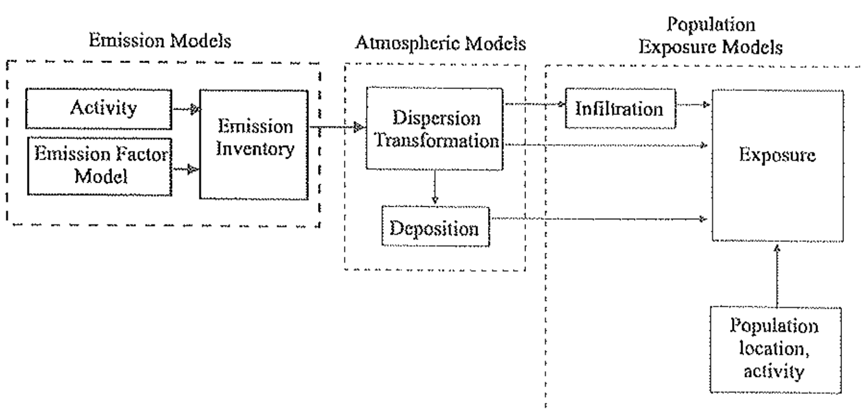
Exposure Modeling



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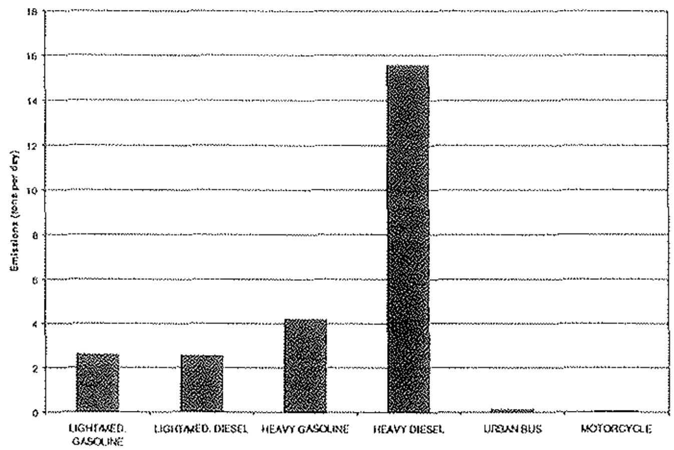
Particulate Matter Emissions by Vehicle Class 1995 South Coast Air Basin



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Federal PM Emission Standards for New Heavy-Duty Diesel Vehicles (HDDVs)

Model Year	PM (g/bhp-hr)
1985 - 1987	None
1988 - 1990	0.60
1991 - 1993	0.25
1994 +	0.10

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Emission Factor Models for Diesel PM_{10} Exhaust

CARB: EMFAC7G is current
EMFAC99 to be released soon

EPA: MOBILE5/PART5 are current
MOBILE6 to be released later this year

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Current Emission Factor Model Estimates of HDDV PM

Data for Basic Emission Rates

- Engine dynamometer testing
- 1984 EPA/EMA study
  - 22 1979/1980 model year HDDV's
- Manufacturer certification testing
- Tampering and malmaintenance study (ARB)

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Current Emission Factor Model Estimates of HDDV PM (continued)

Modeling Methodology

- Convert work-specific emissions to g/mi emission factors
- Calculate sales-weighted average across HDDV weight classes
- Composite across model years to derive fleet emission factor
  - Age distribution
  - Mileage accumulation rates

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Current Emission Factor Model Estimates of HDDV PM (continued)

Modeling Methodology (concluded)

- ARB correction factors
  - Clean diesel fuels
  - Heavy-Duty Vehicle Inspection Program effects
  - Out-of-state vehicle adjustments

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Current Emission Factor Model Estimates of HDDV PM (concluded)

Conversion Factors

Emission Factor (g/mi) =

Work-Specific Emission Level (g/bhp-hr) * Conversion Factor (bhp-hr/mi)

Conversion Factor =

$$\frac{\text{Fuel Density (lb/gal)}}{\text{Brake-Specific Fuel Consumption (lb/bhp-hr)} * \text{Fuel Economy (mi/gal)}}$$

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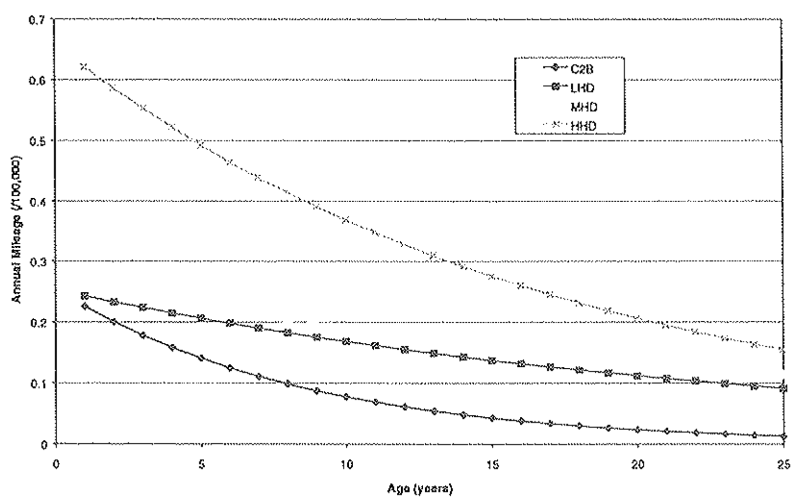
MOBILE5a Default HDDV Mileage Accumulation by Weight Class



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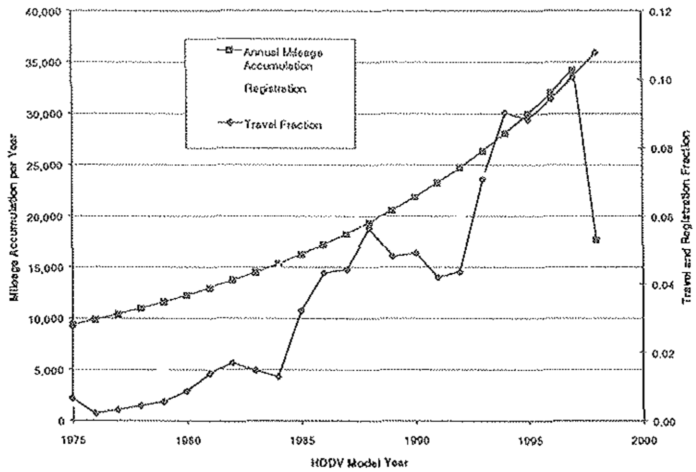
MOBILE5b Default HDDV Travel Characteristics, Jan. 1999 Fleet



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Emission Factor Model Updates

- New chassis dynamometer data
- New certification test data
  - Zero-mile emissions and deterioration rates
- Update conversion factors using updated fuel economy
- Additional weight classes
- Updated sales by weight class
- Updated age and mileage accumulation distributions
- Off-cycle driving effects added

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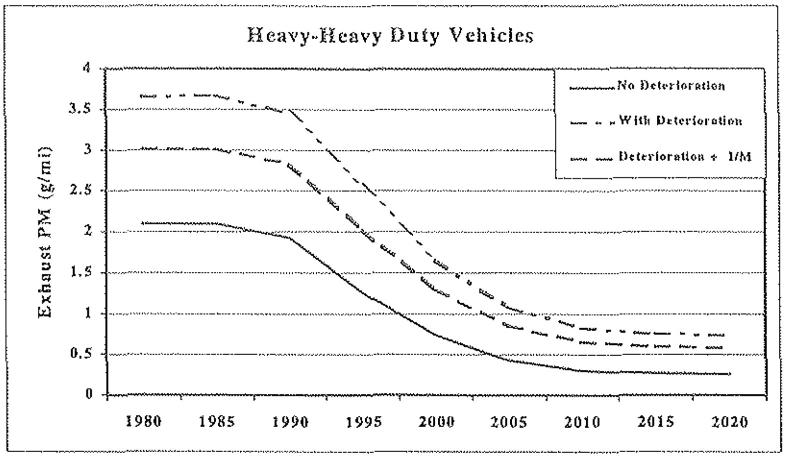
PART5 Projections for Heavy-Heavy Duty Trucks



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Emission Inventory Modeling: Spatial and Temporal Allocation of Emissions

Simple

- VMT by county apportioned to grid cells
- Day of week and time of day profiles

Complex

- Transportation models predict link-specific VMT by time of day

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Emissions Modeling Issues

- Models are based mostly on certification data, not real-world testing
- Need in-use emissions data
- Model validation studies are needed
- Improved activity estimates are needed

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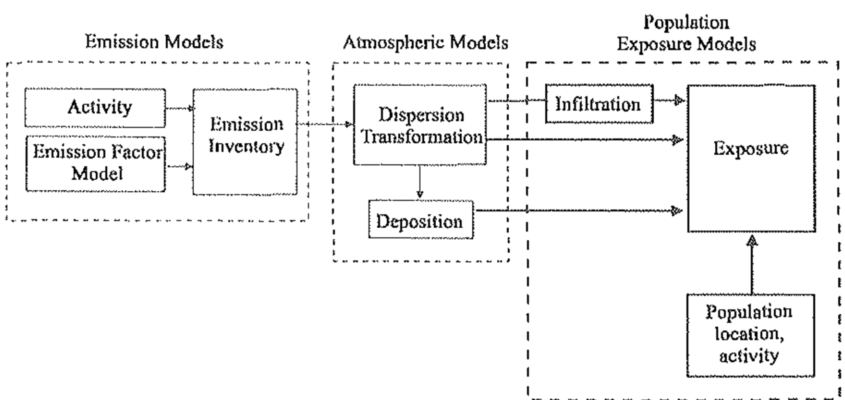
ENVIRON

Exposure Modeling



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Diesel Exhaust Components

- No unique tracers known
- Key Identifying Components

Elemental C	(in absence of other major sources)
Alkyl Penanthrenes	(PAH derivative)
1- Nitropyrene	(Major nitro-PAH constituent)
Alkanes/[Branched + Cyclic HCs]	(high ratio is diesel exhaust)

- Transformation products likely to be significant

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Exposure Modeling Approaches

Source Oriented (Inventory Based)

Pro

- Explicit simulation of transformation, deposition processes possible
- Flexible receptor network arrangement

Con

- Heavy reliance on accuracy of emission inventory
- National modeling requires significant simplifications (e.g. ASPEN)

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ENVIRON

Exposure Modeling Approaches (concluded)

Receptor Oriented

Pro

- Based on observed concentrations
- Factor analysis requires no source profiles (e.g. UNMIX)
- CMB allows apportionment to inventory source categories

Con

- Uncertainties in source apportionment
- Uncertainties in interpolation from monitors to population receptors

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Exposure Studies

- Gray (1986)
  - Los Angeles c. 1982
  - Diesel contribution to observed $PM_{2.1}$ Elemental Carbon from emission inventory is 67%
- Gray et al. (1989)
  - Los Angeles (1986)
  - CMB analysis of PM_{10} data
  - LD diesel contributed 3.6 - 5.6 $\mu g/m^3$ annual average PM_{10}
    6.2 - 22.0 $\mu g/m^3$ 24-hour peak PM_{10}

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Exposure Studies (continued)

- ARB Diesel Toxics (1998)
  - California exposures 1990, 1995, 2000, 2010
  - CMB used to estimate primary PM_{10} from MVs
  - EI estimate of diesel PM_{10} emissions fraction
  - Spatial interpolation from monitoring sites
  - Add indoor exposures via building ventilation rate estimates (CPIEM model)
  - Average diesel PM_{10} exposure = 2.1 ∀ 0.8 μg/m^3
  - "Hot Spot" estimate for Long Beach Freeway
    - 8 μg/m^3 above background (24-hour avg)

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ENVIRON

Average Ambient Outdoor Concentrations of Diesel Exhaust PM_{10} in California for 1990

Air Basin	Air Basin Population	Diesel Exhaust PM_{10} (μg/m^3)
Great Basin Valleys	29,000	0.2
Lake County	51,000	0.3
Lake Tahoe	21,000	1.0
Mojave Desert	557,000	0.8
Mountain Counties	485,000	0.6
North Central Coast	622,000	1.4
North Coast	564,000	1.2
Northeast Plateau	80,000	1.1
Sacramento Valley	2,219,000	2.5
Salton Sea	330,000	2.6
San Diego	2,504,000	2.9
San Francisco Bay Area	5,967,000	2.5
San Joaquin Valley	2,658,000	2.6
South Central Coast	1,232,000	1.8
South Coast	12,809,000	3.6
Statewide Population-Weighted Concentration	**30,131,000**	**3.0**

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Exposure Studies (concluded)

EPA MVRATS (1993)

1. Assume 5% ambient TSP due to diesel exhaust (EI) and adjust for time spent indoors = 1.5 $\mu g/m^3$ national average diesel PM
2. HAPEM - MS
   - Use CO exposure as surrogate for diesel
   - NEM approach to exposure (11 urban areas)
     - Define cohorts based on age, occupation, home/work locations,
     - Activity levels from Cincinnati diary study
     - Ambient CO from NAMS/SLAMS
     - "Exposure factors" from Denver winter personal exposure monitoring study
   - National extrapolation
     - Urban average $TSP_{diesel} = 2\ \mu g/m^3$

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ENVIRON

Estimates of Diesel Exhaust Ambient PM Concentrations by Selected Researchers

Conditions/Method	Year	Concentrations ($\mu g/m^3$)	Reference
Ambient Concentration Estimates Using:			
Dysprosium Tracer – range	1988	5-23	Horvath, et al., 1988
- typical	1988	11	
Lead Surrogate – range	1995	0.7 – 3.9	U.S. EPA, 1983
Elemental Carbon – range Surrogate	1982	3.4 – 5.7	Adapted from Denton, et al., 1992
NAAQS Exposure Model – range	1995	3.1 – 3.7	U.S. EPA, 1983
NAAQS Exposure Model modified	1986	2.6	Ingalls, 1985
Source Apportionment-Urban Areas	1991	4 – 22	Chow et al., 1991

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ENVIRON

Estimated Annual Ambient Concentrations of Diesel Exhaust Particulate Matter ($\mu g/m^3$) (EPA MVRATS, 1993)

Year	Method 1 Rural	Method 1 Urban	Method 1 National	Method 2 National
1990	1.1	2.0	1.8	1.5
1995	0.6	1.2	1.1	
2000	0.4	0.7	0.6	
2010	0.2	0.4	0.4	

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EPA OMS Revised Motor Vehicle Air Toxics Study

- Compute exposures using HAPEM-MS3 model (use CO as surrogate)
  - No differentiation between LD and HD diesel activity patterns
- Use ASPEN national dispersion model
  - Requires allocation of diesel emissions to census tracts
- Use NFRAQS and other recent PM source apportionment studies to estimate ambient diesel PM

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ENVIRON

Exposure Modeling Research Needs

- Diesel contributions to exposure to secondary pollutants (including gas phase and gas ↔ particle interactions)
- Identify better tracers of diesel exhaust
- Updated/expanded population activity data
- Expanded personal exposure monitoring studies

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ENVIRON

Summary Comments on Use of Emissions Models

1. Current on-road PM emissions models are based on very little real-world test data, and are highly uncertain.
2. HD activity estimates are also highly uncertain, but improving.
3. Off-road diesel PM emissions models are also based on very little data.
4. Need to characterize uncertainty in emissions inventory, and factor that into exposure estimates.

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SESSION IV

What Do Published Epidemiology Studies Tell Us About Exposure-Response?

Executive Summary: *Diesel Emissions and Lung Cancer: Epidemiology and Quantitative Risk Assessment.* A Special Report of the Institute's Diesel Epidemiology Expert Panel

John C. Bailar, III, Chair

David Hoel

Ethel Gilbert

Brian Leaderer

Charles Poole

Robert F. Sawyer

G. Marie Swanson

Preliminary findings of the HEI Diesel Epidemiology Expert Panel were presented at the workshop. Here, the Executive Summary of the Panel's final report is included. The full report can be found at *www.healtheffects.org*.

EXECUTIVE SUMMARY

Diesel Emissions and Lung Cancer: A Special Report

Diesel engines are an important part of the world's transportation and industrial infrastructure, especially in heavy-duty applications such as trucks, buses, construction and farm equipment, locomotives, and ships. Energy efficiency and durability account for the dominant use of diesel engines worldwide, and their use may expand in the future. In Europe, 20% to 50% of the new light-duty passenger fleet is powered by diesel engines. Although the percentage of diesel-powered light-duty vehicles is much lower in the United States, advanced technology diesel engines are being proposed as part of the nation's energy conservation and climate change strategies.

The economic advantages of diesel engines are clear; nevertheless, environmental concerns and related health issues must be addressed. Emissions from all types of engines are highly variable and complex mixtures. Diesel engines are more efficient than gasoline engines, and they emit less carbon dioxide (a greenhouse gas), carbon monoxide, and hydrocarbons. Therefore, diesel engines have some advantages over conventional gasoline engines in terms of global warming. However, they emit higher levels of oxides of nitrogen, which are ozone precursors, and particulate matter per vehicle mile traveled than do gasoline engines. The particulates are of special concern in possible health outcomes; they are small enough to be readily respirable, and they have many chemicals adsorbed to their surfaces, including known or suspected mutagens and carcinogens.

Cellular, animal, and human studies have investigated the association between exposure to diesel exhaust and adverse health effects, including cancer. Lung tumors have occurred in rats exposed to diesel exhaust, but the relevance of these lesions to human risk assessment has been questioned. Epidemiologic studies fairly consistently show an elevation in lung cancer rates among occupationally exposed individuals. In most studies, rates are 20% to 50% greater than those in unexposed individuals; however, these studies did not obtain quantitative measurements of exposure during the time period of the study.

Although epidemiologic data have been used generally to identify the hazards associated with exposure to diesel exhaust, questions remain as to whether the human data can be used to develop reliable estimates of the magnitude of any risk for lung cancer (that is, through quantitative risk assessment [QRA]), and whether new research efforts could provide any additional data needed. In response to such issues, the Health Effects Institute initiated the Diesel Epidemiology Project in 1998. The Project includes the evaluation by HEI's Diesel Epidemiology Expert Panel of occupational epidemiologic studies that have been used for QRA, and the development of new research initiatives to improve understanding about the health effects of diesel exhaust.

The Diesel Epidemiology Expert Panel was chaired by John C. Bailar, III, M.D., Ph.D., of The University of Chicago and the HEI Review Committee, and included six other scientists who have expertise in epidemiology, biostatistics, exposure characterization, and exposure assessment. It was charged to (1) review the epidemiologic data that form the basis of current QRAs for diesel exhaust, (2) identify data gaps and sources of uncertainty, (3) make recommendations about the usefulness of extending or conducting further analyses of existing data sets, and (4) make recommendations for the design of new studies that would provide a stronger basis for risk assessment.

Although lung cancer was the health outcome of interest to the Panel's charge, it was not charged to evaluate either the broad toxicologic or epidemiologic literature concerning exposure to diesel exhaust and lung cancer for hazard identification purposes, which has been done by others. State, national, and international agencies have all reviewed the broader animal and human evidence for carcinogenicity and, in either their draft or final reports, have all identified diesel exhaust as a probable human carcinogen or placed it in a comparable category (National Institute for Occupational Safety and Health 1988; International Agency for Research on Cancer 1989; World Health Organization 1996; National Toxicology Program 1998; Office of Environmental Health Hazard Assessment [California Environmental Protection Agency]

1998; U.S. Environmental Protection Agency 1998).

In response to the first charge, the Panel examined published epidemiologic studies of diesel exhaust emissions and lung cancer for possible use in support of QRA. Only two such studies reported any quantitative exposure data associated in some manner with the occupational epidemiologic studies, and they were considered in the Panel's review.

The Panel recognized that no epidemiologic study can be perfect. Therefore, the Panel viewed its task as addressing the question: To what extent can limitations in the design and performance of a particular study affect its contribution to the body of epidemiologic knowledge under examination for QRA? The Panel also recognized that frequently it is very difficult to obtain retrospective data for estimating job-related work exposures, and that this process may require assumptions that cannot be validated. In the studies considered here, which form the core of the Panel's review, reasonable attempts were made to reconstruct past exposures to diesel engine emissions using approaches that were feasible when the studies were conducted. These data subsequently have been used, in some cases, for purposes that were not envisioned by the original investigators. The studies reviewed for this report include:

Railroad Worker Studies

- Case-control: Garshick et al. 1987
- Cohort: Garshick et al. 1988
- Industrial hygiene: Hammond 1988, and Woskie et al. 1988a,b
- Exposure-response analyses: Crump et al. 1991, Office of Environmental Health Hazard Assessment 1998, and Crump 1999

Teamster Studies

- Case-control: Steenland et al. 1990, 1992
- Industrial hygiene: Zaebst et al. 1991
- Exposure-response analysis: Steenland et al. 1998

The reports of these studies were supplemented by published articles and by presentations to the Panel by the principal investigators and others, including secondary analysts of the railroad worker data. The Panel did not consider other completed lung cancer and diesel epidemiologic studies because they included no directly associated quantitative exposure data.

Certain strengths are evident in the studies reviewed by the Panel. The epidemiologic studies include large numbers of study subjects (55,407 subjects, and 1,694 lung cancers, for the railroad worker cohort study; 1,256 deaths from lung cancer for the railroad worker case-control study; and 996 deaths from lung cancer for the teamster case-control study), all of whom were employed in industries where many workers are exposed to diesel exhaust. Job categories with known exposure to asbestos were either excluded or controlled for in the analyses. Both of the case-control studies adjusted data analyses to control for cigarette smoking as a confounding variable. Overall, the results are generally consistent with findings of a weak association between lung cancer and exposure to diesel exhaust. However, published secondary analyses of exposure-response relations in the railroad worker cohort data produced conflicting results (Crump et al. 1991; Office of Environmental Health Hazard Assessment 1998).

Measurements from the industrial hygiene studies in general supported the job exposure categories used in the epidemiologic studies. The industrial hygiene studies measured different markers for diesel exhaust exposure-respirable-sized particles (RSP) for railroad workers and submicron-sized elemental carbon (EC_1) for teamsters. Although the RSP measures were adjusted for the environmental tobacco smoke component, EC_1 is more sensitive and specific to diesel exhaust than adjusted RSP.

In response to the second charge, the Panel developed a framework of general epidemiologic questions about study design, exposure assessment, outcome determination, and analysis. These are meant to help in systematically understanding and revealing the strengths and uncertainties of these studies. This framework was then used to evaluate the studies of railroad workers and teamsters. This process helped to address the third and fourth charges to the Panel, and to assist HEI in focusing its future research directions to inform apparent gaps for QRA.

The original findings of the cohort railroad worker study reported by Garshick and coworkers (1988) indicated a steadily increasing risk of lung cancer for exposed workers with increasing years of employment. This increase with duration of employment, however, was not supported in later, unpublished analyses (Garshick 1991). This increasing risk, plus the availability of some quantitative exposure data in railroad workers (Woskie et al. 1988a,b), prompted additional analyses to explore the exposure-response relation in these data (Crump et al.

1991; Office of Environmental Health Hazard Assessment 1994, 1998; Crump 1999). Crump and colleagues found a negative association between lung cancer risk and several measures of cumulative exposure; that is, risk decreased with increasing cumulative exposure. In contrast, the statistical models used by the Office of Environmental Health Hazard Assessment analysts, using the same data but different assumptions, showed a positive association in which risk increased with increasing cumulative exposure.

The Panel explored these apparent inconsistencies in the exposure-response relation to verify and obtain a better understanding of the previous analyses, and to help clarify differences. These issues are central to whether the railroad worker data can be useful in a QRA for lung cancer.

The Panel's data exploration demonstrated that within the three broad railroad job categories of train workers (e.g., engineers, conductors), shop workers (e.g., electricians, machinists), and clerks and signalmen, the relative risk of lung cancer decreased with increasing duration of employment, and this decrease was statistically significant for the clerks/signalmen and train workers. Although the relative risk decreased with increasing duration of employment, overall risks for train workers, within each duration of employment group, were higher than those for clerks and signalmen, and shop workers had intermediate risks (Figure 1).

These findings are not consistent with a steadily increasing association between cumulative diesel exposure and lung cancer risk. Furthermore, if the difference in risk between train workers and clerks/signalmen was due primarily to differences in exposure to diesel emissions, one would expect the relative risk for train workers compared with that for clerks and signalmen to be reduced or even eliminated after adjusting for exposure. In fact, adjustment for exposure increased this relative risk. Such a systematic pattern of decreasing risk with increasing exposure suggests that some form of bias is present in the data, which makes it difficult to determine the true nature of an exposure-response relation. Bias can result from uncontrolled confounding by cigarette smoking or by other occupational exposure, differential misclassification of exposures by job category, longer survival of "healthier" workers, or differential ascertainment of lung cancer as a cause of death.

Initial findings from the teamster case-control study (Steenland et al. 1990) showed an increased risk of lung cancer with increasing years of employment. The investigators published an exposure-response analysis for the teamster study (Steenland et al. 1998) after the Panel's work started, thus the evaluation of this set of studies was necessarily less extensive.

Reconstructing past exposures for which actual data are limited or nonexistent requires several assumptions. The Panel had concerns about several of the assumptions used by Steenland and colleagues in the exposure-response analysis of the teamster data. These concerns include (1) the data on 1990 emissions used to estimate past exposures to diesel exhaust may underestimate average exposures over a range of work histories, given that more recent data show higher emissions for that time; (2) the date assumed for dieselization in the trucking industry, which, if too early, may overestimate exposures; (3) the degree to which vehicle miles traveled accurately reflects actual exposure to diesel exhaust for various job groups, which may affect exposure estimates in either direction; (4) the possible effects of using various scenarios of emission levels to account for long fleet turnover times in the trucking industry; and (5) the difficulty in distinguishing truck driver exposures from background levels, because measured estimates are close. Also, among the assumptions Steenland and colleagues used, nondiesel sources of elemental carbon in ambient air, especially from gasoline engine emissions, were not considered.

The Panel also was concerned about the controls used in the case-control study. Lung and bladder cancers and motor vehicle accidents were excluded as control causes of death, and controls were selected from other causes. If those causes of death were associated with exposure to diesel emissions, smoking, or both, the study findings could be biased.

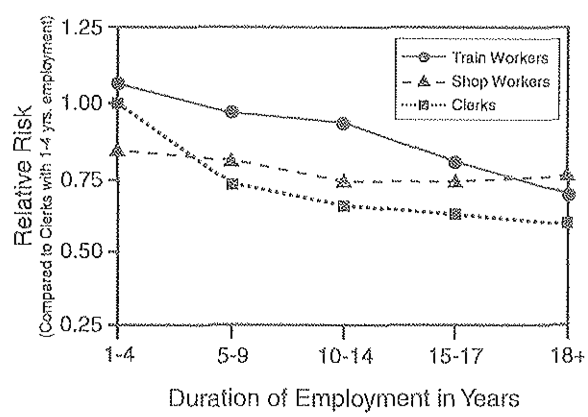


Figure 1. Panel's analysis depicting consistently elevated risk of lung cancer for train workers compared with clerks for each time period, but decreasing risk by job category over duration of employment. See Appendix C in the full report for details.

Important work is currently under way to study the health effects of exposure to diesel exhaust in nonmetal miners in Germany (Säverin et al. 1998) and in the United States (National Cancer Institute–National Institute for Occupational Safety and Health 1997). The Panel did not review these studies because they are still in progress. However, the Panel heard presentations from these investigators at the HEI Diesel Workshop: Building a Research Strategy to Improve Risk Assessment (HEI 1999) at Stone Mountain, GA, March 7–9, 1999. In particular, the National Cancer Institute–National Institute for Occupational Safety and Health study is large and appears to be well designed and comprehensive. It includes a cohort and nested case-control component, as well as extensive current measurements of exposure to diesel exhaust, detailed reconstruction of historical exposure, and biomarker development. These studies in progress are likely to inform hazard identification, exposure estimation, and exposure-response analyses, all components of risk assessment.

The Panel recognizes that regulatory decisions need to be made in spite of the limitations and uncertainties of the few studies with quantitative data currently available. The findings described here and the systematic evaluation of these and other studies are designed to inform the ongoing process and provide a means to weigh a study's strengths and limitations.

FINDINGS

General

Enhanced exposure and epidemiologic data and analyses are needed for the purposes of QRA; these might come from further exploration of existing studies or from new studies.

Railroad Worker Studies

At present, the railroad worker cohort study (Garshick et al. 1988), though part of a larger body of hazard identification studies, has very limited utility for QRA of lifetime lung cancer risk from exposure to ambient levels of diesel exhaust for the following reasons.

- The various exposure-response analyses are limited by the scope and quality of currently available exposure data. Quantitative exposure data were not obtained during the cohort study period. Also, there is a paucity of qualitative data on individual exposures before 1959, and on the variation in exposure by railroad site, by season, and over time. The potential impact of concurrent exposures (for example, to grease, dust, other fumes, asbestos, and active and passive cigarette smoke) were not examined in depth. The diesel exhaust exposure data are suitable for a crude categorical measure of exposure by job category; but other measures, including duration of employment in a job category exposed to diesel exhaust, intensity of exposure concentration (mg/m3), and lifetime exposure ([mg/m3]-years), are not adequate to support quantitative exposure-response analyses.
- The Panel's analysis of the exposure-response association in the railroad worker data showed that the evidence for a positive association of lung cancer with cumulative exposure to diesel exhaust depends entirely on differences in risks among job categories. Train workers (with higher exposures) have higher risks compared with clerks (with low or no exposure). However, within all job categories, the relation of lung cancer risk to duration of employment is negative.
- Factors that might explain a negative association between duration of employment and lung cancer in these data include bias introduced by systematic differences in exposure misclassification among and within job categories; differentially incomplete ascertainment of lung cancer deaths by job category; lack of information on other occupational exposures and air pollutants; the presence of a healthy worker survivor effect; confounding by cigarette smoking; and analysis of relative risks rather than absolute risks. Also, in a case-control study, if causes of death among controls were associated with exposure to diesel exhaust, smoking, or both, the results could be biased.

Teamster Studies

The investigators' analysis of the teamster data reported an exposure-response relation (Steenland et al. 1998) that may be useful for QRA; this relation will be better understood with further exploration of uncertainties and assumptions, particularly those relating to the reconstruction of past exposures and the selection of controls. Exposures of teamsters are more similar to ambient exposures of the public than are exposures of railroad workers, and the diesel exhaust to which teamsters are exposed *comes from a source that is likely to be relevant to regulatory issues.*

The Panel reviewed the teamster study without the benefit of additional analyses and interpretations, and its comments are not as detailed as those about the railroad worker studies. Understanding the teamster study will evolve with time; however, some conclusions can be drawn now.

- The set of teamster studies may provide reasonable estimates of worker exposure to diesel exhaust, but significant further evaluation and development are needed. The marker for diesel exhaust that was selected for study by Steenland and associates, EC1, is more sensitive and specific than RSP adjusted for environmental tobacco smoke, but has several limitations (e.g., the contribution of diesel emissions to ambient EC1 concentrations has not been constant over time). The industrial hygiene study, which was conducted after the period when workers in the case-control study were exposed, identified a range of exposures for various job categories, but did not consider (1) site-to-site variations, (2) seasonal variations, (3) concurrent exposures to other agents, (4) historical ambient particle concentrations, or (5) intra- and interindividual variability. The estimation of historical exposures needs to incorporate recent data on diesel emissions from vehicles in use, reassessment of when dieselization occurred, alternatives to estimating exposure by vehicle miles traveled, and historical regional ambient pollution data.
- The exposure-response relation reported in the teamster study increases in a linear manner. However, more can be learned from other analysts examining these data using different approaches.
- Neither a roster of the study population nor an alternative method of selecting controls to represent it was available to the researchers. It cannot be established with certainty whether the causes of death used for controls adequately represent the joint distribution of exposure to diesel exhaust and smoking in the case-control study. If smoking, or diesel exhaust exposure as determined by job category, or both were associated with causes of death used for controls, results could be biased.

RECOMMENDATIONS

The Panel's recommendations reflect its general understanding, as expressed in its framework for evaluating studies, of what constitute adequate data for QRA. They also reflect the preceding evaluation of the studies of railroad workers and teamsters. The Panel is aware that research currently in progress will respond to some of these research needs; however, results are not yet available, and it is not yet clear whether all of the proposed needs will be met.

Completed Studies

1. The Panel recommends against using the current railroad worker data as the basis for QRA in ambient settings.
2. Further scrutiny of the teamster data, including estimation of uncertainty in both the exposure estimates and selection of controls, is recommended in order to improve the use of these data in QRA. Strengths of the teamster study include the relevance of exposure levels to the general population and the use of an exposure marker for diesel engine emissions that was an improvement over RSP. The teamster study exposure-response analysis is relatively new, and its further review and analysis by both the original investigators and others should be accelerated. Alternative retrospective exposure models need to be developed that use the alternative assumptions described above and in more detail in the body of the text.

Needs for New Techniques and Data

3. Better measures of exposure to constituents of diesel emissions, with careful attention to selection of the sample studied, are needed. Of particular importance are the selection and validation of a chemical marker of exposure to the complex mix of diesel exhaust emissions. Exposure models may include data from personal monitors, area monitors placed where diesel exposure is likely to occur, and current and historical data regarding emission sources. In any such modeling effort, the effects of environmental tobacco smoke should be removed as completely as possible.
4. Reliable estimates of past emissions and of factors affecting historical exposures in

a range of settings are needed to improve the characterization of uncertainties, both quantitative and qualitative, in historical models of exposures.

5. Although biomarker technology was not available when the studies reviewed were conducted, appropriate, validated, and specific biomarkers of diesel exposures, health outcomes, and susceptibility are needed.

Design Needs for New Studies of Exposure-Response Analyses

6. Exposures should be adequately and accurately characterized with respect to magnitude, frequency, and duration, rather than solely by duration of employment. Errors and uncertainties in exposure measurements should be quantified where possible; these should be fully reported to users, and taken into account in both power calculations and exposure-response analyses.
7. Cigarette smoking is a potent risk factor for lung cancer, and it must be controlled for in any study of risk factors for this disease. Smoking histories obtained for a cohort study subset that uses a case-control or case-cohort design will strengthen the interpretation of results.
8. The exposures considered should be close to levels of regulatory concern, including a range of exposures to provide a base for understanding the relation between exposure and health effects.

Needs for New Studies

A prospective epidemiologic study of the development of lung cancer in exposed and unexposed individuals could have many strengths. Information on confounders and exposures could be more complete than for a retrospective study, and many of the biases and uncertainties discussed in this report could be eliminated or reduced. These advantages, however, need to be weighed against the disadvantages, which include high costs and a long period of follow-up. Other study designs that include retrospective components are possible for a new epidemiologic study of lung cancer, but they are likely to include uncertainties and sources of bias that investigators will need to explore completely and acknowledge in their reporting.

9. The Panel recommends that a new, large, epidemiologic study of diesel exhaust emissions and lung cancer be considered after (1) currently ongoing or existing studies, including HEI's feasibility studies (to be completed in the spring of 2000), are evaluated, and (2) attempts to retrofit improved exposure assessments to existing epidemiologic studies are evaluated, including whether they can provide sufficiently accurate, complete, and relevant exposure data to support QRA.
10. Studies of lung cancer risk in general populations exposed to ambient diesel exhaust particulate matter will be difficult to conduct; however, such studies could usefully investigate other, noncancer health effects that occur in a shorter time after exposure.

SESSION V

What Will Epidemiology Studies Now Underway Tell Us About Exposure-Response?

Charles Poole, Co-Chair

Gerald van Belle, Co-Chair

Robert Säverin

Dirk Dahmann

Jorgen Olsen

Herman Autrup

Diane Mundt

Sessions V and VI were devoted to presentations of new epidemiologic studies of workers exposed to diesel emissions. The goal was to evaluate how these studies might improve risk assessments and their suitability for quantitative risk assessment. Session V included epidemiologic studies currently underway, one with German potash miners and one with Danish bus drivers. The session ended with poster presentations by the investigators of the six feasibility studies that HEI funded in response to RFA 98-3.

German Potash Miners: Cancer Mortality

Robert Säverin

Robert Säverin

Diesel Exhaust and Lung Cancer Mortality in German Potash Miners

During 1969 to 1970, six potash mines in the South Harz Mountains area of Germany changed technology to the use of mobile diesel powered vehicles. From then until the mines closed in 1991, the underground workforce was exposed to diesel exhaust.

The cohort was defined based on medical company records. The miners were required to undergo medical examination every other year. The works' doctors kept medical files recording workplaces and smoking habit. A team of medical personnel familiar with the mining technology went through the records to recruit a cohort comprising male miners who had worked underground for at least one year after 1969. This criterion was met by 5,981 men with 55 % of them exposed as early as 1970. The individual chronological series of workplaces occupied since 1970 of the recruited person was reconstructed from the record. Additionally, information on smoking habit, pre-mining occupations, and personal data like name, date of birth, and address was extracted. Diesel exhaust exposure was not mentioned explicitly in the records.

Each subject was classified as an active smoker, a former smoker, or a non-smoker according to the medical record. Because smoking was asked for routinely during medical examinations only after 1982, no smoking information could be obtained for 14.6 % of the subjects.

The follow-up started in 1970, and mortality was ascertained up to 1994. Persons lost to follow-up (1.9 %) and those with implausible or incomplete workplace history (5.5 %) were excluded leaving 5,536 cohort subjects to be followed. Among them, a subcohort of 3,258 miners worked underground at least ten years, held one single job during at least 80 % of their underground time, and held not more than three underground jobs in total.

The potash mines consisted of a widespread net of tunnel-like roadways connecting several production sections, the workshop section, and the shafts with each other. The salt was produced with the room-and-pillar method of mining allowing spacious rooms for production and transportation. The main sources of diesel exhaust were huge front-end loaders with 200 to 650 HP operating in the production section. They were used to scoop up and carry the debris after blasting, frequently turning and passing through their own emission cloud. Drill carriages were a second minor exhaust source. They were diesel propelled but drilling was done purely electrically. In the production section, the main occupations were loaders, drill operators, and blasters. They experienced the highest exposure to diesel exhaust, and their jobs were designated the *production* category of exposure.

By natural and forced ventilation, the emissions were diluted and distributed exposing several other workplaces to a lower but still considerable level. The *maintenance* category covers workplaces close to the *production* section where repairmen and electricians secured the infrastructure of the ever expanding mine. The foremen performed surveillance duties and moved throughout the mine. They too were considered to be exposed in the *maintenance* category of exposure.

The front-end loaders were maintained and repaired in a remote workshop section where emissions only occurred from engine checks and in the course of coming and going. Jobs in the maintenance section were designated as the *workshop* category and had the lowest exposure levels.

In 1992, measurements of the concentration of total carbon, i.e., elemental and organic carbon in total, in the airborne fine dust fraction were performed. With personal dust sampling, and area dust sampling where suitable, a set of 255 concentration values covering all workplaces was obtained. The measured values were averaged for the defined exposure categories *production, maintenance, workshop* providing three representative average concentrations. The mining technology and the type of machinery used did not change substantially after 1970. Therefore, the concentrations measured in 1992 were chosen to represent exposure during follow-up.

The exposure measure chosen for correlation with lung cancer mortality was cumulative exposure, i.e., the workplace concentration multiplied by years of exposure time. Using the individual chronological series of workplaces occupied since 1970, the increase in cumulative exposure was calculated for each miner. Thus, for each single year of observation, the cumulative exposure of each subject was known.

When the mines ceased production in 1991, most of the miners were dismissed and abandoned underground work and exposure. The mean exposure time was 15 years, and in 1994, the mean time since first exposure was 19 years. The mean age in 1994 was 49 years.

The measured total-carbon concentrations ranged from 0.038 to 1.28 mg/m^3. The representative *production*-category value was 0.39 mg/m^3 total carbon in the airborne fine dust fraction, the *maintenance* value was 0.23 mg/m^3 and the *workshop* value was 0.12 mg/m^3.

Validation revealed that the mean difference of the cumulative exposure in 1994 calculated from the medical files to that calculated from the interview data was 0.29 ymg/m^3, i.e., the medical files overestimated the cumulative exposure by only 8.5 % on average. About 90 % of the differences were in the range from −2.39 to 1.33 ymg/m^3 . In summary, though considerable individual exposure misclassification occured due to inaccuracies in the medical files, the exposure assessment was not substantially biased.

The percentage of smokers increased with cumulative exposure by a mere 0.08 % per ymg/m^3 (95%-confidence interval: 0.02 – 0.12 % per ymg/m^3), i.e., smoking habit was equally distributed over cumulative exposure and so the cohort proved to be homogenious regarding smoking.

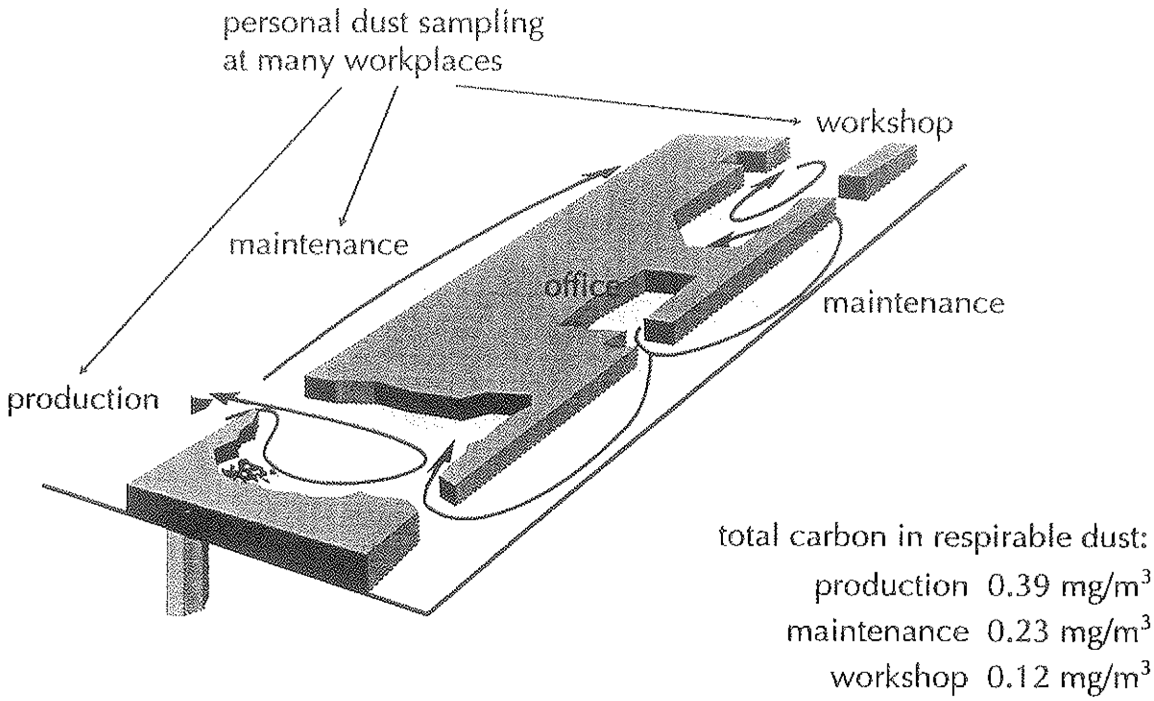
Whether or not a person's medical record contained smoking information was not associated with the cumulative exposure in 1994 of that person. The interviews revealed that former smokers were often classified as non-smokers in the medical records, mostly if they had stopped smoking many years earlier. The comparison of interview data and medical data showed that 28 % of apparent non-smokers were really misclassified former smokers. The mean exposure in 1994 of correctly classified non-smokers was 3.48 ymg/m^3, and that of misclassified non-smokers was 3.60 ymg/m^3. Hence, like the missing data, the misclassified subjects were equally distributed over the cumulative exposure scale.

A total of 424 deaths occurred between 1970 and 1994, including 133 from cancer and 38 cases from lung cancer. The cohort all-cause mortality was about half the mortality of the general male population. The SMR calculated from all deaths before 1983 was significantly smaller than that from 1983 onwards. The cohort's mortaliy from lung cancer was unexceptional.

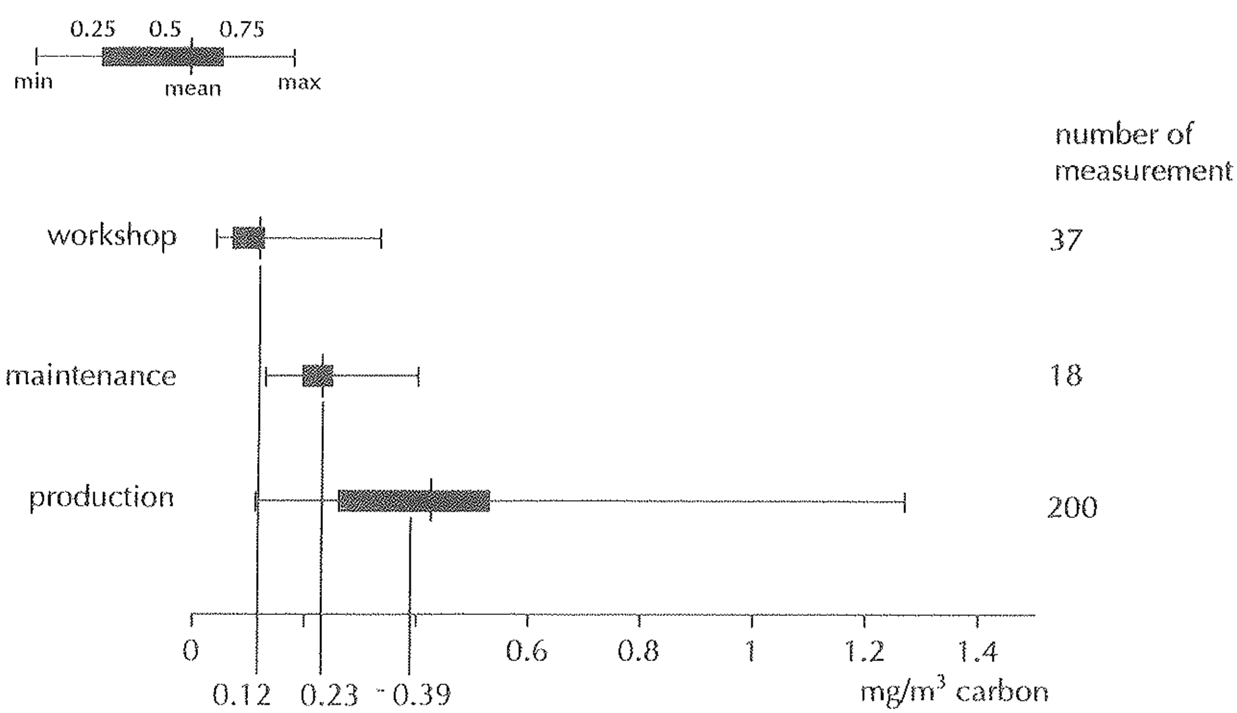
The relative risk for lung cancer in a highly exposed *production* group compared to the *workshop* group was 2.17 (95%-confidence interval 0.79 – 5.99) based on 11 and 6 deaths respectively. The all-cause relative risk was close to unity. The mean cumulative exposure was 4.38 ymg/m^3 in the *production* group and 2.12 ymg/m^3 in the *workshop* group. The *workshop* group had 64.3 % smokers, slightly less than the *production* group with 69.8 % smokers. In each group, the interviewed smokers smoked 12 cigarettes per day on average.

Poisson regression was used to calculate relative risks for twenty years of exposure in the highest *production* category of exposure, i.e., for a cumulative exposure of 4.9 ymg/m^3. In the overall cohort, the relative risk for lung cancer was 1.16 (0.38 - 3.5) whereas in the subcohort, the relative risk was 1.89 (0.46 - 11.9). The corresponding relative risks derived by Cox regression were 1.85 (0.56 - 6.1) for the overall cohort and 2.57 (0.51 - 13.0) for the subcohort.

The Workplaces in a Potash Mine

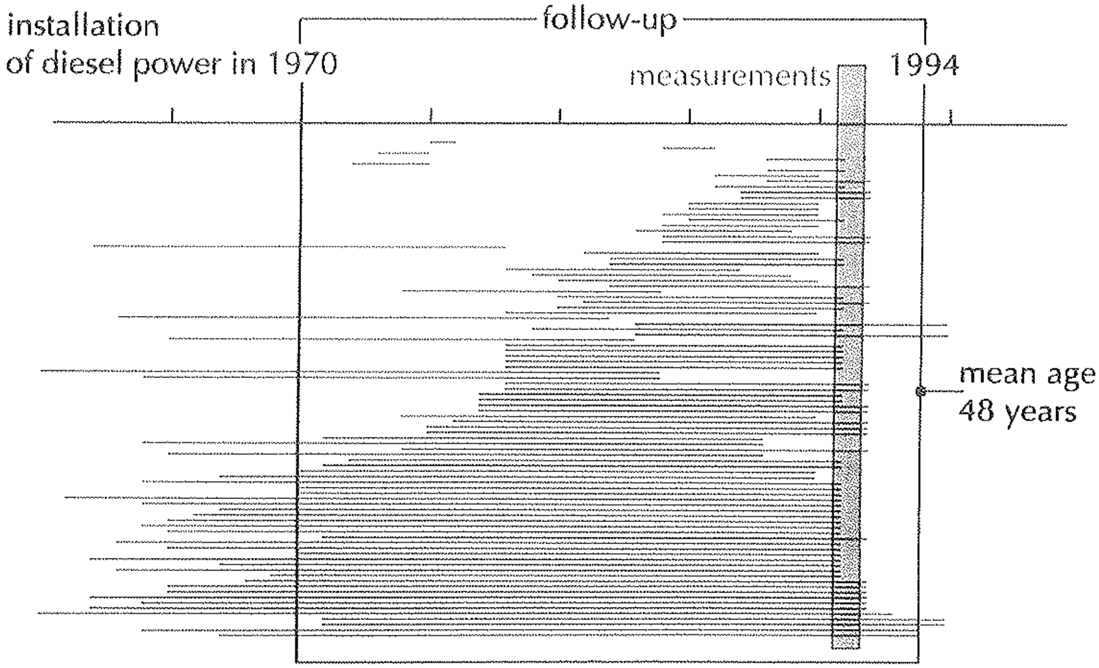


The Frequency Distribution of the Measurements

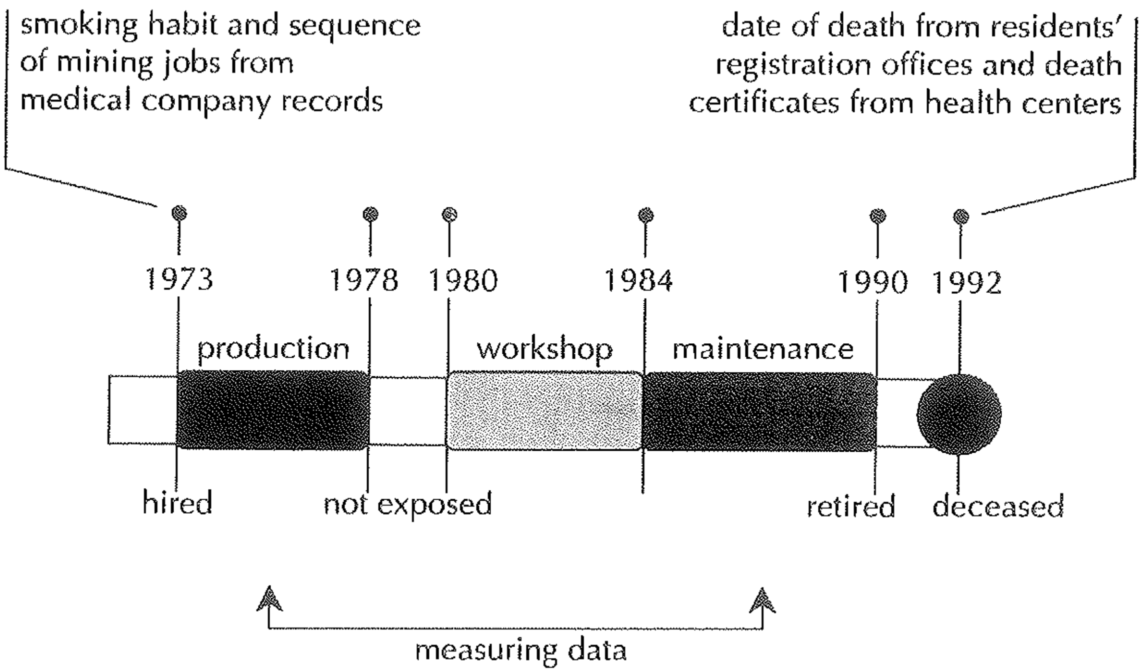


Individual Occupational Histories

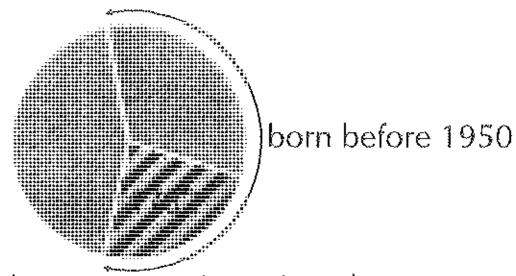
sample from the cohort of 5536 subjects



The Individual Occupational History



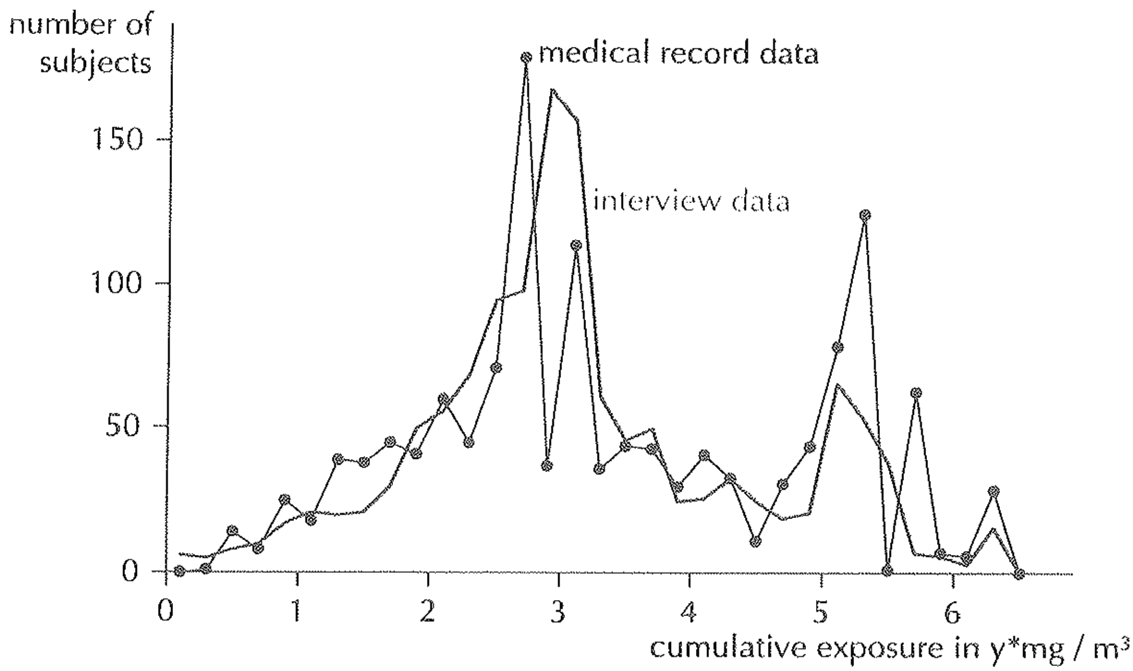
The Validation Study



medical record data	interview data
underground jobs, yearly (20 different job terms)	underground jobs, monthly (322 different job terms)
non-mining jobs (yes/no in five economy sectors)	non-mining jobs, monthly (company, exposures)
non-smoker or casual smoker / former smoker / smoker	number of cigarettes, yearly

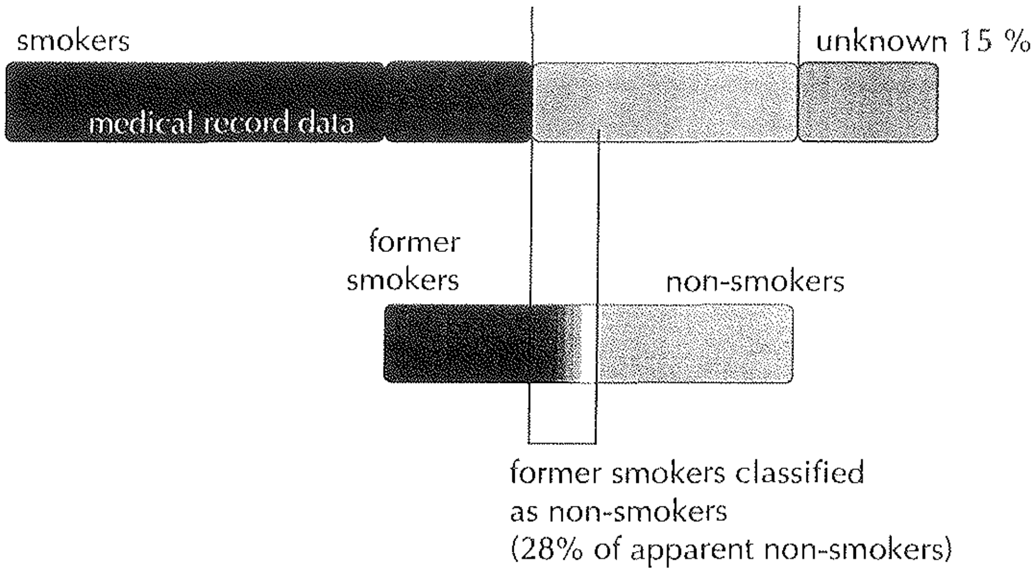
The Validation of the Exposure Data

frequency distribution of cumulative exposure



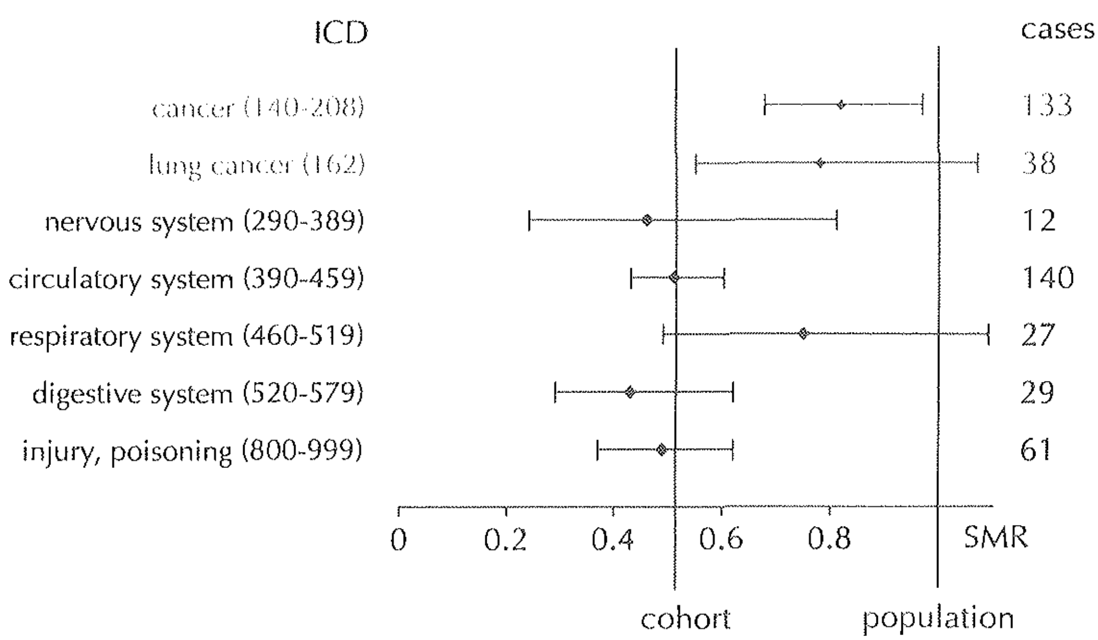
The Data on Smoking Habit

medical record data validated with interview data

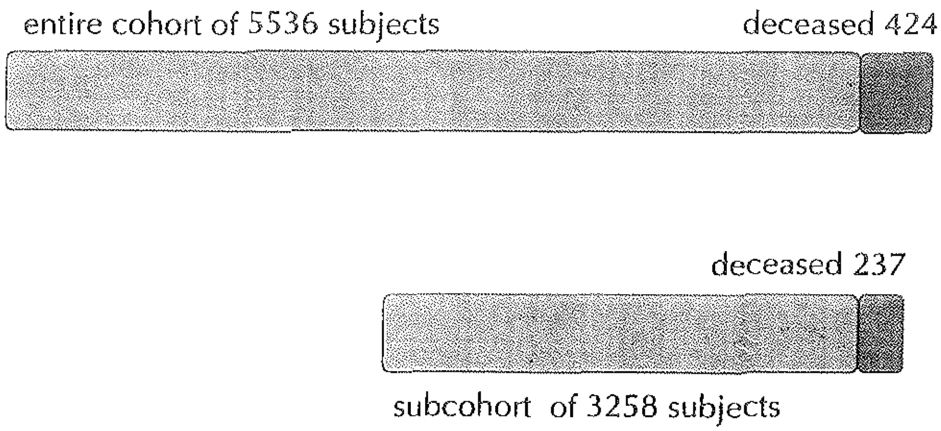


The Comparison With Population

standardized mortality ratio SMR



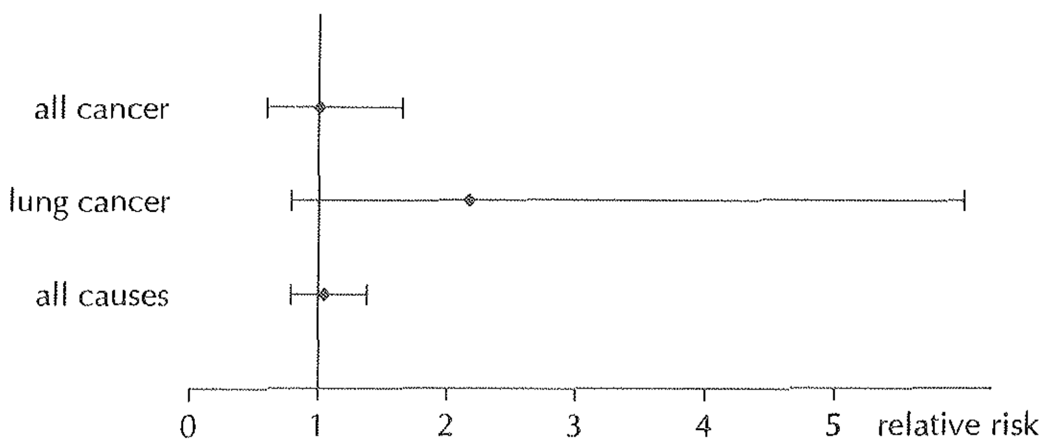
The Entire Cohort and the Subcohort



subcohort subjects worked underground at least ten years, and 80 per cent of their underground time in one single job

The Comparison Between Production and Workshop

Mantel-Haenszel relative risk



subjects worked underground at least ten years and 80 per cent of their underground time in the workshop or production exposure category

Poisson Regression Results

risk

$$\lambda(D,A) = e^{\gamma} A^{\delta} e^{\alpha D} \approx e^{\gamma} A^{\delta} + e^{\gamma} A^{\delta} \alpha D$$

relative risk

$$RR = \frac{\lambda(D,A)}{\lambda(D=0,A)} \approx 1 + \alpha D$$

relative risk of

lung cancer	1.89 (0.46 - 11.9)
all cancer	1.65 (0.82 - 4.40)
all causes	1.39 (0.92 - 2.3)

after twenty years
in the production exposure category (subcohort)

Cox Regression Results

hazard function

$$h(D,A) = \xi[D(A)]\, h_0(A)$$

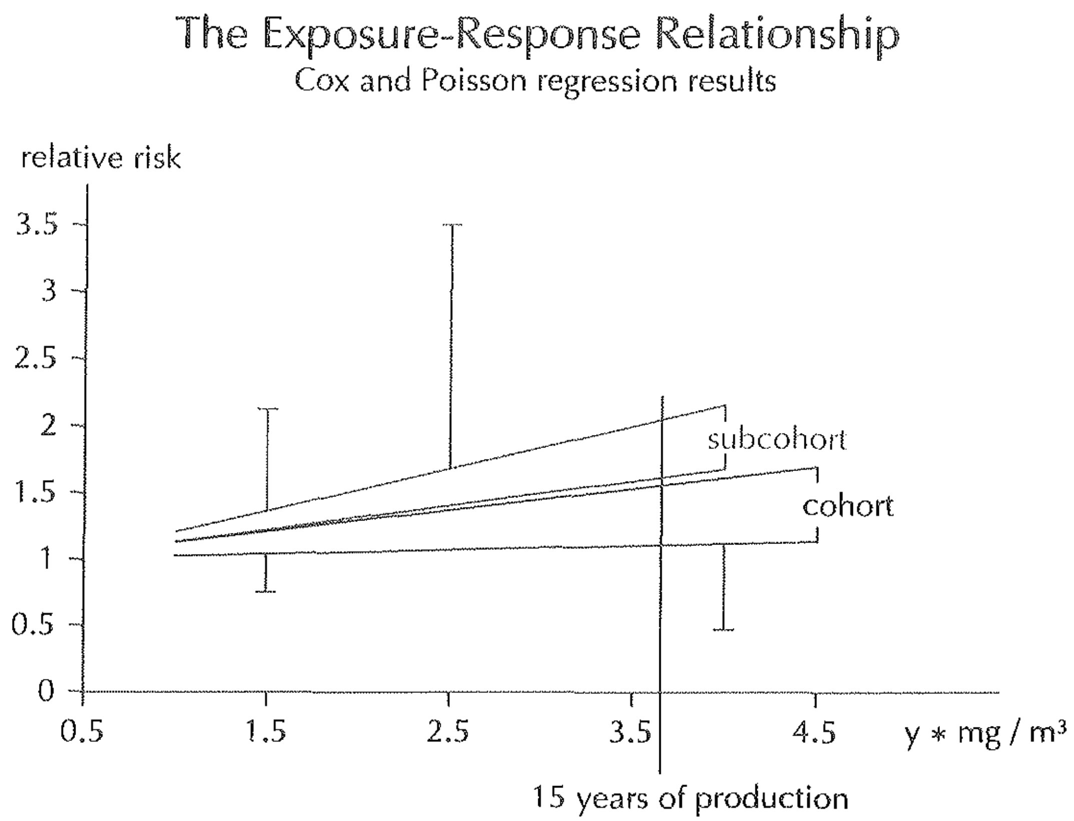
relative risk

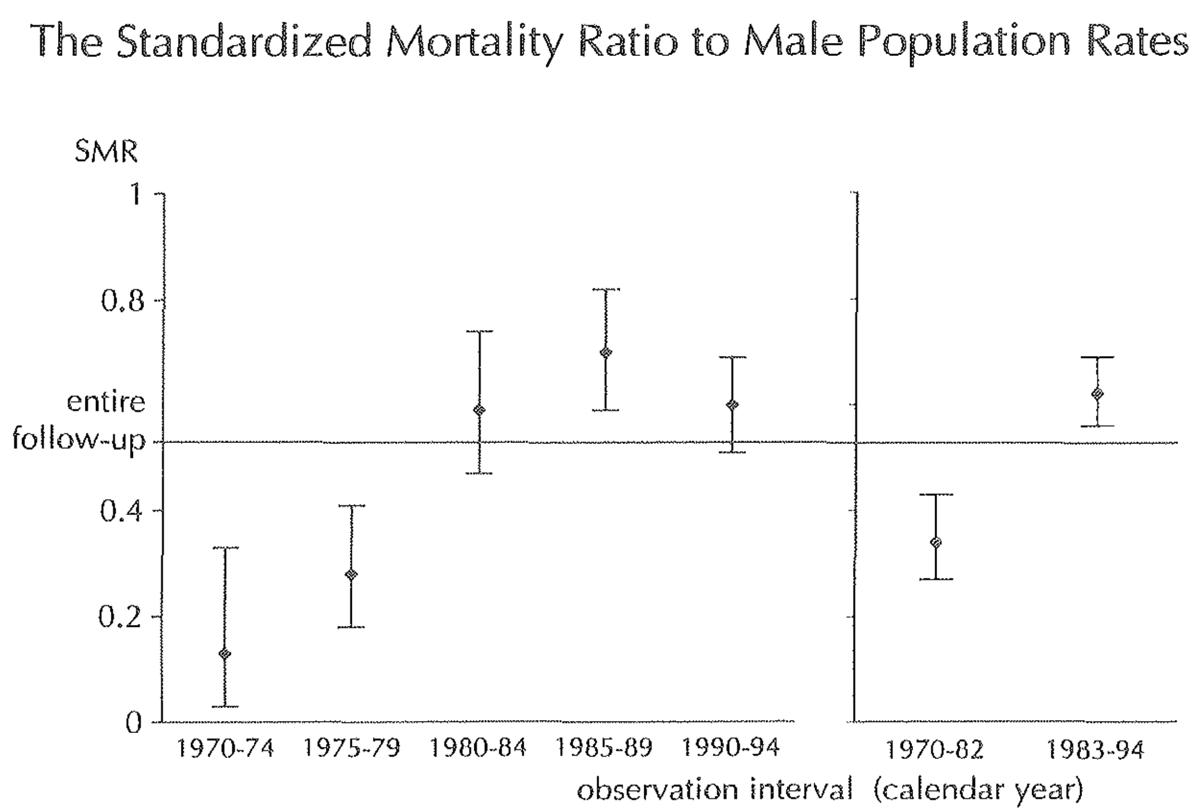
$$RR = \frac{h(D_1,A)}{h(D_0,A)} = \frac{\xi(D_1)}{\xi(D_0)}$$

relative risk of

lung cancer	2.81 (0.55 - 14.4)
all cancer	1.66 (0.71 - 3.90)
all causes	1.95 (1.20 - 3.2)

after twenty years
in the production exposure category (subcohort)


The Exposure-Response Relationship
Cox and Poisson regression results
relative risk
3.5
3
2.5
2
1.5
1
0.5
0
0.5
1.5
2.5
3.5
4.5
y * mg / m³
subcohort
cohort
15 years of production


The Standardized Mortality Ratio to Male Population Rates
SMR
1
0.8
entire
follow-up
0.4
0.2
0
1970-74
1975-79
1980-84
1985-89
1990-94
1970-82
1983-94
observation interval (calendar year)

German Potash Miners: Exposure

Dirk Dahmann

Diesel Exhaust and Lung Cancer Mortality in German Potash Mines
Exposure Assessment

Dirk Dahmann, Institut für Gefahrstoff-Forschung, Bochum

The exposure levels to diesel exhaust in six mines of the former German Democratic Republic had to be investigated. The study was peformed between 1991 and 1993. The situation in German potash and salt mining is characterized by a rapid transition process. The number of mines decreased, and those on the territory of the former GDR were thoroughly restructured or closed. Nevertheless, the mining process in the last decade and before is fairly comparable. Salt/potash ore is blasted during change of shifts, and the loose material is transported to the surface heavy diesel equipment and conveyor belts. Among others, job categories include loader drivers, drillers, road and ceiling maintenance personnel, expolisves workers, repair shop workers, people doing repair work on machines or electrical devices in the field and supervisors. As mentioned, a very critical point is the rapid replacement of obsolete machinery by modern types within the period of time of the study whose aim was to lower diesel exhaust and the parallel development of reduced ventilation as a cost factor. Summarized, the resulting situation can be characterized as fairly homogenous.

The measurement procedure was according to the official German method of diesel particulate matter in workplaces (ZH 1/120.44, Part 1). It is best described as coulometric determination of the total carbon present in the respirable dust collected on quartz fibre filters. This method has been published and validated in several interlaboratory tests.

We used personal sampling (lower detection limit 5 $\mu g/m^3$ in an 8h shift) and occasionally stationary sampling (ldl 0.2 $\mu g/m^3$ per 8 h). These methods have been shown to lead to equivalent results on the conditions of the measurements. All investigations were performed according to the requirements for compliance measurements of official German regulations. Personal sampling was preferred, all values are 8h shift averages. The data were obtained between the years 1991 and 1993 in a total of twelve potash/salt mines. However, as the mines of the study were partly no longer operating on exposure conditions similar to those of the real production years, most of the measurements had to be performed in other mines with comparable conditions. The relevant job categories could be covered by about 570 measurement data but in rather different densities. As a consequence and because the scattering of results within several of these categories was shown to be large, it was decided to group the outcome. Three groups of decreasing exposure levels were created, i.e. workers "in the field" (especially drivers of loading scoops), workers with changing (but lower) exposure levels and workers in the fresh air region near the intake shaft (basically repair shop personnel). These exposure levels will be presented by Dr. Säverin.

Two important questions need to be discussed: Are we convinced that the measurement campaign resulted in representative exposure data on the mines in question on the conditions of modern potash mining? The answer is undoubtedly yes. There is a twofold reason for this confidence. First, exposure conditions have always been governed by the factors "emission of the engines used" and "ventilation offered to that particular workplace". These factors have been under strict control by the minining companies and their supervising authorities. Second, we have shown by numerous measurements since 1993 that the situation is indeed very stable.

The second question is more difficult to answer: Can we be sure that the situation BEFORE 1990 was identical to that during our measurements? We are fairly sure. Even under the consideration that the modernisation process mentioned above and the exposure levels did NOT change dramatically within that period, we do not know the exact conditions of the former GDR. For this purpose, we would have to check production, ventilation and fuel consumption data which are no longer available. However, we asked experienced technicians for their educated guess, and they agreed to our proceedings. In our opinion, we used the best avialable model. It should be realized, however, that exposure data on previous years show a higher degree of uncertainty.

Diesel Exhaust and Lung Cancer Mortality in German Potash Miners

Exposure Assessment

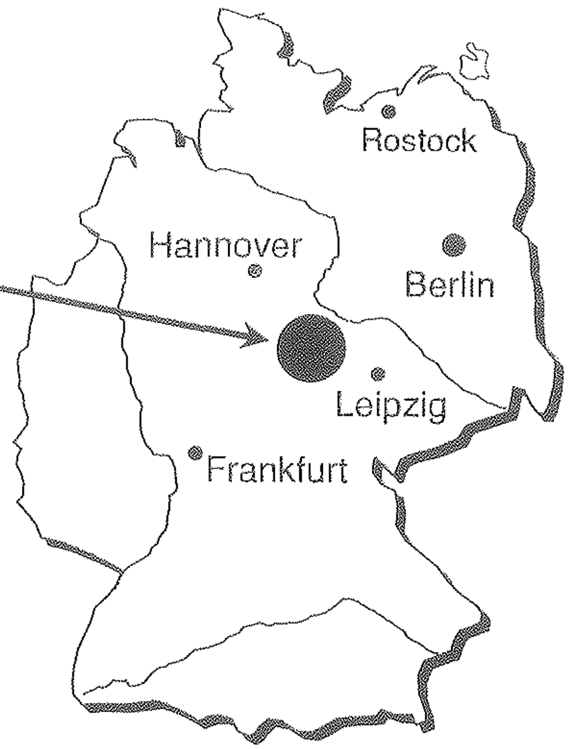
Dirk Dahmann, Bochum

Contents

- Mining Conditions
- Exposure Situation and Threshold Limits
- Sampling and Analytical Procedure
- Measurement Campaign
- Conclusions

Where are the mines?

south of the
Harz-mountains on
the territory of the
former GDR



Volkenroda
Sollstedt
Roßleben
Bleicherode
Sondershausen
Bischofferode

none of them is producing anymore

Mining Conditions 1999 and 1990

- In 1990 16 potash mines and 8 salt mines
  - a typical mine about 1500 miners
- Currently still 6 potash mines and 7 salt mines of varying size
  - the same mine under 500 miners

The Mining Process

- Blasting of salt/potash during change of shifts
- Loading and transport of loose salt/potash by large loaders to belts
- Simultaneously preparation of new blasting by drilling and related processes
- Simultaneously mine maintenance work
- Simultaneously maintenance of engines and electrical equipment

Job Categories (Selection)

- "In the field"
  - driving of loaders etc.
  - drilling
  - road maintenance
  - maintenance of ceiling etc.
  - filling of drilled holes with explosives etc.
- "Near fresh air shaft"
  - repair shop workers
- "Changing exposure"
  - supervisors
  - repair of "in field" facilities
  - transportation to and from field

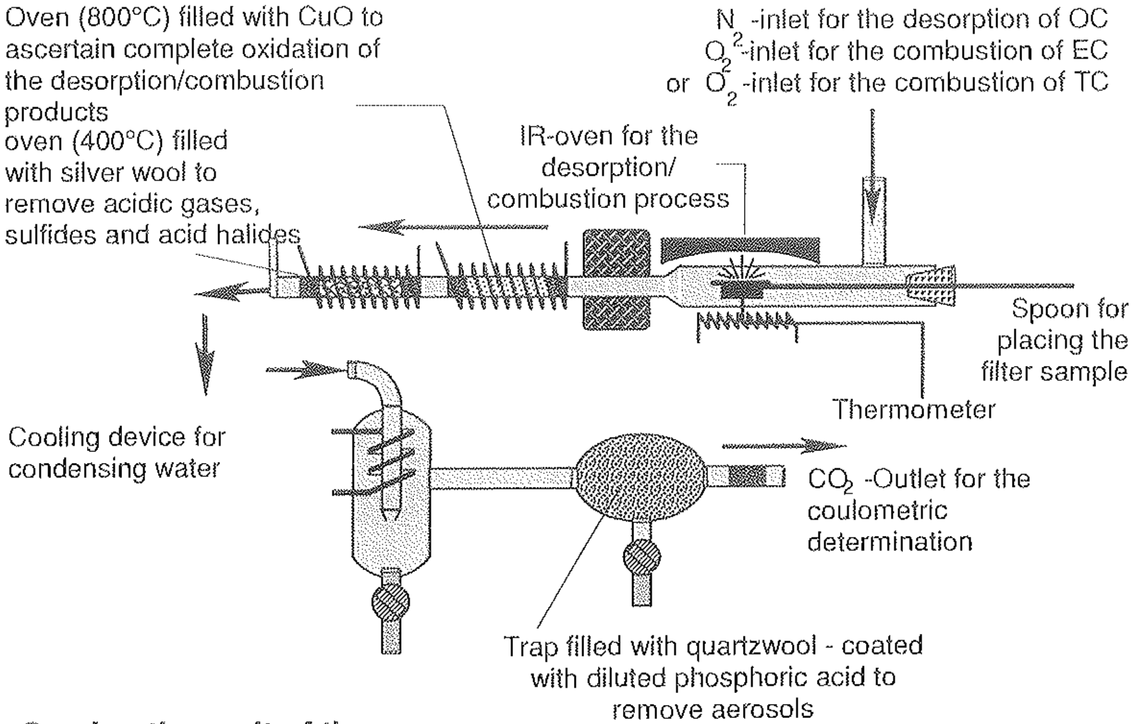
General Remarks to the Exposure Situation

- Rapid transition of mining equipment and conditions from "GDR" to "FRG" conditions in the period investigated
  - **however**: old equipment was replaced by modern types and ventilation was decreased at the same time
- Result: Fairly homogenous conditions within the mines under investigation

DPM - 1990-German practice

Component	Value (TRK) in ml/m³	Value (TRK) in mg/m³	Reference to TRGS 102	Remark
Diesel Engine Emissions [10, 11]			27)	"Investigated by coulometric analysis of the total carbon (TC) in the **respirable dust fraction**" (ZH 1/120.44)
"-non-coal-mining underground and construction work underground [12]"		0,6		
"-other workplaces"		0,2		

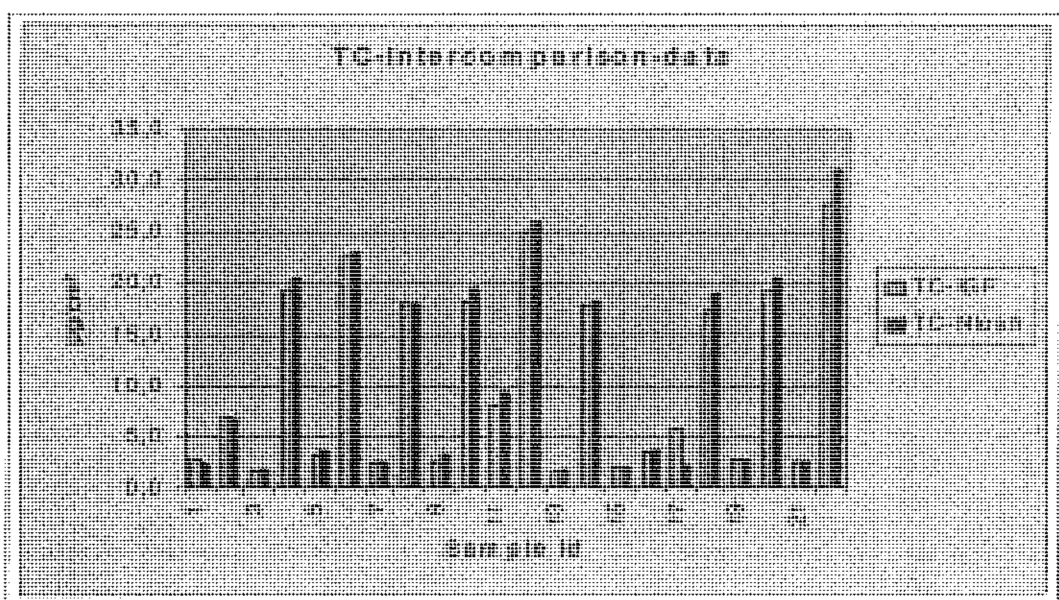
[10] A special threshold value for the triggering of preventive occupational examinations has been provided (0.1 mg/m³).
[11] Because of the interferences of the acknowledged analytical procedure in coal mining neither measurements can be performed nor the state of art of the technical equipment can be investigated in this branch. A threshold limit therefore cannot be fixed.
[12] Checking this value in 1995 (has been done)



Combustion unit of the coulometric device

How Valid is the Method?

Results of a recent intercomparison with NIOSH 5040



Sampling Procedures

- Personal Sampling - Casella Cyclone
- Flowrate: 2 l/min,
- Comment: to be used only under controlled environmental conditions e.g. in salt- potash-mines without non-Diesel sources of OC, EC etc.

- **Static Samplers**:
- - MPG II (horizontal elutriator according to Johannesburg convention)
- Flowrate: 46,5 l/min

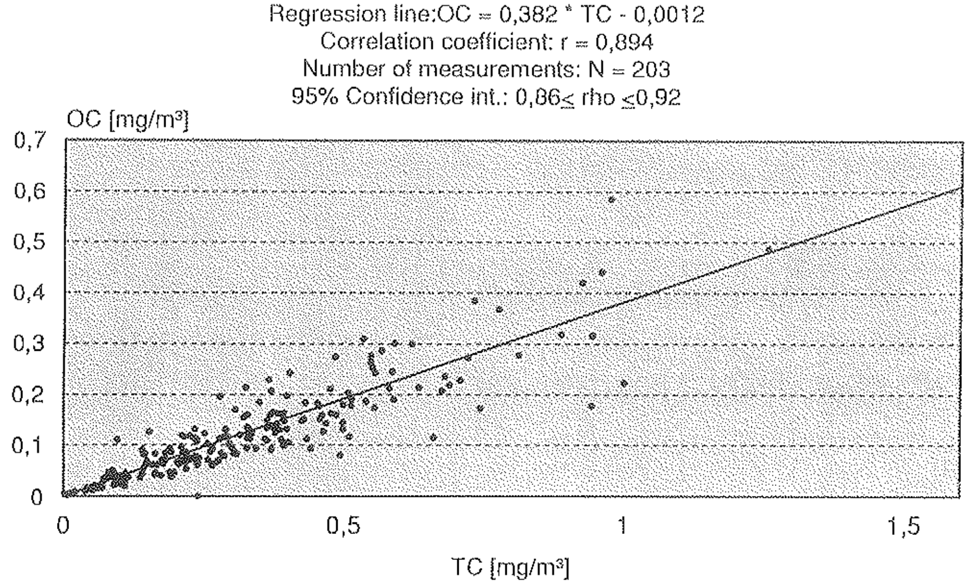
Existing Official Methods

- ZH 1/120.44 "Verfahren zur Bestimmung von Kohlenstoff im Feinstaub - anwendbar für partikelförmige Dieselmotor-Emissionen in Arbeitsbereichen -"
  - Part 1 (TC-Determination)
  - Part 2 (EC/OC-Determination)

  - **Comment: Though the study displays only the TC-values, EC and OC have been determined for most of the data as well!**

Organic Carbon as Part of Total Carbon

Correlation between OC and TC



Performance Characteristics

- Lower Detection Limit:
  - typically in the range of about 5 µg C per filter for TC, EC and OC
  - usually OC higher than EC or TC

- that means:
  - personal sampling: 5 µg/m³ for 8 hours sampling
  - static: 0.2 µg/m³ for 8 hours sampling

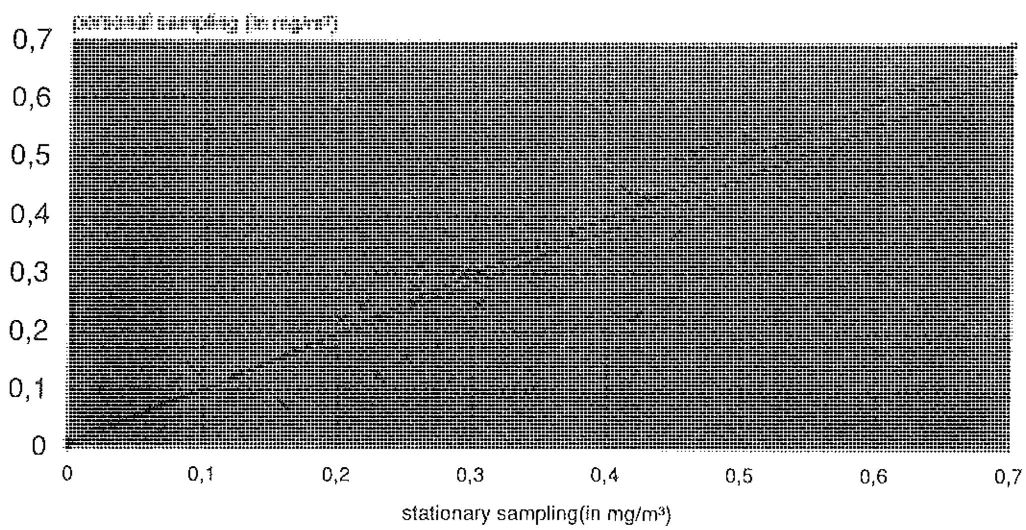
Measurement Campaign

All measurements were performed according to the official German regulations for compliance measurements (TRGS 402)

- personal sampling preferred
- static sampling done were necessary (but then "conservatively")
- all values are 8h-shift average
- data from 12 potash/salt mines (comparable ones but not necessarily the mines of the study)

Comparison of Stationary and Personal Sampling - an Example

parallel sampling in identical workplaces - comparison of 8h-shift values



Measurement Campaign II

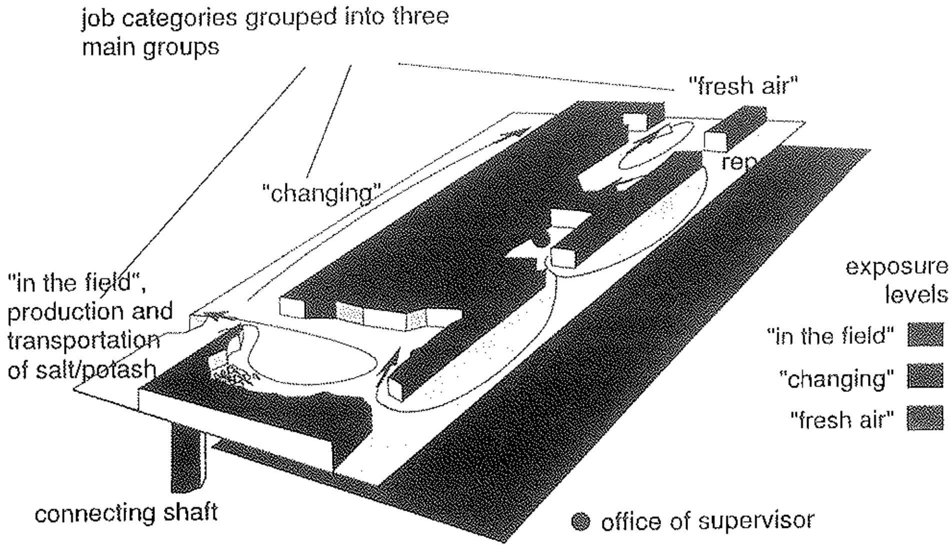
- a total of 570 exposure values were obtained
- measurements for this study were taken during 1991-1993
- all the relevant jobs were covered, however in very different density
- grouping of job categories to exposure groups was consequently necessary

Measurement Campaign III

From measurement results towards exposure levels:

- jobs were grouped according to three exposure levels:
  - "in the field"
  - "changing"
  - "fresh air"
- the available exposure data were used to get "typical" levels for these three "mine-departments"
- comparison of these levels for the mines were performed and the levels were then fixed accordingly (see Dr. Säverin)
- the resulting levels were assumed to have been constant from the start of exposure to now

Potash Mine (V E R Y schematic)



Questions:

Are the exposure levels representative for the situation in the mines investigated in the study?

- <u>Yes!</u>
  - There are only two main influencing factors:
    - ✓ the specific emission of the engines
    - ✓ the ventilation on-site
  - Ventilation requirements have always been fulfilled according to the power of the engines in use
  - All measurements since 1993 confirm that the situation is very stable

Questions

How sure can we be that the former situation was identical to the conditions during measurements?

- <u>Fairly!</u>
  - We monitored the process of change from 1990 to about 1995 and levels did not change dramatically.
  - However, we do not exactly know how conditions were in the GDR. This would need very intense investigation of production or ventilation data which simply is no longer available.
  - We did however ask technicians for their educated guess on this question.
  - **So we think this is currently the best available model**

Danish Bus Drivers: Cancer Incidence

Jørgen Olsen

Cancer incidence in Danish urban bus drivers and tramway employees

Authors:
Jørgen H. Olsen [1], Helle Soll- Johanning [2], Elsa Bach [2], Finn Tuchsen [2], and Herman Autrup [3]

[1] Institute of Cancer Epidemiology, Danish Cancer Society
[2] National Institute of Occupational Health
[3] University of Aarhus

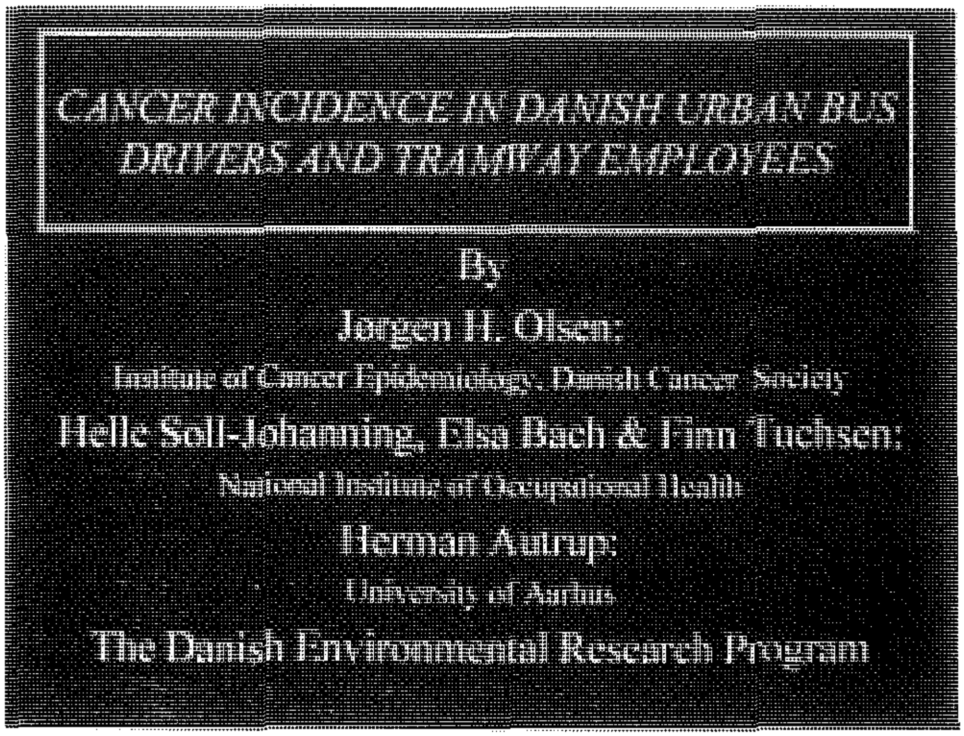
The objective of the present study was to investigate the risk of cancer associated with exposure to air pollution among bus drivers and tramway employees. A particular effort was done in the design and conduct of the study in order to adjust for any effect of tobacco smoking. A retrospective cohort of 18,120 bus drivers or tramway employees in Copenhagen, Denmark, in the period 1900-1994, was identified in the company files and followed-up in the national Danish Cancer Registry. Observed numbers of cancer among cohort members were compared with expected numbers based on the age- and sex-specific cancer iucidence rates for the national population (in some runs alternatively for the population of Copenhagen), and the standardised incidence ratios of cancer, taken as the observed-to-expected cancers, were calculated.

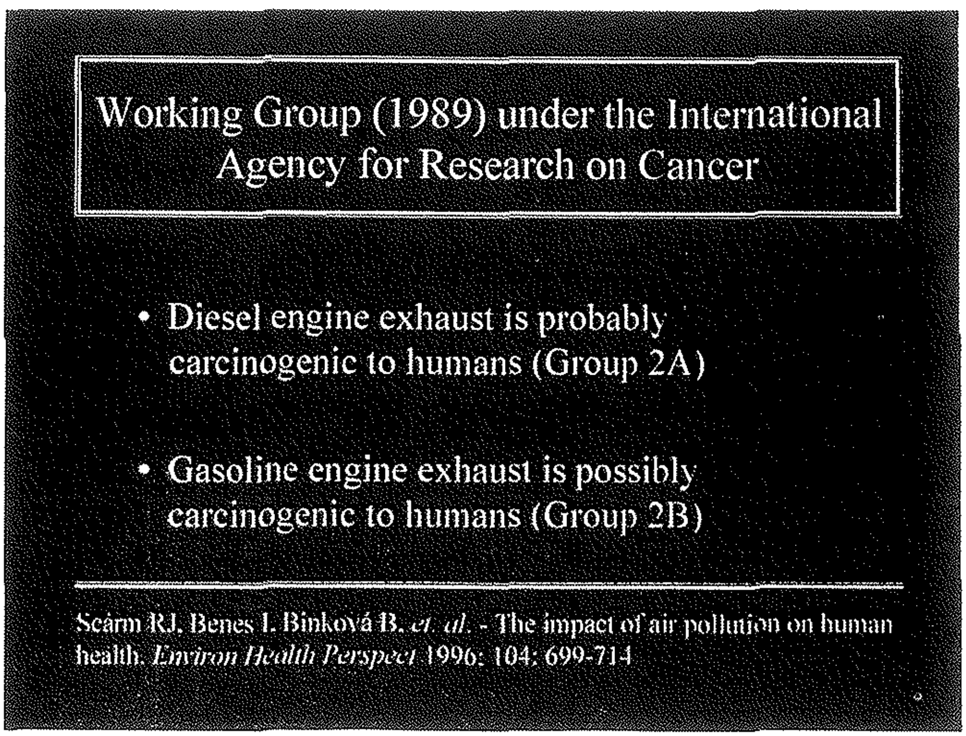
Cohort analysis. Findings showed that the bus drivers or tramway employees had an increased risk of all malignant neoplasms (standardised incidence ratio (SIR) 1.24, 95% confidence interval (95% CI) 1.19 to 1.30). The relative risk was significantly increased for both men and women (SIR 1.24, 95% CI 1.19 to 1.30 and 1.28, 1.06 to 1.53, respectively). People employed for < 3 months had no increased risk of cancer (1.04, 0.81 to 1.31). For men who were employed for > 3 months the risk of lung cancer (1.6, 1.5 to 1.8), laryngeal cancer (1.4, 1.0 to 1.9), kidney cancer (1.6, 1.3 to 2.0), bladder cancer (1.4, 1.2 to 1.6), skin cancer (1.1, 1.0 to 1.2), pharyngal cancer (1.9, 1.2 to 2.8), rectal cancer (1.2, 1.0 to 1.5) and liver cancer (1.6, 1.2 to 2.2) was significantly increased. However, by application of cancer reference rates for Copenhagen, only lung cancer remained significantly increased among male drivers with a SIR of 1.2 (1.1 to 1.3).

Measurements. Measurements of level of respirable dust, nitrogen dioxide and PAHs on one bus line were performed over a three-weeks period. Measured levels of these compounds, measured inside and outside the bus, were compared with levels calculated by use of the Operational Street Pollution Model (OSPM). It was concluded (i) that exposure levels of bus drivers to traffic related air pollution in Copenhagen is relatively low, (ii) that the observed nitrogen dioxide concentrations correlates very well with the calculated concentrations, and (iii) that there was no difference between the observed nitrogen dioxide and PAH concentrations inside and outside the bus.

Nested case-control study. Preliminary results from a case-control study of 240 cases of lung and bladder cancers, and 651 control subjects, nested in the study cohort showed that 98% of cases and 93% of bus driver controls were ever smokers. This finding strongly indicates that smoking is more prevalent among bus drivers than among the general population.

Overall conclusion. Based on the combined results of the study activities presented, the following conclusions were made: (i) the cohort study shows clearly increased risk for lung cancer, (ii) the measurement program indicates that levels of air pollution is relatively low in the study area, (iii) smoking habits among bus drivers seem to be substantially above normal, even among men in Copenhagen, and (iiii) the case-control study indicates, that at least part of the observed excess must be ascribed to tobacco smoking, however, any firm conclusion must await the results of the case-control analysis.


CANCER INCIDENCE IN DANISH URBAN BUS DRIVERS AND TRAMWAY EMPLOYEES
By
Jørgen H. Olsen:
Institute of Cancer Epidemiology, Danish Cancer Society
Helle Soll-Johanning, Elsa Bach & Finn Tuchsen:
National Institute of Occupational Health
Herman Autrup:
University of Aarhus
The Danish Environmental Research Program


Working Group (1989) under the International Agency for Research on Cancer
• Diesel engine exhaust is probably carcinogenic to humans (Group 2A)
• Gasoline engine exhaust is possibly carcinogenic to humans (Group 2B)
Scárm RJ, Benes I, Binková B, et. al. - The impact of air pollution on human health. Environ Health Perspect 1996; 104; 699-714

Danish study of urban bus drivers and tramway employees

- Study design
- Cancer findings
- Measurements

The Copenhagen Traffic Company (CTC) - District 1

- 80% of all bus companies in Copenhagen in 1974
- All employees (n=19,489), 1900 to 1994
- Exclusions:

– Duplicate registrations – Incorrect data – Missing information	n = 832 (4.3%)
– Loss to follow-up	n = 483 (2.5%)
– Death before 1 Jan. 1943	n = 54 (0.3%)
• Included i study	n = 18,120 (92.9%)

Descriptive characteristics

Characteristic	n	%
Sex:		
Male	16203	89
Female	1917	11
First employment (y):		
1900-19	526	3
1920-39	1325	7
1940-59	4310	24
1960-79	5909	33
1980-92	6050	33
Duration of employment (y):		
< 0.25	1060	6
0.25 - < 1	3358	19
1 - 9	8497	47
10 - 19	2044	11
≥ 20	3161	17
Total	18120	100

Follow-up for cancer incidence

- Follow-up from date of 1st employment (or 1 January 1943)
- Till date of death (or 31 December 1992)
- Linked with national Cancer Registry files
- Observed numbers of cancers compared with the expected numbers (based on national rates)
- Standardized incidence ratio (SIR) for cancer calculated as the ratio of observed-to-expected cancers
- 95% confidence intervals

Cancer at all sites combined
Total cohort (n = 18120)

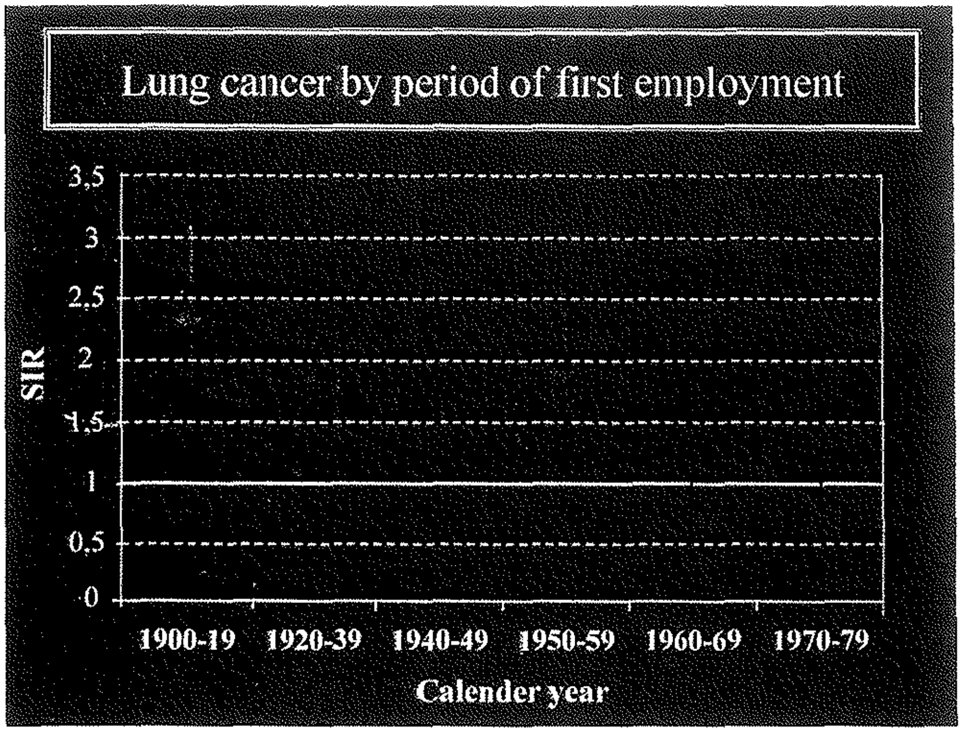
Stratification	Obs	Exp	SIR	(95% CI)
Total cohort	2149	1727[1]	1.24	(1.19-1.30)
Sex:				
Men	2031	1635	1.24	(1.19-1.30)
Women	118	92	1.28	(1.06-1.53)
Duration (y)				
< 0.25	72	69	1.04	(0.81-1.31)
≥ 0.25	2077	1658	1.25	(1.20-1.31)

[1] Reference: all Denmark

Cancers of selected sites
Men employed ≥ 3 months (n = 15249)

Site of cancer		All Denmark		Copenhagen	
	Obs	Exp	SIR	Exp	SIR
All malignant neoplasms	1966	1567	1.3*	1877	1.1*
Larynx	39	29	1.4*	40	1.0
Lung	473	292	1.6*	390	1.2*
Uninary bladder	177	130	1.4*	165	1.1
Esophagus	27	20	1.4	28	1.0
Liver	40	25	1.6*	37	1.1
Oral cavity	16	10	1.7*	15	1.1
Pharynx	22	12	1.9*	20	1.1
Pancreas	57	46	1.2	56	1.0

$*p < 0.05$



Cancer by time since first employment

	Time since first employment (y)					
	0-14		15-29		≥ 30	
Site of cancer	Obs	SIR	Obs	SIR	Obs	SIR
All malignant neoplasms	260	1.1	340	1.2*	1366	1.3**
Larynx	9	2.2	12	1.9*	18	1.0
Lung	35	1.2	77	1.5*	361	1.7**
Uninary baldder	19	1.3	28	1.3	130	1.4*

* $p < 0.05$; ** $p < 0.001$

Lung cancer by duration of employment and time since first employment

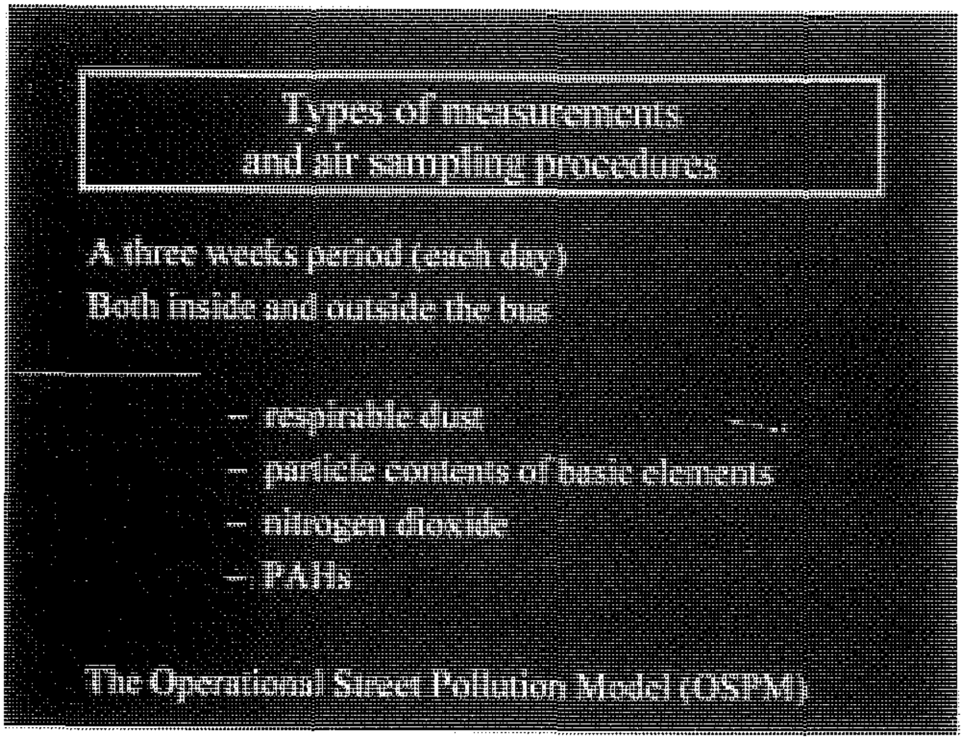
Duration of employment (y)	Time since first employment (y) 0-14 Obs	0-14 SIR	15-29 Obs	15-29 SIR	≥ 30 Obs	≥ 30 SIR
< 0.25	2	1.2	1	0.6	9	1.1
0.25 - < 1.5	5	1.0	11	1.5	57	2.0*
1.5 - 4	9	1.3	11	1.5	48	1.7*
5 - 14	17	1.3	26	2.5	70	2.1*
≥ 15	3	0.7	28	1.1	188	1.5*

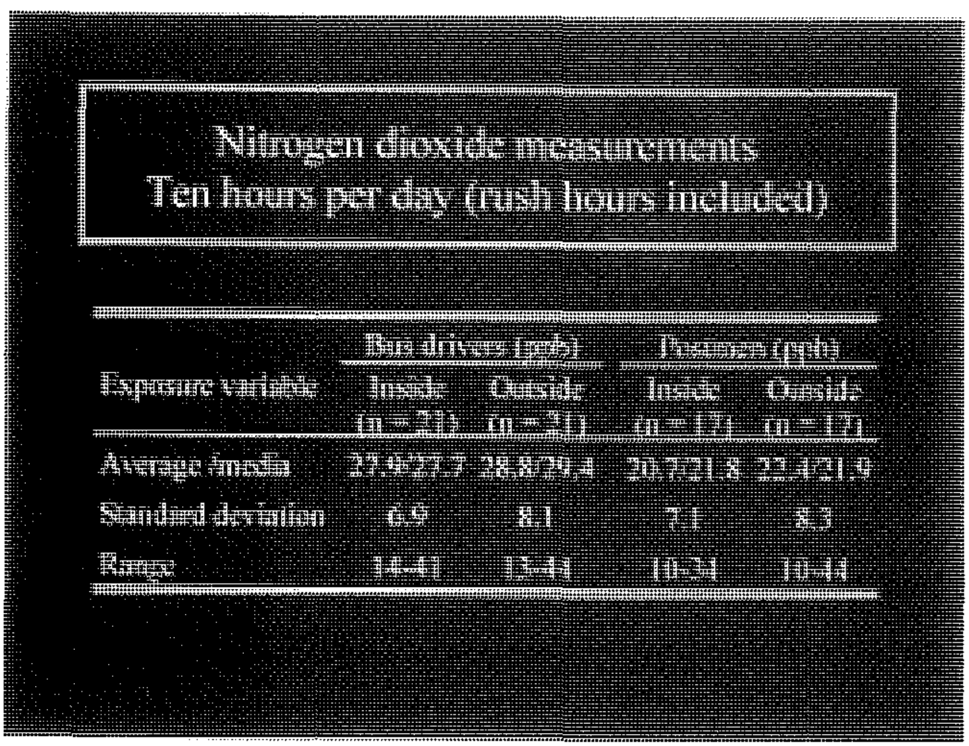
* $p < 0.05$

EXPOSSURE ASSESSMENT STUDY IN BUS DRIVERS AND TRAMWAY EMPLOYEES

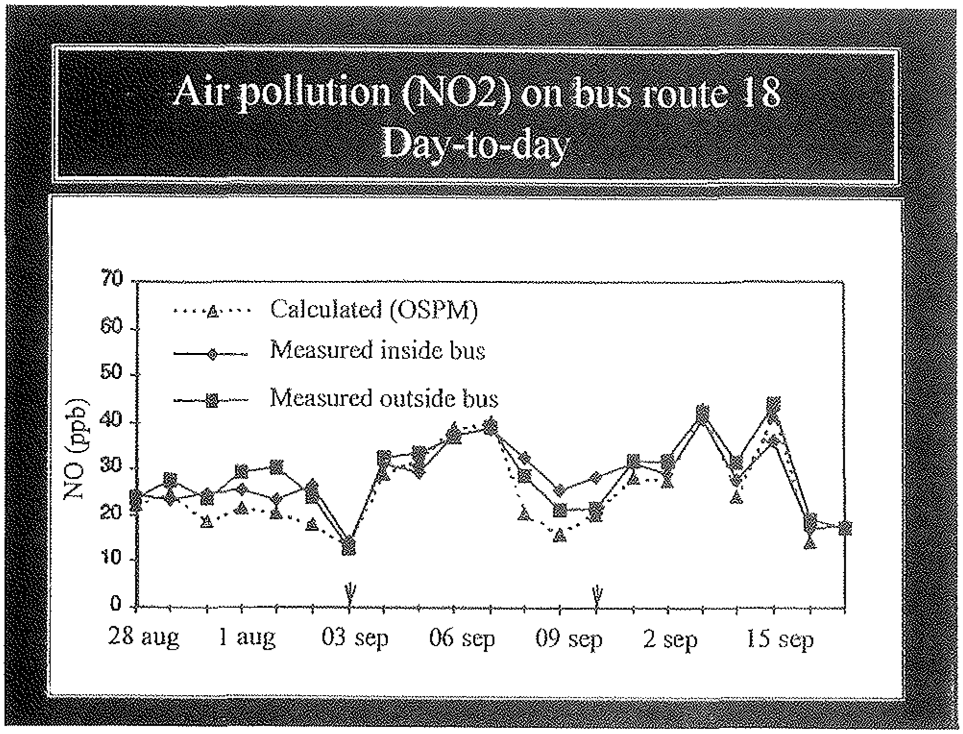
By

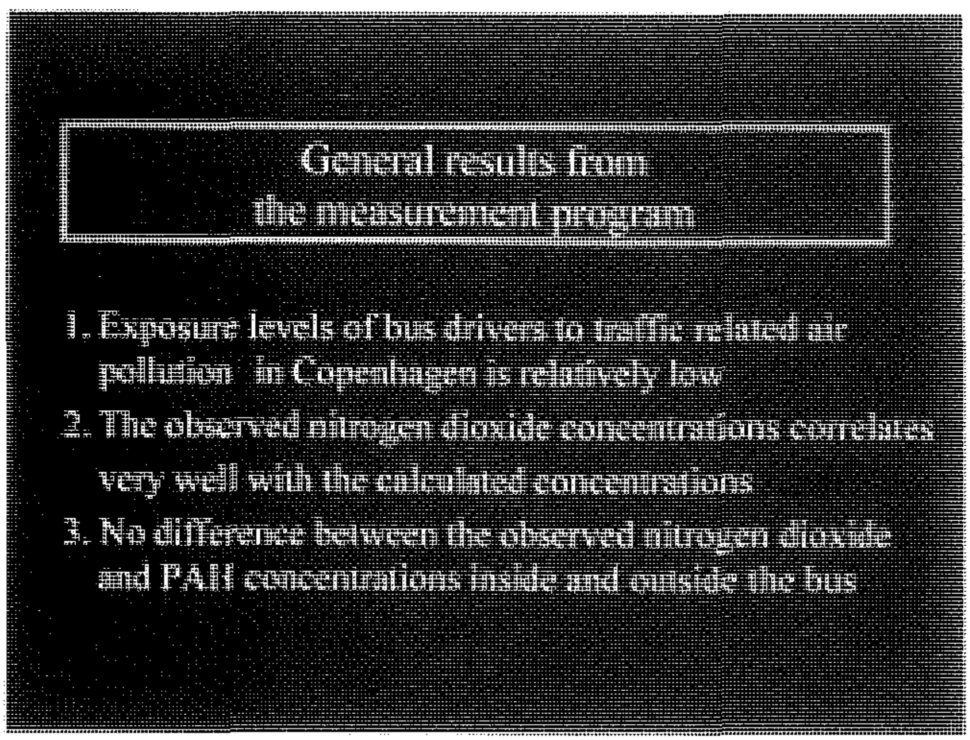
Peter Wilhardt, Niels Oluf Breum,
Åse Marie Hansen & Lisbeth E. Knudsen
National Institute of Occupational Health
Ole Hertel
National Environment Research Institute

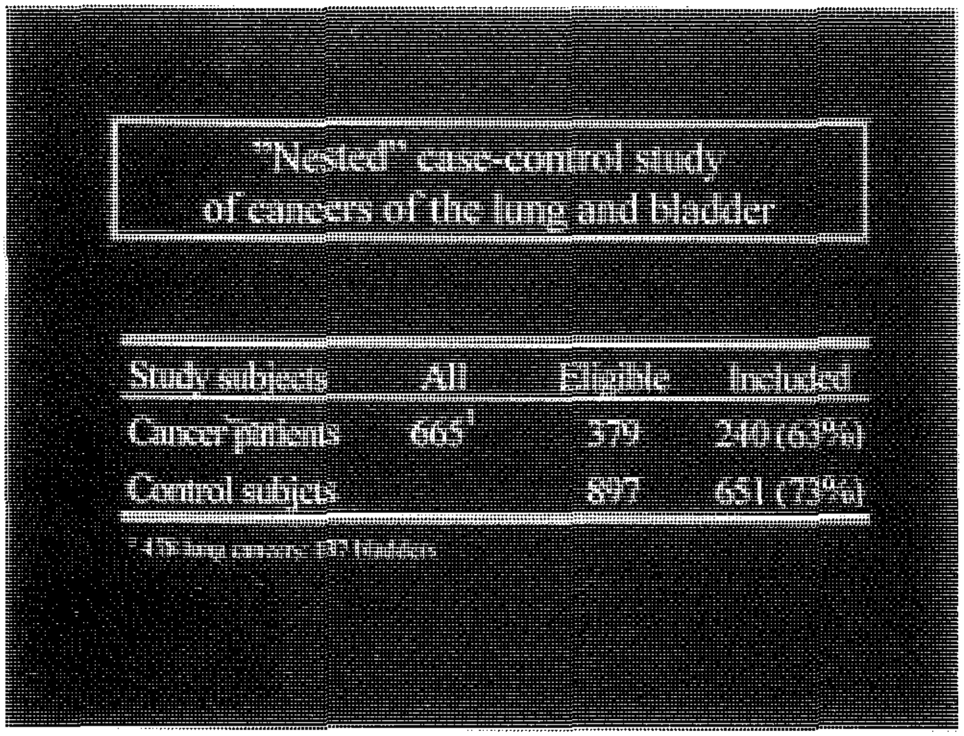




	Bus drivers (ppb)		Postmen (ppb)	
Exposure variable	Inside (n = 21)	Outside (n = 21)	Inside (n = 17)	Outside (n = 17)
Average /media	27.9/27.7	28.8/29.4	20.7/21.8	22.4/21.9
Standard deviation	6.9	8.1	7.1	8.3
Range	14-41	13-44	10-34	10-44

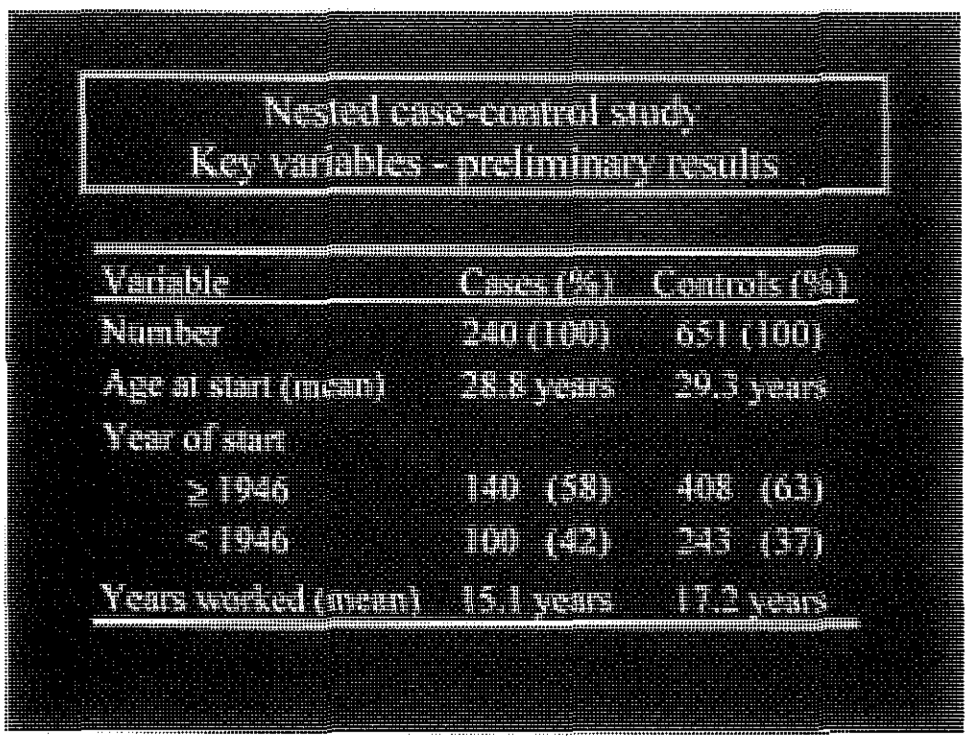

Air pollution (NO2) on bus route 18
Day-to-day
Calculated (OSPM)
Measured inside bus
Measured outside bus
NO (ppb)
70
60
50
40
30
20
10
0
28 aug
1 aug
03 sep
06 sep
09 sep
2 sep
15 sep


General results from
the measurement program
1. Exposure levels of bus drivers to traffic related air
pollution in Copenhagen is relatively low
2. The observed nitrogen dioxide concentrations correlates
very well with the calculated concentrations
3. No difference between the observed nitrogen dioxide
and PAH concentrations inside and outside the bus



"Nested" case-control study
of cancers of the lung and bladder

Study subjects	All	Eligible	Included
Cancer patients	665[1]	379	240 (63%)
Control subjects		897	651 (73%)



Nested case-control study
Key variables - preliminary results

Variable	Cases (%)	Controls (%)
Number	240 (100)	651 (100)
Age at start (mean)	28.8 years	29.3 years
Year of start		
≥ 1946	140 (58)	408 (63)
< 1946	100 (42)	243 (37)
Years worked (mean)	15.1 years	17.2 years

Nested case-control study
Smoking habits-preliminary results

Variable	Cases (%)	Controls (%)
Number	240 (100)	651 (100)
Ever smokers		
Yes	235 (98)	604 (93)
No	5 (2)	47 (7)
Pack-years		
Non-smokers	5 (2)	47 (7)
> 50	67 (28)	157 (24)
≤ 50	129 (54)	349 (54)
Missing	39 (16)	98 (15)

Cancer in Danish bus drivers and tramway employees

Conclusions

- Clearly increased risk for lung cancer
- Air pollution is relatively low
- Smoking habits above normal
- Causality question: must await proper analysis of the nested case-control study

Danish Bus Drivers: Biomarkers

Herman Autrup

DIESEL EXHAUST AND CANCER IN DANISH BUS DRIVERS – Biomarker results

Herman Autrup, Department of Environmental Medicine, University of Aarhus, Århus, Denmark

Ambient air contains a complex mixture of chemical compounds either in the gaseous form or bound to particulate matters. Air pollution in urban areas originates mostly from automobile exhaust with heavy diesel engines, buses and trucks contributing approximately 50-60% of the traffic emission in the urban areas. Some of the compounds found in diesel exhaust are carcinogenic in experimental animals. The level and type of the carcinogenic compounds depends on the engine construction, fuel type and driving conditions.

Different biomarkers have been used to assess exposure to genotoxic compounds in the general and occupational environment, ranging from the presence of mutagenicity in urine to induction of chromosomal damage in peripheral lymphocytes. The objective of this study was to evaluate the burden with genotoxic compounds in a Danish population assumed to be exposed extensively to ambient air pollution, mostly generated by incomplete combustion of fossil fuels.

A total of 209 non-smoking people was enrolled in the study, 107 busdrivers and 102 postal workers. Blood samples collected from these workers were divided in lymphocytes, erythocytes and serum. In addition urine samples were collected. The biomarkers used in this project were divided into markers of biological effective dose, biological effects and oxidative stress. The level of carcinogen-DNA adducts were determined in the lymphocytes using the P32-postlabelling method with butanol enrichment. This method will detect bulky adducts formed by polycyclic aromatic hydroearbons and aromatic amines.

The major adducts in the busdrivers were located along the X-Y axis, similar to the pattern seen in tobacco smokers. A significantly higher adduct level was observed in bus drivers working in the central Copenhagen, compared to the bus drivers working in the suburban areas and postal workers. The adduct level in this group however was several fold higher than in a rural population living on the island of Funen. The adduct level in the busdrivers in the central Copenhagen was also higher than in people living in areas with high level of air pollution, Bangkok and Athens. The adduct level was significantly higher in people who are deficient in detoxification of PAH, e.g. glutathione S-transferase M1 null. The adduct pattern in the Copenhagen bus drivers was also different from the pattern seen in the latter groups. No adducts comigrating with the 1-nitropyrene DNA adduct were observed in lymphocytes from Danish bus drivers. In contrast to the DNA-adduct level, bus drivers working outside the central Copenhagen had a higher level of benzo(a)pyrene albumin measured by ELISA assay. A negative correlation between the two markers were observed. Nitro-PAH are characteristic for diesel exhaust. After reduction of the nitrogroup, these compounds form adducts with hemoglobin. The group of busdrivers (30) had an increased level of 1-aminopyrene adduct compared to bus garage workers and miners, but did not differ for other amino-PAHs. Some of the consequences of DNA-adduct formation are chromosomal mutations and mutations in selected genes, e.g. the p53 cancer suppressor gene. An increased level of chromosomal aberrations was observed in the bus drivers in central Copenhagen compared to the postal workers. Exposure to traffic exhaust is associated with an increased risk of bladder and lung cancer. In a case-control study the mutational pattern I the p53 gene was compared in bus drivers with people not occupationally exposed to traffic exhaust. No significant difference in the mutation pattern could be observed.

While the PAH associated with particles generate DNA adducts following metabolic activation, the the nenuded particle will stimulate the formation of reactive oxygen species.Oxidative damage was measured to DNA, excretion of 8-oxo-7,8-dihydro-2'-deoxyguanosine in urine, lipids formation of malondialdehyde and proteins, 2-aminoapidic semialdehyde.
No significant difference in 8-oxo-dG and malondialdehyd level between bus drivers in city center and rural/suburban areas was found. In contrast a significantly higher level of 2-aminoapidic semialdehyde was observed in bus drivers in the city center.

Different biomarkers for genotoxic exposure and oxidative stress have been used to demonstrate that busdrivers in the central part of Copenhagen, with high level of exposure for diesel exhaust has an significantly increased level. These markers are not specific for compounds present in diesel exposure, but the marker level is associated with a higher level of ambient air pollution mainly generated by diesel buses and trucks in the street canyons of central Copenhagen.

Acknowledgement: The project has been supported by a grant to Centre for Biochemical and Occupational Epidemiology from the Danish Strategic Environmental Research Programme (1992-97).

Key publications:

Nielsen PS, de Pater N, Okkels H and Autrup H (1995) Environmental air pollution and DNA adducts in Copenhagen bus drivers – effect of GSTM1 and NAT2 genotypes on adduct levels. Carcinogenesis, 17: 1021-1027.

Autrup H, Daneshvar B, Dragsted LO et al (1999) Biomarkers for exposure to ambient air pollution – Comparison of carcinogen-DNA adduct levels with other exposure markers and markers for oxidative stress. Environ. Health Perspectives, 107: In press.

Knudsen LE, Norrpa H, Gamborg MO et al (1999) Chromosomal aberrations induced by urban air pollution in humans: influence of DNA repair and polymorphisms of glutathione S-transferase M1 and N-acetyltransferase 2. Cancer Epi Biomarker Prevention In press.

Loft S, Poulsen HE, Vistisen K and Knudsen LE (1999) Increased excretion of 8-oxo-7,8-dihydro-2'-deoxyguanosine, a biomarker of oxidative stress, in urban bus drivers. Mutation Res. In press.

DIESEL EXHAUST AND CANCER IN DANISH BUS DRIVERS
Biomarkers results

Herman Autrup
University of Aarhus
Aarhus Denmark

STUDY SUBJECTS
All non - smokers

Busdrivers:	107 (81 men and 26 women)
Mean age	45 years (range 27 - 60)
High exposure	47
Low exposure	60
Postal workers:	102 (70 men and 32 women)
Mean age	38 years (range 20 - 60)

PAH - ENVIRONMENT

	Naphthalene	Phenanthrene
Bus drivers	1365	48
Postal workers	408	56

BIOMARKERS OF EXPOSURE - 1

Biological effective dose:

- Bulky carcinogen - DNA adducts
- BPDE - albumin adducts
- Nitro - PAH hemoglobin adducts

Biological effects:

- Chromosomal aberrations
- p53 Mutation spectra

BIOMARKERS OF EXPOSURE - 2

Oxidative stress:

- Malondialdehyde (lipid)
- 8 - oxo - 2´ deoxyguanosine (DNA)
- 2 - amino - apidic semialdehyde (protein)

Others:

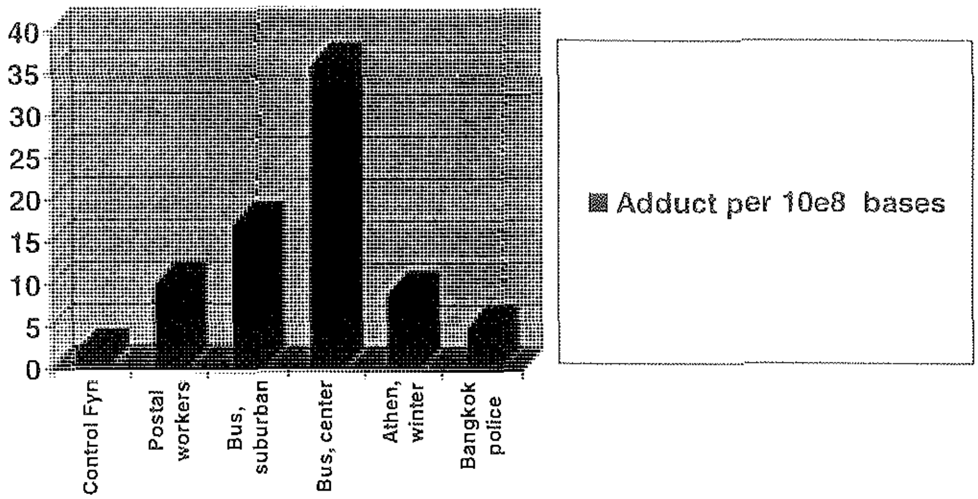
- 1 - Hydroxypyrene
- Mutagenicity - urine

CARCINOGEN DNA - ADDUCT LEVEL

	Number	Adduct level[a]	P values
City center	49	75.4 ± 122.0	
Rural / suburban	47	26.0 ± 40.5	0.012
Postal workers	97	30.9 ± 51.8	0.005

[a] Adducts per 10^8 nucleotides

BIOLOGICAL EFFECTIVE DOSE ADDUCTS



OXIDATION OF PLASMA PROTEINS

2 - aminoapidic semialdehyde

		Pmoles / mg protein	P value
City center	56	55.8 ± 24.1	
Rural / suburban	44	42.4 ± 13.8	0.0016
Postal workers	101	31.0 ± 5.3	0.0001

MARKERS OF OXIDATIVE STRESS

Malondialdehyd	#	Level	P
City center	55	0.87 ± 0.19	
Rural / suburban	45	0.96 ± 0.25	0.093
Postal workers	101	0.71 ± 0.16	0.0001
8 - oxodG			
City center	29	1.90 ± 1.08	
Rural / suburban	20	1.46 ± 0.89	0.05
Postal workers	82	2.25 ± 1.13	0.86

NITRO PAH Hb - ADDUCT LEVELS

Diesel exhaust

	Miners (N = 30)	Bus drivers (N = 30)	Bus garage workers (N = 29)
1 - aminopyrene	0.24[a]	0.45	0.13
2 - aminofluorene	0.14	0.09	0.04
3 - aminofluoranthene	0.14	0.03	0.03

[a] p mol/g Hb

Neuman et al Arch Toxical Suppl 20 179-187, 1997

CHROMOSOMAL ABERRATION

Total + gaps

	#	RR	95% CI
Postal workers	101	1.00	
Bus drivers	106	1.21	1.01 - 1.45

Knudsen et al Cancer Epi Biomakers Prevention In press

CORRELATION - BIOMARKERS

	Cases	r	p
DNA adduct vs PAH albumin	192	- 0.20	0.005
DNA adduct vs AAS - plasma	192	0.13	0.07
DNA adduct vs 8-oxodG	102	- 0.10	0.32

DNA ADDUCT LEVEL

Glutathione S - transferase

GSTM1	null	44 ± 58 (87)	
	plus	29 ± 53 (105)	P = 0.04
GSTT1	null	41 ± 57 (30)	
	plus	37 ± 57 (162)	P = 0.33
GSTP1	a,a	36 ± 48 (72)	
	a,b	35 ± 54 (99)	
	b,b	54 ± 86 (21)	P = 0.25 (trend)

CHROMOSOMAL ABERRATION

Effect of genotype

	NAT2 slow	GSTM1 null
Bus drivers, all	1.39 (1.09 - 1.78)	1.29 (1.00 - 1.66)
Bus drivers, city	1.43 (1.01 - 2.03)	1.41 (1.00 - 1.99)
Postal workers	1.68 (1.23 - 2.31)	1.08 (0.83 - 1.41)

p53 MUTATION SPECTRUM

Lung and bladder cancer

Case: Bus drivers

Referent: Matched by gender, age at diagnosis and time of diagnosis. Exclusion: Taxi and truck drivers

132 mutational analysis

p53 MUTATION ANALYSIS

Characterization of groups

	Cancercases		References	
	#	Age	#	Age
Squamous	27	64 ± 7	26	61 ± 6
Adeno	22	61 ± 9	15	59 ± 6
Bladder	27	62 ± 10	15	61 ± 8

p53 MUTATION

Identification of archival material

↓

DNA extraction of paraffin embedded tissue

↓

PCR, Exon 5 - 8

↓

SSCP analysis

↓

DNA sequencing

p53 MUTATION IN LUNG CANCER

	SSC		AC	
Mutation rate:				
Cases:	15/27 (55%)		7/22 (32%)	
Referent:	15/26 (57%)		6/15 (40%)	
Mutations - Exons:	5	6	7	8
Case:	6 (40%)	1 (7%)	4 (26%)	4 (26%)
Referent:	6 (40%)	2 (13%)	4 (26%)	3 (20%)

p53 MUTATION IN LUNG CANCER

Mutation pattern - SCC

	Case	Referent
Deletion	2/15	3/15
A → G	2/15	3/15
G → T	6/15	6/15
G → A	0	3/15
Others	5/15	1/15

p53 MUTATIONS IN BLADDER CANCER

	Case	Referent
Deletion	9/27	8/27
G → T	1/11	0/9
G → A	5/11	4/9
G → A transition:		
CpG site	1	4
non - CpG site	4	0

Fischer´s exact test P = 0.048

CONCLUSION

- Biomarker levels were significantly higher in bus drivers in central Copenhagen
- DNA adduct pattern indicates bulky adduct
- No correlations between different biomakers
- Large interindividual variations
- p53 mutation pattern similar in bus drivers and referent group

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HEI
DIESEL FEASIBILITY STUDIES

Diane J. Mundt, Ph.D.

THE DIESEL EPIDEMIOLOGY PROJECT OVERVIEW

HEI
- Diesel Epidemiology Expert Panel
- Feasibility Studies
- PM Research Program

WORLDWIDE RESEARCH
- Occupational Studies
- Environmental studies
- Emissions Characterization
- Biomarkers
- PM

RESEARCH STRATEGY WORKSHOP

FUTURE DIESEL RESEARCH

RFA 98-3

- Identify populations exposed at ambient or low occupational levels
  * exposure data available
- Develop exposure assessment strategy
  * validate measurement techniques
  * propose approaches for developing quantitative estimates of exposure

RFA 98-3

- Fifteen applications - six studies funded
- Approximately $1.4 million
  * Core sponsors: EPA, motor vehicle industry
  * Additional commitments: CARB, EMA

FEASIBILITY STUDIES

- Identification of 3 new cohorts
  - * Canadian railroad workers
    Murray Finkelstein, McMaster University, Canada
  - * U.S. truckers
    Eric Garshick, Channing Lab
  - * Transportation and construction workers in Central Europe
    Paolo Boffetta, IARC, France

FEASIBILITY STUDIES

- Exposure characterization/assessment
  - * Validate measurement techniques; characterize DPM with IH methods
    David Kittelson, University of Minnesota
  - * Ambient exposures to DPM in U.S. tunnel (compared with 25 years ago)
    William Pierson, Alan Gertler, DRI
  - * Personal monitor development and exposure characterization
    Barbara Zielinska, DRI

POSTER SESSION

HEI Diesel Feasibility Studies

Paolo Boffetta

Murray Finkelstein

Eric Garshick

David Kittelson

William Pierson

Barbara Zielinska