



United States
Environmental Protection
Agency

Health Assessment Document For Diesel Engine Exhaust

CONTENTS

LIST OF TABLES	viii
LIST OF FIGURES	xi
FOREWORD	xiv
PREFACE	xvi
AUTHORS, CONTRIBUTORS, AND REVIEWERS	xvii
ACKNOWLEDGMENTS	xxii
1. EXECUTIVE SUMMARY	1-1
1.1. INTRODUCTION	1-1
1.2. COMPOSITION OF DIESEL EXHAUST	1-1
1.3. DIESEL EXHAUST AS A COMPONENT OF AMBIENT PARTICULATE MATTER	1-2
1.4. ATMOSPHERIC TRANSFORMATION OF DIESEL EXHAUST	1-2
1.5. EXPOSURE TO DIESEL EXHAUST	1-3
1.6. HEALTH EFFECTS OF DIESEL EXHAUST	1-3
1.6.1. Acute (Short-Term Exposure) Effects	1-4
1.6.2. Chronic (Long-Term Exposure) Noncancer Respiratory Effects	1-4
1.6.3. Chronic (Long-Term Exposure) Carcinogenic Effects	1-4
1.7. SOURCES OF UNCERTAINTY	1-6
2. DIESEL EXHAUST EMISSIONS CHARACTERIZATION, ATMOSPHERIC TRANSFORMATION, AND EXPOSURES	2-1
2.1. INTRODUCTION	2-1
2.2. PRIMARY DIESEL EXHAUST EMISSIONS	2-3
2.2.1. History of Dieselization	2-3
2.2.2. Diesel Combustion and Formation of Primary Emissions	2-9
2.2.3. Diesel Emission Standards and Emission Trends Inventory	2-15
2.2.4. Historical Trends in Diesel Fuel Use and Impact of Fuel Properties on Emissions	2-25
2.2.5. Chronological Assessment of Emission Factors	2-29
2.2.6. Engine Technology Description and Chronology	2-43
2.2.7. Air Toxic Emissions	2-53
2.2.8. Physical and Chemical Composition of Diesel Exhaust Particles	2-59
2.3. ATMOSPHERIC TRANSFORMATION OF DIESEL EXHAUST	2-85
2.3.1. Gas-Phase Diesel Exhaust	2-85
2.3.2. Particle-Phase Diesel Exhaust	2-91
2.3.3. Diesel Exhaust Aging	2-94
2.4. AMBIENT DIESEL EXHAUST CONCENTRATIONS AND EXPOSURES	2-95
2.4.1. Diesel Exhaust Gases in the Ambient Atmosphere	2-95
2.4.2. Ambient Concentrations of DPM	2-96
2.4.3. Exposures to Diesel Exhaust	2-107
2.5. SUMMARY AND DISCUSSION	2-119
2.5.1. History of Diesel Engine Use, Standards, and Technology	2-120
2.5.2. Physical and Chemical Composition of Diesel Exhaust	2-121

CONTENTS (continued)

2.5.3. Atmospheric Transformation of Diesel Exhaust	2-125
2.5.4. Ambient Concentrations and Exposure to Diesel Exhaust	2-126
REFERENCES FOR CHAPTER 2	2-128
3. DOSIMETRY OF DIESEL PARTICULATE MATTER	3-1
3.1. INTRODUCTION	3-1
3.2. CHARACTERISTICS OF INHALED DIESEL PARTICULATE MATTER	3-2
3.3. REGIONAL DEPOSITION OF INHALED DIESEL PARTICULATE MATTER ...	3-2
3.3.1. Deposition Mechanisms	3-3
3.3.2. Particle Clearance and Translocation Mechanisms	3-9
3.3.3. Translocations of Particles to Extra-Alveolar Macrophage Compartment Sites	3-22
3.4. PARTICLE "OVERLOAD"	3-26
3.4.1. Introduction	3-26
3.4.2. Relevance to Humans	3-28
3.4.3. Potential Mechanisms for an AM Sequestration Compartment for Particles During Particle Overload	3-30
3.5. BIOAVAILABILITY OF ORGANIC CONSTITUENTS PRESENT ON DIESEL EXHAUST PARTICLES	3-31
3.5.1. In Vivo Studies	3-32
3.5.2. In Vitro Studies	3-34
3.5.3. Modeling Studies	3-36
3.5.4. Summary and Bioavailability	3-37
3.6. MODELING THE DEPOSITION AND CLEARANCE OF PARTICLES IN THE RESPIRATORY TRACT	3-38
3.6.1. Introduction	3-38
3.6.2. Dosimetry Models for DPM	3-38
3.7. SUMMARY AND DISCUSSION	3-54
REFERENCES FOR CHAPTER 3	3-56
4. MUTAGENICITY	4-1
4.1. GENE MUTATIONS	4-2
4.2. CHROMOSOME EFFECTS	4-5
4.3. OTHER GENOTOXIC EFFECTS	4-7
4.4. SUMMARY AND DISCUSSION	4-8
REFERENCES FOR CHAPTER 4	4-9
5. NONCANCER HEALTH EFFECTS OF DIESEL EXHAUST	5-1
5.1. HEALTH EFFECTS OF WHOLE DIESEL EXHAUST	5-2
5.1.1. Human Studies	5-2
5.1.2. Traffic Studies	5-23
5.1.3. Laboratory Animal Studies	5-24
5.2. MODE OF ACTION OF DIESEL EXHAUST-INDUCED NONCANCER EFFECTS	5-84
5.2.1. Comparison of Health Effects of Filtered and Unfiltered Diesel Exhaust ...	5-84

CONTENTS (continued)

5.2.2. Mode of Action for the Noncarcinogenic Effects of DPM	5-89
5.3. INTERACTIVE EFFECTS OF DIESEL EXHAUST	5-90
5.4. COMPARATIVE RESPONSIVENESS AMONG SPECIES TO THE HISTOPATHOLOGIC EFFECTS OF DIESEL EXHAUST	5-92
5.5. DOSE-RATE AND PARTICULATE CAUSATIVE ISSUES	5-93
5.6. SUMMARY AND DISCUSSION	5-97
5.6.1. Effects of Diesel Exhaust on Humans	5-97
5.6.2. Effects of Diesel Exhaust on Laboratory Animals	5-99
5.6.3. Comparison of Filtered and Unfiltered Diesel Exhaust	5-102
5.6.4. Interactive Effects of Diesel Exhaust	5-103
5.6.5. Conclusions	5-103
REFERENCES FOR CHAPTER 5	5-104
6. ESTIMATING HUMAN NONCANCER HEALTH RISKS OF DIESEL EXHAUST ...	6-1
6.1. INTRODUCTION	6-1
6.2. THE INHALATION REFERENCE CONCENTRATION APPROACH	6-3
6.3. CHRONIC REFERENCE CONCENTRATION FOR DIESEL EXHAUST	6-5
6.3.1. Principal Studies for Dose-Response Analysis: Chronic, Multiple-Dose Level Rat Studies	6-6
6.3.2. Derivation of Human Continuous Equivalent Concentrations, HECs	6-9
6.3.3. Dose-Response Analysis—Choice of an Effect Level	6-11
6.3.4. Uncertainty Factors (UF) for the RFC—A Composite Factor of 30	6-14
6.3.5. Derivation of the RfC for Diesel Exhaust	6-16
6.4. EPIDEMIOLOGICAL EVIDENCE AND NAAQS FOR FINE PM	6-17
6.4.1. Epidemiological Evidence for Fine PM	6-18
6.4.2. NAAQS for Fine PM	6-25
6.4.3. DPM as a Component of Fine PM	6-30
6.5. CHARACTERIZATION OF THE NONCANCER ASSESSMENT FOR DIESEL EXHAUST	6-30
6.6. SUMMARY	6-32
REFERENCES FOR CHAPTER 6	6-33
7. CARCINOGENICITY OF DIESEL EXHAUST	7-1
7.1. INTRODUCTION	7-1
7.1.1. Overview	7-1
7.1.2. Ambient PM-Lung Cancer Relationships	7-1
7.2. EPIDEMIOLOGIC STUDIES OF THE CARCINOGENICITY OF EXPOSURE TO DIESEL EXHAUST	7-3
7.2.1. Cohort Studies	7-6
7.2.2. Case-Control Studies of Lung Cancer	7-32
7.2.3. Summaries of Studies and Meta-Analyses of Lung Cancer	7-61
7.2.4. Summary and Discussion	7-66

CONTENTS (continued)

7.3. CARCINOGENICITY OF DIESEL EXHAUST IN LABORATORY ANIMALS	7-83
7.3.1. Inhalation Studies (Whole Diesel Exhaust)	7-84
7.3.2. Inhalation Studies (Filtered Diesel Exhaust)	7-108
7.3.3. Inhalation Studies (DE Plus Cocarcinogens)	7-109
7.3.4. Lung Implantation or Intratracheal Instillation Studies	7-111
7.3.5. Subcutaneous and Intraperitoneal Injection Studies	7-117
7.3.6. Dermal Studies	7-119
7.3.7. Summary and Conclusions of Laboratory Animal Carcinogenicity Studies ..	7-121
7.4. MODE OF ACTION OF DIESEL EXHAUST-INDUCED CARCINOGENESIS	7-128
7.4.1. Potential Role of Organic Exhaust Components in Lung Cancer Induction ..	7-129
7.4.2. Role of Inflammatory Cytokines and Proteolytic Enzymes in the Induction of Lung Cancer in Rats by Diesel Exhaust	7-132
7.4.3. Role of Reactive Oxygen Species in Lung Cancer Induction by Diesel Exhaust	7-133
7.4.4. Relationship of Physical Characteristics of Particles to Cancer Induction ..	7-136
7.4.5. Integrative Hypothesis for Diesel-Induced Lung Cancer	7-137
7.4.6. Summary	7-139
7.5. WEIGHT-OF-EVIDENCE EVALUATION FOR POTENTIAL HUMAN CARCINOGENICITY	7-140
7.5.1. Human Evidence	7-141
7.5.2. Animal Evidence	7-142
7.5.3. Other Key Data	7-143
7.5.4. Mode of Action	7-143
7.5.5. Characterization of Overall Weight of Evidence: EPA's 1986 Guidelines for Carcinogen Risk Assessment	7-144
7.5.6. Weight-of-Evidence Hazard Narrative: EPA's Proposed Guidelines for Carcinogen Risk Assessment (1996b, 1999)	7-144
7.6. EVALUATIONS BY OTHER ORGANIZATIONS	7-146
7.7. CONCLUSION	7-147
REFERENCES FOR CHAPTER 7	7-148
8. DOSE-RESPONSE ASSESSMENT: CARCINOGENIC EFFECTS	8-1
8.1. INTRODUCTION	8-1
8.2. MODE OF ACTION AND DOSE-RESPONSE APPROACH	8-2
8.3. USE OF EPIDEMIOLOGIC STUDIES FOR QUANTITATIVE RISK ASSESSMENT	8-4
8.3.1. Sources of Uncertainty	8-4
8.3.2. Evaluation of Key Epidemiologic Studies for Potential Use in Quantitative Risk Estimates	8-5
8.3.3. Conclusion	8-11
8.4. PERSPECTIVES ON CANCER RISK	8-11
8.5. SUMMARY AND DISCUSSION	8-16
REFERENCES FOR CHAPTER 8	8-17

CONTENTS (continued)

9. CHARACTERIZATION OF POTENTIAL HUMAN HEALTH EFFECTS OF DIESEL EXHAUST: HAZARD AND DOSE-RESPONSE ASSESSMENTS	9-1
9.1. INTRODUCTION	9-1
9.2. PHYSICAL AND CHEMICAL COMPOSITION OF DIESEL EXHAUST	9-2
9.2.1. Diesel Exhaust Components of Possible Health Concern	9-2
9.2.2. "Fresh" Versus "Aged" Diesel Exhaust	9-4
9.2.3. Changes of Diesel Exhaust Emissions and Composition Over Time	9-5
9.3. AMBIENT CONCENTRATIONS AND EXPOSURE TO DIESEL EXHAUST	9-6
9.4. HAZARD CHARACTERIZATION	9-8
9.4.1. Acute and Short-Term Exposures	9-8
9.4.2. Chronic Exposure	9-10
9.5. DOSE-RESPONSE ASSESSMENT	9-16
9.5.1. Evaluation of Risk for Noncancer Health Effects	9-17
9.5.2. Evaluation of Cancer Risks	9-20
9.6. SUMMARY AND CONCLUSIONS	9-24
REFERENCES FOR CHAPTER 9	9-27
APPENDIX A: CALCULATION OF HUMAN EQUIVALENT CONTINUOUS EXPOSURE CONCENTRATIONS (HECs)	A-1
APPENDIX B: BENCHMARK CONCENTRATION ANALYSIS OF DIESEL DATA	B-1
APPENDIX C: A SUMMARY REVIEW OF CANCER DOSE-RESPONSE ANALYSES ON DIESEL EXHAUST	C-1

1. EXECUTIVE SUMMARY

1.1. INTRODUCTION

This Health Assessment Document for Diesel Engine Exhaust (DE) represents EPA's first comprehensive review of the potential health effects from ambient exposure to exhaust from diesel engines. The assessment was developed to provide information about the potential for DE to pose environmental health hazards, information that would be useful in evaluating regulatory needs under provisions of the Clean Air Act. The assessment identifies and characterizes the potential human health hazards of DE (i.e., hazard assessment) and seeks to estimate the relationship between exposure and disease response for the key health effects (i.e., dose-response assessment). A full exposure assessment and risk characterization, the other two components of a complete risk assessment, are beyond the scope of this document.

The report has nine chapters and three appendices. Chapter 2 provides a characterization of diesel emissions, atmospheric transformation, and human exposures to provide a context for the hazard evaluation of DE. Chapters 3, 4, 5, and 7 provide a review of relevant information for the evaluation of potential health hazards of DE, including dosimetry (Chapter 3), mutagenicity (Chapter 4), noncancer effects (Chapter 5), and carcinogenic effects (Chapter 7). Chapters 6 and 8 contain dose-response analyses to provide insight about the significance of the key noncancer and cancer hazards. Chapter 9 summarizes and characterizes the overall nature of the health hazard potential in the environment and the overall confidence and/or uncertainties associated with the conclusions.

1.2. COMPOSITION OF DIESEL EXHAUST

DE is a complex mixture of hundreds of constituents in either a gas or particle form. Gaseous components of DE include carbon dioxide, oxygen, nitrogen, water vapor, carbon monoxide, nitrogen compounds, sulfur compounds, and numerous low-molecular-weight hydrocarbons. Among the gaseous hydrocarbon components of DE that are individually known to be of toxicologic relevance are the aldehydes (e.g., formaldehyde, acetaldehyde, acrolein), benzene, 1,3-butadiene, and polycyclic aromatic hydrocarbons (PAHs) and nitro-PAHs.

The particles present in DE (i.e., diesel particulate matter [DPM]) are composed of a center core of elemental carbon and adsorbed organic compounds, as well as small amounts of sulfate, nitrate, metals, and other trace elements. DPM consists of fine particles (fine particles have a diameter $<2.5 \mu\text{m}$), including a subgroup with a large number of ultrafine particles (ultrafine particles have a diameter $<0.1 \mu\text{m}$). Collectively, these particles have a large surface area which makes them an excellent medium for adsorbing organics. Also, their small size makes them highly respirable and able to reach the deep lung. A number of potentially

toxicologically relevant organic compounds are on the particles. The organics, in general, range from about 20% to 40 % of the particle weight, though higher and lower percentages are also reported. Many of the organic compounds present on the particle and in the gases are individually known to have mutagenic and carcinogenic properties. For example, PAHs, nitro-PAHs, and oxidized PAH derivatives are present on the diesel particles, with the PAHs and their derivatives comprising about 1% or less of the DPM mass.

DE emissions vary significantly in chemical composition and particle sizes between different engine types (heavy-duty, light-duty), engine operating conditions (idle, accelerate, decelerate), and fuel formulations (high/low sulfur fuel). Also, there are emission differences between on-road and nonroad engines simply because the nonroad engines to date are generally of older technology. The mass of particles emitted and the organic components on the particles from on-road diesel engines have been reduced over the years. Available data for on-road engines indicate that toxicologically relevant organic components of DE (e.g., PAHs, nitro-PAHs) emitted from older vehicle engines are still present in emissions from newer engines, though relative amounts have decreased. There is currently insufficient information to characterize the changes in the composition of DE from nonroad diesel engines over time.

1.3. DIESEL EXHAUST AS A COMPONENT OF AMBIENT PARTICULATE MATTER

DE is emitted from “on-road” diesel engines (vehicle engines) or “nonroad” diesel engines (e.g., locomotives, marine vessels, heavy-duty equipment, etc.). Nationwide, data in 1998 indicated that DE as measured by DPM made up about 6% of the total ambient $PM_{2.5}$ inventory (i.e., particles with aerodynamic diameter of 2.5 micrometers or less) and about 23% of the inventory, if natural and miscellaneous sources of $PM_{2.5}$ are excluded. Estimates of the DPM percentage of the total inventory in urban centers are higher. For example, estimates range from 10% to 36% in some urban areas in California, Colorado, and Arizona. Available data also indicate that over the years there have been significant reductions in DPM emissions from the exhaust of on-road diesel engines, whereas limited data suggest that exhaust emissions from nonroad engines have increased.

1.4. ATMOSPHERIC TRANSFORMATION OF DIESEL EXHAUST

After emission from the tailpipe, DE undergoes dilution and chemical and physical transformations in the atmosphere, as well as dispersion and transport in the atmosphere. The atmospheric lifetime for some compounds present in DE ranges from hours to days. DPM is directly emitted from diesel-powered engines (primary particulate matter) and can be formed from the gaseous compounds emitted by diesel engines (secondary particulate matter). Limited information is available about the physical and chemical transformation of DE in the

atmosphere. It is not clear what the overall toxicological consequences of DE's transformations are because some compounds in the DE mixture are altered to more toxic forms while others are made less toxic.

1.5. EXPOSURE TO DIESEL EXHAUST

DPM mass (expressed as $\mu\text{g DPM}/\text{m}^3$) has historically been used as a surrogate measure of exposure for whole DE. Although uncertainty exists as to whether DPM is the most appropriate parameter to correlate with human health effects, it is considered a reasonable choice until more definitive information about the mechanisms of toxicity or mode(s) of action of DE becomes available. In the ambient environment, human exposure to DE comes from both on-road and nonroad engine exhaust. A large percentage of the U.S. population also is exposed to ambient $\text{PM}_{2.5}$, of which DPM is typically a significant constituent. Although this document does not provide an exposure assessment, DE exposure information is included to provide a context for the health effects information. Exposure estimates for the early to mid-1990s suggest that national annual average DE exposure from on-road engines alone was in the range of about 0.5 to 0.8 $\mu\text{g DPM}/\text{m}^3$ of inhaled air in many rural and urban areas, respectively. Exposures could be higher if there is a nonroad DE source that adds to the exposure from on-road vehicles. For example, preliminary estimates show that, on a national average basis, accounting for nonroad DE emissions adds another twofold to the on-road exposure. For localized urban areas where people spend a large portion of their time outdoors, the exposures are higher and, for example, may range up to 4.0 $\mu\text{g DPM}/\text{m}^3$ of inhaled air.

1.6. HEALTH EFFECTS OF DIESEL EXHAUST

Available evidence indicates that there are human health hazards associated with exposure to DE. The hazards include acute exposure-related symptoms, chronic exposure-related noncancer respiratory effects, and lung cancer. The health hazard conclusions are based on exhaust emissions from diesel engines built prior to the mid-1990s. With current engine use including some new and many more older engines (engines typically stay in service for a long time), the health hazard conclusions, in general, are applicable to engines currently in use. As new and cleaner diesel engines, together with different diesel fuels, replace a substantial number of existing engines, the general applicability of the health hazard conclusions will need to be re-evaluated. With new engine and fuel technology expected to produce significantly cleaner engine exhaust by 2007 (e.g., in response to new federal heavy duty engine regulations), significant reductions in public health hazards are expected for those engine uses affected by the regulations.

1.6.1. Acute (Short-Term Exposure) Effects

Information is limited for characterizing the potential health effects associated with acute or short-term exposure. However, on the basis of available human and animal evidence, it is concluded that acute or short-term (e.g., episodic) exposure to DE can cause acute irritation (e.g., eye, throat, bronchial), neurophysiological symptoms (e.g., lightheadedness, nausea), and respiratory symptoms (cough, phlegm). There also is evidence for an immunologic effect—the exacerbation of allergenic responses to known allergens and asthma-like symptoms. The lack of adequate exposure-response information in the acute health effect studies precludes the development of recommendations about levels of exposure that would be presumed safe for these effects.

1.6.2. Chronic (Long-Term Exposure) Noncancer Respiratory Effects

Information from the available human studies is inadequate for a definitive evaluation of possible noncancer health effects from chronic exposure to DE. However, on the basis of extensive animal evidence, DE is judged to pose a chronic respiratory hazard to humans. Chronic-exposure, animal inhalation studies show a spectrum of dose-dependent inflammation and histopathological changes in the lung in several animal species including rats, mice, hamsters, and monkeys.

This assessment provides an estimate of inhalation exposure of DE (as measured by DPM) to which humans may be exposed throughout their lifetime without being likely to experience adverse noncancer respiratory effects. This exposure level, known as the reference concentration (RfC) for DE of $5 \mu\text{g}/\text{m}^3$ of DPM was derived on the basis of dose-response data on inflammatory and histopathological changes in the lung from rat inhalation studies. In recognition of the presence of DPM in ambient $\text{PM}_{2.5}$, it also is appropriate to consider the wealth of $\text{PM}_{2.5}$ human health effects data. In this regard, the 1997 National Ambient Air Quality Standard for $\text{PM}_{2.5}$ of $15 \mu\text{g}/\text{m}^3$ (annual average concentration) also would be expected to provide a measure of protection from DPM, reflecting DPM's current approximate proportion to $\text{PM}_{2.5}$.

1.6.3. Chronic (Long-Term Exposure) Carcinogenic Effects

This assessment concludes that DE is “likely to be carcinogenic to humans by inhalation” and that this hazard applies to environmental exposures. This conclusion is based on the totality of evidence from human, animal, and other supporting studies. There is considerable evidence demonstrating an association between DE exposure and increased lung cancer risk among workers in varied occupations where diesel engines historically have been used. The human evidence from occupational studies is considered strongly supportive of a finding that DE

exposure is causally associated with lung cancer, though the evidence is less than that needed to definitively conclude that DE is carcinogenic to humans. There is some uncertainty about the degree to which confounders are having an influence on the observed cancer risk in the occupational studies, and there is uncertainty evolving from the lack of actual DE exposure data for the workers. In addition to the human evidence, there is supporting evidence of DPM's carcinogenicity and associated DPM organic compound extracts in rats and mice by noninhalation routes of exposure. Other supporting evidence includes the demonstrated mutagenic and chromosomal effects of DE and its organic constituents, and the suggestive evidence for bioavailability of the DPM organics in humans and animals. Although high-exposure chronic rat inhalation studies show a significant lung cancer response, this is not thought predictive of a human hazard at lower environmental exposures. The rat response is considered to result from an overload of particles in the lung resulting from the high exposure, and such an overload is not expected to occur in humans at environmental exposures.

Although the available human evidence shows a lung cancer hazard to be present at occupational exposures that are generally higher than environmental levels, it is reasonable to presume that the hazard extends to environmental exposure levels. While there is an incomplete understanding of the mode of action for DE-induced lung cancer that may occur in humans, there is the potential for a nonthreshold mutagenic mode of action stemming from the organics in the DE mixture. A case for an environmental hazard also is shown by the simple observation that the estimated higher environmental exposure levels are close to, if not overlapping, the lower range of occupational exposures for which lung cancer increases are reported. These considerations taken together support the prudent public health choice of presuming a cancer hazard for DE at environmental levels of exposure. Overall, the evidence for a potential cancer hazard to humans resulting from chronic inhalation exposure to DE is persuasive, even though assumptions and uncertainties are involved. While the hazard evidence is persuasive, this does not lead to similar confidence in understanding the exposure/dose-response relationship.

Given a carcinogenicity hazard, EPA typically performs a dose-response assessment of the human or animal data to develop a cancer unit risk estimate that can be used with exposure information to characterize the potential cancer disease impact on an exposed population. The DE human exposure-response data are considered too uncertain to derive a confident quantitative estimate of cancer unit risk, and with the chronic rat inhalation studies not being predictive for environmental levels of exposure, EPA has not developed a quantitative estimate of cancer unit risk.

In the absence of a cancer unit risk, simple exploratory analyses were used to provide a perspective of the range of possible lung cancer risk from environmental exposure to DE. The analyses make use of reported lung cancer risk increases in occupational epidemiologic studies,

and the differences between occupational and environmental exposure. The purpose of having a risk perspective is to illustrate and have a sense of the possible significance of the lung cancer hazard from environmental exposure. The risk perspective cannot be viewed as a definitive quantitative characterization of cancer risk nor is it suitable for estimation of exposure-specific population risks.

1.7. SOURCES OF UNCERTAINTY

Even though the overall evidence for potential human health effects of DE is persuasive, many uncertainties exist because of the use of assumptions to bridge data and knowledge gaps about human exposures to DE and the general lack of understanding about underlying mechanisms by which DE causes observed toxicities in humans and animals. A notable uncertainty of this assessment is whether the health hazards identified from studies using emissions from older engines can be applied to present-day environmental emissions and related exposures, as some physical and chemical characteristics of the emissions from certain sources have changed over time. Available data are not sufficient to provide definitive answers to this question because changes in DE composition over time cannot be confidently quantified, and the relationship between the DE components and the mode(s) of action for DE toxicity is/are unclear. While recognizing the uncertainty, for this assessment a judgment is made that prior-year toxicologic and epidemiologic findings can be applied to more current exposures, both of which use DPM mass in air as the measure of DE exposure.

Other uncertainties include the assumptions that health effects observed at high doses may be applicable to low doses, and that toxicologic findings in laboratory animals generally are predictive of human responses. In the absence of a more complete understanding of how DE may cause adverse health effects in humans and laboratory animals, related assumptions (i.e., the presence of a biological threshold for chronic respiratory effects based on cumulative dosage and absence of a threshold for lung cancer stemming from subtle and irreversible effects) are considered reasonable and prudent.

Although parts of this assessment, particularly the noncancer RfC estimate, have been derived with a generic consideration of sensitive subgroups within the population, the actual spectrum of the population that may have a greater susceptibility to DE is unknown and cannot be better characterized until more information is available regarding the adverse effects of DPM in humans. Increased susceptibility, for example, could result from above-average increases in DE deposition and retention in the respiratory system or intrinsic differences in respiratory system tissue sensitivity. There is no DE-specific information that provides direct insight to the question of differential human susceptibility. Given the nature of DE's noncancer effects on the respiratory system it would be reasonable, for example, to consider possible vulnerable

subgroups to include infants/children, the elderly, or individuals with preexisting health conditions, particularly respiratory conditions.

In developing a perspective on the possible significance of the environmental cancer hazard of DE, this assessment uses information about the differences in the magnitude of DE exposures between the occupational and environmental settings. Although an appreciation for differences in exposure is needed only at an order-of-magnitude level for this assessment, one should recognize that individual exposure is a function of both the variable concentrations in the environment and the related breathing and particle retention patterns of the individual. Because of variations in these factors across the population, different subgroups could receive lower or higher exposure to DE than those groups mentioned in this assessment.

Lastly, this assessment considers only potential health effects from exposures to DE alone. Effects of DE exposure could be additive to or synergistic with concurrent exposures to many other air pollutants. However, in the absence of more definitive data demonstrating interactive effects (e.g., potentiation of allergenicity effects, potentiation of DPM toxicity by ambient ozone and oxides of nitrogen) from combined exposures to DE and other pollutants, it is not possible to address this issue. Further research is needed to improve the knowledge and data on DE exposures and potential human health effects, and thereby reduce uncertainties of future assessments of the DE health effects data.

2. DIESEL EXHAUST EMISSIONS CHARACTERIZATION, ATMOSPHERIC TRANSFORMATION, AND EXPOSURES

2.1. INTRODUCTION

This chapter provides background information relating to the diesel engine, the pollutants it emits, the history of its use in highway vehicles and railroad locomotives, diesel exhaust composition and emissions trends, and air pollution regulatory standards for diesel engines in the United States. The chapter also provides specific information about the physical and chemical composition of diesel exhaust, descriptions of its atmospheric transformations, observations of measured and modeled ambient concentrations (considered alone and as a component of atmospheric particles in general), some estimates of population exposures as well as a comparison of DPM with ambient fine particulate matter (PM_{2.5}). In addition, this chapter gives background information that is used in conjunction with toxicology and epidemiology data to formulate conclusions about human health hazards that are discussed in later chapters of this document. The exposure information does not represent a formal or rigorous exposure assessment; it is intended only to provide a context for the health effects data and health hazard findings.

For the purposes of this document, carbonaceous matter, diesel exhaust, diesel particulate matter, elemental carbon, organic carbon, soluble organic fraction, and soot are defined below.

Carbonaceous matter: Carbon-containing compounds that are associated with particulate matter in diesel exhaust. In this document, the term carbonaceous matter includes all organic and elemental carbon-containing compounds that are found in the particle phase. In other documents, this term is sometimes used interchangeably to refer to the insoluble fraction of diesel particulate matter or the soot fraction.

Diesel engine exhaust (DE): Gaseous and particle-phase emissions resulting from the combustion of diesel fuel in an internal-combustion, compression-ignition engine. DE includes emissions from a diesel engine or diesel vehicle (inclusive of aftertreatment devices), but does not include emissions from brake and tire wear.

Diesel particulate matter (DPM): The particle-phase compounds emitted in DE. DPM can refer to both primary emissions and secondary particles that are formed by atmospheric processes. In this document, DPM refers to primary particles. Primary diesel particles are considered fresh after being emitted and aged after

undergoing oxidation, nitration, or other chemical and physical changes in the atmosphere. As used in this document, DPM refers to both fresh and aged DPM unless a distinction is made.

Elemental carbon (EC): Carbon that has undergone pyrolysis (i.e., has been stripped of hydrogen). In pure form, EC contains only carbon atoms, although EC as it exists in combustion particulate matter is likely to contain some hydrogen atoms.

Organic carbon (OC): Carbon- and hydrogen-containing molecules emitted in DE largely as the result of unburned diesel fuel and, to a lesser extent, from engine lubrication oil. OC compounds also can contain oxygen, nitrogen, and sulfur, as well as other elements in small quantities.

Soluble organic fraction (SOF): The organic portion of DPM that can be extracted from the particle matrix into solution. Extraction solutions and procedures vary and are described in Section 2.2.8.1.

Soot: Agglomerations of EC and OC particles. Soot also is often characterized as the insoluble portion of DPM, and is therefore considered to be mainly EC by some investigators.

This chapter begins with a history of dieselization for on-road vehicles and locomotives, followed by an introductory discussion of the formation of primary diesel emissions to assist the reader in understanding the complex factors that influence the formation of particulate matter (PM) and other DE emissions. The next section is a summary of EPA emission standards for on-road and locomotive diesel engines and a description of the national trends in emissions from on-road and nonroad diesel engine sources based on inventory modeling. The chapter continues with a discussion of diesel fuel use and the impact of fuel properties on emissions. The chronological assessment of emissions factors is presented in summaries of chassis and engine dynamometer testing and tunnel tests. This is followed by a description of engine technologies and their effect on emissions, and a description of the chemical and physical nature of emissions. The data describing the important atmospheric transformations of DE are summarized. The chapter concludes with a summary of the available literature regarding atmospheric concentrations of DPM and exposures to DE. EPA has assessed national and urban-area annual average exposure to DPM using the Hazardous Air Pollutant Exposure Model, and this assessment is presented in Section 2.4.3. A full exposure assessment would include the

distribution of ambient DE exposures in different geographic regions and among different demographic groups, the most highly exposed (90th percentile), exposures in microenvironments for short and long durations, the maximum exposure range (98th percentile), and the number of maximum-exposed individuals. However, such an assessment is not currently available. EPA is developing tools to provide a more complete exposure assessment.

2.2. PRIMARY DIESEL EXHAUST EMISSIONS

2.2.1. History of Dieselization

The diesel engine was patented in 1892 by Rudolf Diesel, who conceived it as a prime mover that would provide much improved fuel efficiency compared with spark-ignition (SI) engines. To the present day, the diesel engine's excellent fuel economy remains one of its strongest selling points. In the United States, the diesel engine is used mainly in trucks, buses, agricultural and other nonroad equipment, locomotives, and ships.

The chief advantages of the diesel engine over the gasoline engine are its fuel economy and durability. Diesel engines, however, emit more PM per mile driven compared with gasoline engines of a similar weight. Over the past decade, modifications of engine components have substantially reduced particle emissions from both diesel and gasoline engines (Hammerle et al., 1994; Sawyer and Johnson, 1995).

The diesel engine compresses air to high pressure and temperature. Fuel, when injected into this compressed air, autoignites, releasing its chemical energy. The expanding combustion gases do work on the piston before being exhausted to the atmosphere. Power output is controlled by the amount of injected fuel rather than by throttling the air intake. Compared to its SI counterpart, the diesel engine's superior efficiency derives from a higher compression ratio and no part-load throttling. To ensure structural integrity for prolonged reliable operation at the higher peak pressures brought about by a higher compression ratio and autoignition, the structure of a diesel engine generally is more massive than its SI counterpart.

Diesel engines (also called compression-ignition) may be broadly identified as being either two- or four-stroke cycle, injected directly or indirectly, and naturally aspirated or supercharged. They also are classified according to service requirements such as light-duty (LD) or heavy-duty (HD) automotive/truck, small or large industrial, and rail or marine.

All diesel engines use hydraulic fuel injection in one form or another. The fuel system must meet four objectives if a diesel engine is to function properly over its entire operating range. It must: (1) meter the correct quantity of fuel, (2) distribute the fuel to the correct cylinder, (3) inject the fuel at the correct time, and (4) inject the fuel so that it is atomized and mixes well with the in-cylinder air. The first two objectives are functions of a well-designed injection pump, and the last two are mostly functions of the injection nozzle. Fuel injection

systems are moving toward the use of electronic components for more flexible control than is available with purely mechanical systems to obtain lower exhaust emissions without diminishing fuel efficiency.

Both the fuel and the lubricants that service diesel engines are highly finished petroleum-based products combined with chemical additives. Diesel fuel is a mixture of many different hydrocarbon molecules from about C₇ to about C₃₅, with a boiling range from roughly 350 °F to 650 °F. Many of the fuel and oil properties, such as specific energy content (which is higher than gasoline), ignition quality, and specific gravity, are related to hydrocarbon composition. Therefore, fuel and lubricant composition affect many aspects of engine performance, including fuel economy and exhaust emissions.

Complete and incomplete combustion of fuel in the diesel engine results in the formation of a complex mixture of gaseous (gas-phase hydrocarbons, CO, CO₂, NO, NO₂, SO₂) and particulate exhaust (carbonaceous matter, sulfate, and trace elements). Because of concerns over health effects associated with DE, EPA began regulating emissions from diesel engines in 1970 (for smoke) and then added regulations for gaseous emissions. EPA first regulated particulate emissions from HD diesels in 1988.

2.2.1.1. Dieselization of the On-Road Fleet

Because of their durability and fuel economy, the use of diesel engines, particularly in long-distance applications, has increased over the years. The Census of Transportation, Truck Inventory and Use Survey (TIUS) indicates that among Class 3-8 trucks, diesel engine use has increased more rapidly than gasoline engine use in the past 20 years. Truck classes are defined by gross vehicle weight as described in Table 2-1. Dieselization first occurred among Class 7 and 8 trucks. The TIUS indicates that 81.5% of diesel trucks on the road in 1963 were Class 7 or 8 trucks (Table 2-2). Class 7 sales became predominantly (>50%) diesel in the 1970s and Class 8 sales became predominantly diesel in the 1960s. Diesels did not make up a majority of class 5 and 6 sales until the 1990s (Figures 2-1 and 2-2). HD trucks have historically constituted the majority of diesel sales and mileage. However, an increasing number of LD diesel trucks have been sold domestically in recent years. In the 1990s, approximately one in three diesel trucks sold was a Class 1 or Class 2 vehicle. Diesel trucks have historically been driven more miles per truck than gasoline trucks. For example, the TIUS indicates that 59% of diesel trucks were driven more than 50,000 miles in 1963, compared with 3% of gasoline trucks.

Table 2-1. Vehicle classification and weights for on-road trucks

Class	Gross vehicle weight (lb)
1	<6,000
2	6,001–10,000
3	10,001–14,000
4	14,001–16,000
5	16,001–19,500
6	19,501–26,000
7	26,001–33,000
8A ^a	33,001–60,000
8B ^a	>60,000
Medium duty (MD)	10,001–19,500 (same as Classes 3–5)
Light-heavy duty (LHD)	19,501–26,000 (same as Class 6)
Heavy-heavy duty (HHD)	>26,001 (same as Class 7–8)

^aClass 8A and Class 8B are often considered together.

Table 2-2. Total (gas and diesel) diesel trucks in the fleet in 1992

Truck class	1992 gas and diesel trucks	1992 diesel trucks	% Diesels
Class 1 and 2 (Light duty)	55,193,300	1,387,600	3
Class 3, 4, and 5 (Medium duty)	1,258,500	326,300	26
Class 6 (Light heavy-duty)	732,300	273,800	37
Class 7 and 8 (Heavy heavy-duty)	2,016,600	1,725,300	86

Source: Census of Transportation, 1995.

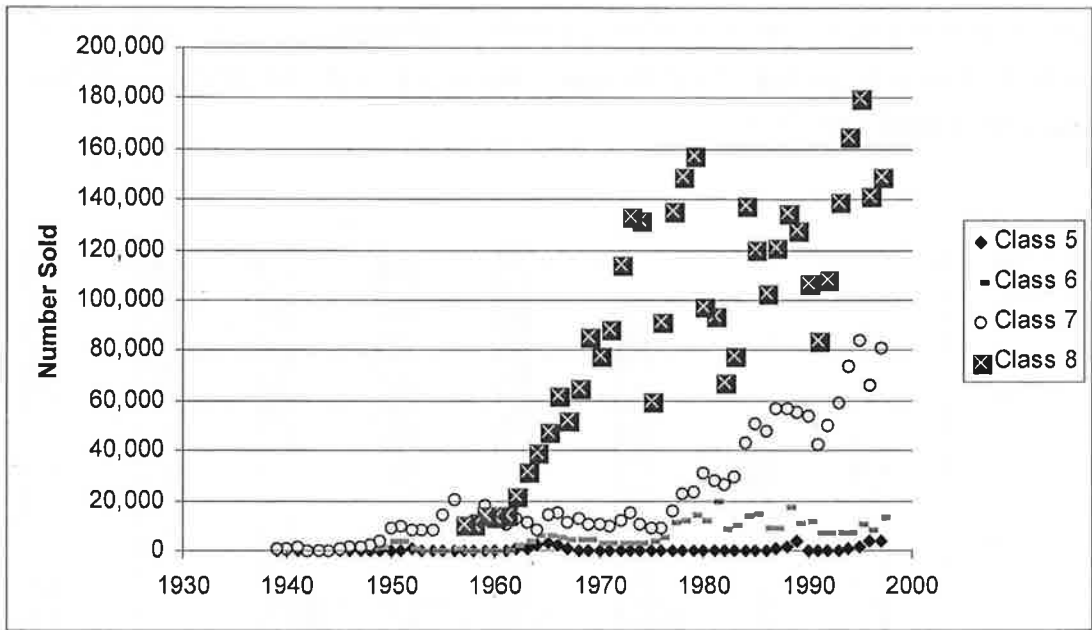


Figure 2-1. Diesel truck sales (domestic) for the years 1939-1997.

Source: AAMA, 1927-1974 and 1975-1998.

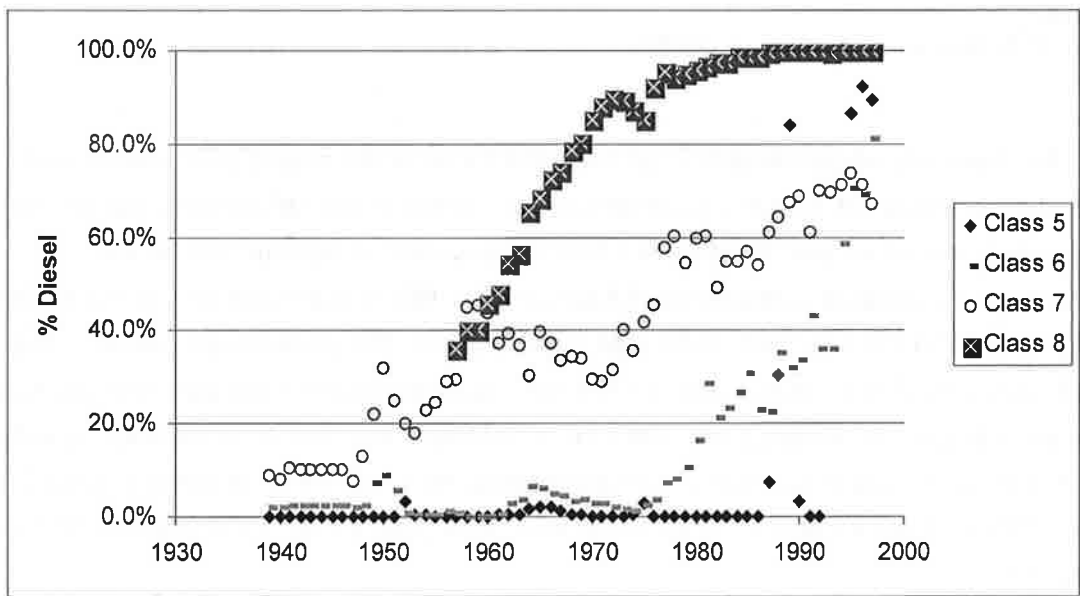


Figure 2-2. Diesel truck sales as a percentage of total truck sales for the years 1939-1997.

Source: AAMA, 1927-1974 and 1975-1998.

Among combination trucks, consisting of tractor-trailers and single-unit trucks with trailers, diesel vehicles have driven a majority of the miles since at least 1963, the first year in which TIUS was conducted (Figure 2-3).

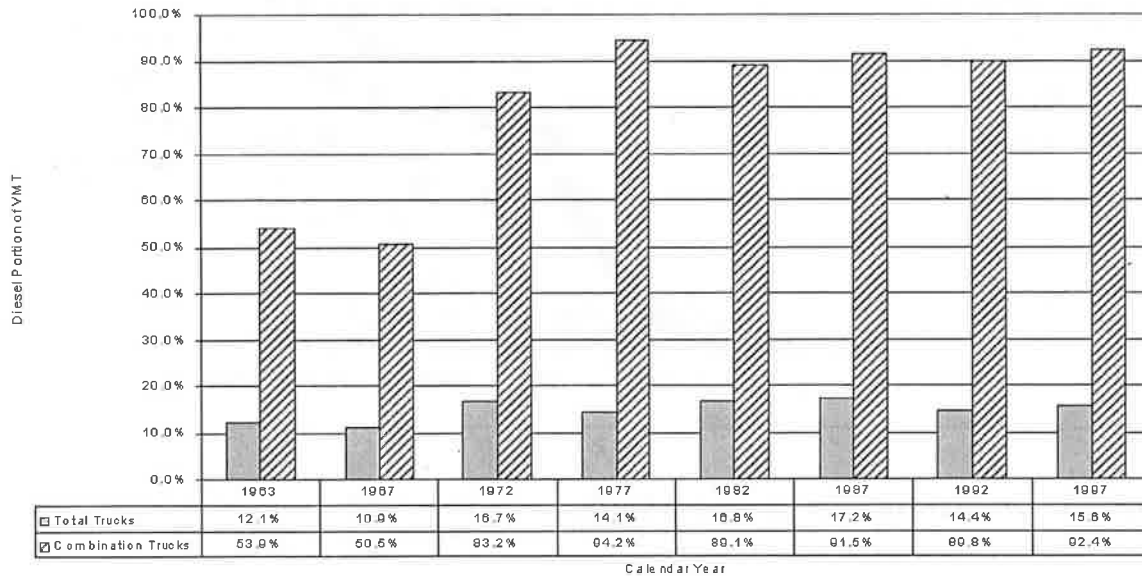


Figure 2-3. Percentage of truck miles attributable to diesel trucks. VMT= vehicle miles traveled.

Source: U.S. Bureau of the Census, 1999b.

The longevity of diesel trucks is an important factor to understand past, current, and projected exposures to DE because older vehicles are subject to less stringent regulations and may remain in use for several decades after their manufacture. American Automobile Manufacturers Association publications (AAMA, 1927-1997) indicate that 53% of trucks from model years 1947-1956 were still on the road after 14 years. The proportion of trucks in use after 14 years was 63% for model years 1974-1983, suggesting that the lifespan of trucks built in later years is longer. According to the 1997 TIUS, vehicles older than 10 years made up 40% of Class 7 and 8 trucks and 16% of Class 7-8 vehicle miles traveled (VMT) (Figures 2-4 and 2-5). Almost all Class 7 and 8 trucks were diesel vehicles in the period 1982-1997 (93% in 1982 and 99% in 1997).

2.2.1.2. Dieselization of Railroad Locomotive Engines

Early in the 20th century the political and economic pressure on the railroads to replace steam locomotives was substantial. Railroads were losing business to other forms of transport. The diesel-electric locomotive provided 90% in-service time, compared with only 50% for steam locomotives, and had three times the thermal efficiency (Klein, 1991; Kirkland, 1983).

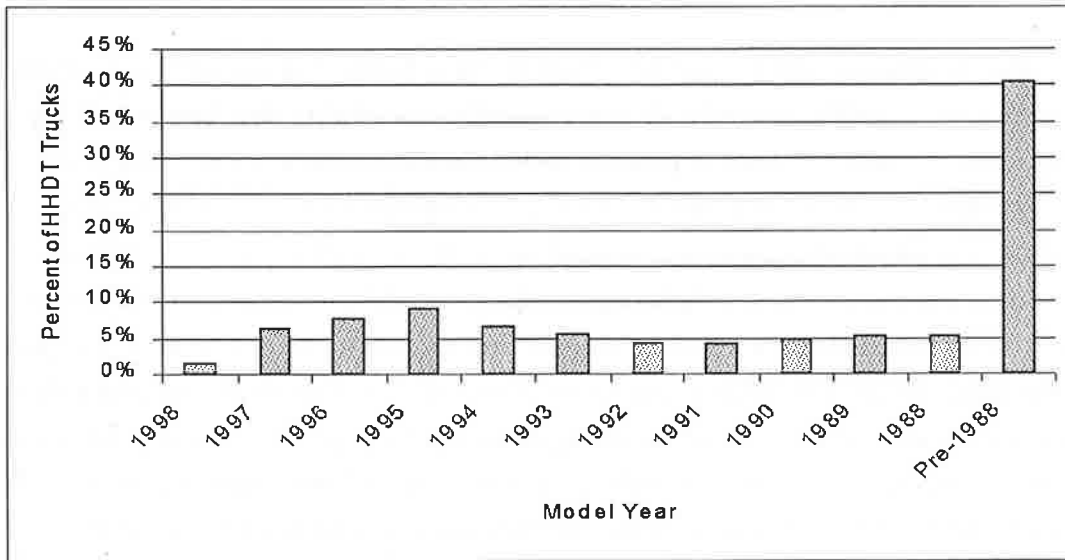


Figure 2-4. Model year distribution of in-use HD truck fleet in 1997.
 Source: U.S. Bureau of the Census, 1999b.

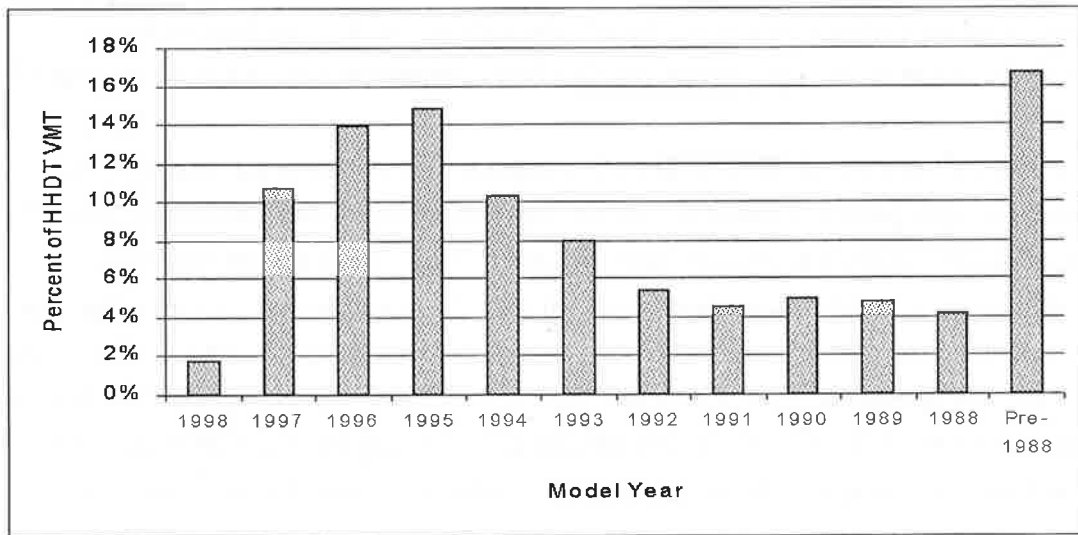


Figure 2-5. Model year distribution of vehicle miles traveled by the in-use HD truck fleet in 1997.
 Source: U.S. Bureau of Census, 1999b.

Additionally, several cities had passed laws barring steam locomotives within the city limits because the large quantities of smoke obscured visibility, creating a safety hazard. The first prototype diesel locomotive was completed in 1917. By 1924 General Electric (GE) was producing a standard line of switching locomotives on a production basis. Electro-Motive Corporation was founded the same year to produce diesel locomotives in competition with GE. This company was purchased in 1929 by General Motors (GM) and became the Electro-Motive Division. After this acquisition, GM began to develop the two-stroke engine for this application. Up to this time, all locomotive diesel engines were four-stroke. Two-strokes offered a much higher power-to-weight ratio, and GM's strategy was to get a large increase in power by moving to the two-stroke cycle. The first true high-speed, two-stroke, diesel-electric locomotives were produced by GM in 1935. However, because of the economic climate of the Great Depression, few of these were sold until after the Second World War. At the end of the war, most locomotives were still steam-driven but were more than 15 years old, and the railroads were ready to replace the entire locomotive fleet. Few, if any, steam locomotives were sold after 1945 because the entire fleet was converted to diesel (Coifman, 1994).

The locomotive fleet has included significant percentages of both two- and four-stroke engines. The four-stroke diesel engines were naturally aspirated in the 1940s and 1950s. It is unlikely that any of the two-stroke engines used in locomotive applications were strictly naturally aspirated. Nearly all two-stroke diesel locomotive engines are uniflow scavenged, with a positive-displacement blower for scavenging assistance. In 1975, it was estimated that 75% of the locomotives in service were two-stroke, of which about one-half used one or more turbochargers in addition to the existing positive-displacement blower for additional intake boost pressure.

Almost all of the four-stroke locomotive engines were naturally aspirated in 1975. Electronic fuel injection for locomotive engines was first offered in the 1994 model year (U.S. EPA, 1998b). All locomotive engines manufactured in recent years are turbocharged, aftercooled or intercooled four-stroke engines. In part, this is because of the somewhat greater durability of four-strokes, although impending emissions regulations may have also been a factor in this shift. The typical lifespan of a locomotive has been estimated to be more than 40 years (U.S. EPA, 1998b). Many of the smaller railroads are still using engines built in the 1940s, although the engines may have been rebuilt several times since their original manufacture.

2.2.2. Diesel Combustion and Formation of Primary Emissions

A basic understanding of diesel combustion processes can assist in understanding the complex factors that influence the formation of DPM and other DE emissions. Unlike SI combustion, diesel combustion is a fairly nonhomogenous process. Fuel is sprayed at high

pressure into the compressed cylinder contents (primarily air with some residual combustion products) as the piston nears the top of the compression stroke. The turbulent mixing of fuel and air that takes place is enhanced by injection pressure, the orientation of the intake ports (inducement of intake-swirl tangential to the cylinder wall), piston motion, and piston bowl shape. In some cases, fuel and air mixing is induced via injection of the fuel into a turbulence-generating pre-chamber or swirl chamber located adjacent to the main chamber (primarily in older, higher speed engines and some LD diesels). Examples of typical direct injection and indirect injection combustion systems are compared in Figure 2-6. Diesel combustion can be considered to consist of the following phases (Heywood, 1988; Watson and Janota, 1982):

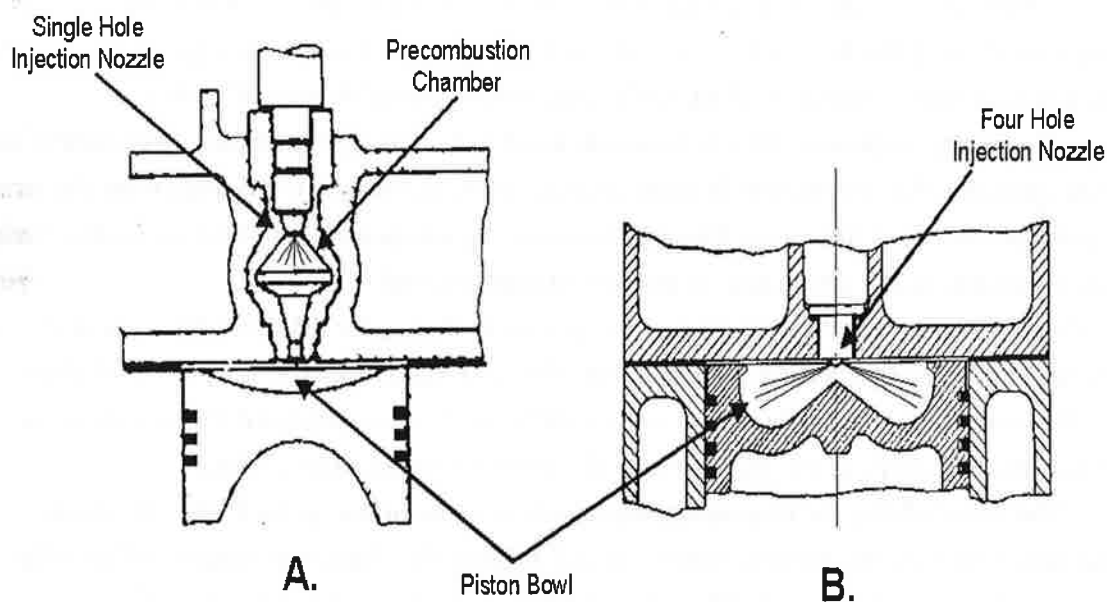


Figure 2-6. A comparison of IDI (A) and DI (B) combustion systems of high-speed HD diesel truck engines. DI engines almost completely replaced IDI engines for these applications by the early 1980s. (IDI = indirect injection, DI=direct injection)

- An ignition delay period, which starts after the initial injection of fuel and continues until the initiation of combustion. The delay period is governed by the rate of fuel and air mixing, diffusion, turbulence, heat transfer, chemical kinetics, fuel vaporization, and fuel composition. Fuel cetane rating is an indication of ignition delay.
- Rapid, premixed burning of the fuel and air mixture from the ignition delay period.
- Diffusion-controlled burning, where the fuel burns as it is injected and diffuses into the cylinder.
- A very small amount of rate-controlled burning during the expansion stroke, after the end of injection.

Engine speed and load are controlled by the quantity of fuel injected. Thus, the overall fuel-to-air ratio varies greatly as engine speed and load vary. On a macro scale, the cylinder contents are always fuel-lean. Depending on the time available for combustion and the proximity of oxygen, the fuel droplets are either completely or partially oxidized. At temperatures above 1,300 K, much of the unburned fuel that is not oxidized is pyrolyzed (stripped of hydrogen) to form EC (Dec and Espey, 1995). In addition to EC, other carbonaceous matter is present, largely from unburned fuel. The agglomeration of elemental and OC forms particles that are frequently referred to as “soot” particles. In this document, the terms “EC” and “OC” are used to refer to the carbon-containing components of DPM, and collectively, they are referred to as the carbonaceous fraction of a diesel particle.

Carbonaceous particle formation occurs primarily during the diffusion-burn phase of combustion, and is highest during high load and other conditions consistent with high fuel-air ratios. Most of the carbonaceous matter formed (80% to 98%) is oxidized during combustion, most likely by hydroxyl radicals (Kittelson et al., 1986; Foster and Tree, 1994).

DPM is defined by the measurement procedures summarized in the Code of Federal Regulations, Title 40 CFR, Part 86, Subpart N (CFR 40:86.N). These procedures define DPM emissions as the mass of material collected on a filter at a temperature of 52 °C or less after dilution of the exhaust with air. DPM is formed by a number of physical processes acting in concert as the exhaust is cooled and diluted. These are nucleation, coagulation, condensation, and adsorption. The core DE particles are formed by nucleation and coagulation from primary spherical particles consisting of solid carbonaceous (EC) material and ash (trace metals and other elements). To these, through coagulation, adsorption, and condensation, are added organic and sulfur compounds (sulfate) combined with other condensed material (Figure 2-7). Because of

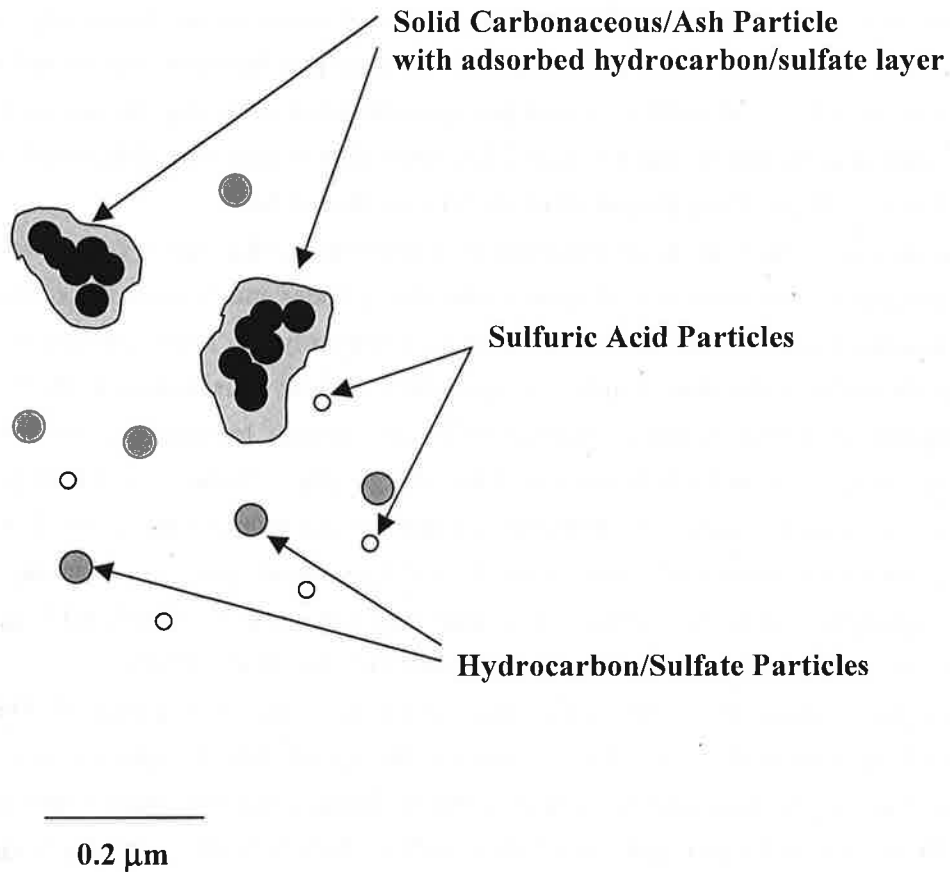


Figure 2-7. Schematic diagram of diesel engine exhaust particles.

Source: Modified from Kittelson, 1998.

their size, $<0.5 \mu\text{m}$, these particles have a very large surface area per gram of mass, which makes them able to adsorb large quantities of ash, organic compounds, and sulfate. The specific surface area of the EC core has been measured to be approximately $30\text{--}50 \text{ m}^2/\text{g}$ (Frey and Corn, 1967). Pierson and Brachaczek (1976) report that after the extraction of adsorbed organic material, the surface area of the diesel particle core is approximately $90 \text{ m}^2/\text{g}$.

The organic material associated with diesel particles originates from unburned fuel, engine lubrication oil, and small quantities of partial combustion and pyrolysis products. This is frequently quantified as the SOF, which is discussed in much more detail in Section 2.2.7. The formation of sulfate in DE depends primarily on fuel sulfur content. During combustion, sulfur compounds present in the fuel are oxidized to sulfur dioxide (SO_2). Approximately 1% to 4% of fuel sulfur is oxidized to form sulfuric acid (H_2SO_4) (Wall et al., 1987; Khatri et al., 1978; Baranescu, 1988; Barry et al., 1985). Upon cooling, sulfuric acid and water condense into an aerosol that is nonvolatile under ambient conditions. The mass of sulfuric acid DPM is more than doubled by the mass of water associated with the sulfuric acid under typical DPM measurement conditions (50% relative humidity, $20\text{--}25 \text{ }^\circ\text{C}$) (Wall et al., 1987).

Emissions from combustion engines produce oxide of nitrogen (NO_x) primarily (at least initially) as of NO. High combustion temperatures cause reactions between oxygen and nitrogen to form NO and some NO_2 . Most NO_2 formed during combustion is rapidly decomposed. NO can also decompose to N_2 and O_2 , but the rate of decomposition is very slow (Heywood, 1988; Watson and Janota, 1982). Thus, almost all of the NO_x emitted is NO.

Some organic compounds from unburned fuel and from lubricating oil consumed by the engine can be trapped in crevices or cool spots within the cylinder and thus are not sufficiently available to conditions that would lead to their oxidation or pyrolysis. These compounds are emitted from the engine and either contribute to gas-phase organic emissions or to DPM emissions, depending on their volatility. Within the exhaust system, temperatures are sufficiently high that these compounds are entirely present within the gas phase (Johnson and Kittelson, 1996). Upon cooling and mixing with ambient air in the exhaust plume, some of the less volatile organic compounds can adsorb to the surfaces of the EC agglomerate particles. Lacking sufficient EC adsorption sites, the organic compounds may condense on sulfuric acid nuclei to form a heterogeneously nucleated organic aerosol (Abdul-Khalek et al., 1999).

Although not unique to DE, the high content of EC associated with typical DPM emissions has long been used by some investigators to distinguish diesel engine sources of this particle from other combustion aerosols. Diesel particles from newer HD engines are typically composed of ~75% EC (EC can range from 33% to 90%), ~20% OC (OC can range from 7% to 49%), and small amounts of sulfate, nitrate, trace elements, water, and unidentified components (Figure 2-8). Metallic compounds from engine component wear, and from compounds in the fuel and lubricant, contribute to DPM mass. Ash from oil combustion also contributes trace amounts.

Ambient $\text{PM}_{2.5}$ measured in the eastern United States is dominated by sulfate (34%), whereas ambient $\text{PM}_{2.5}$ in the western United States is dominated by OC (39%) (Table 2-3) (U.S. EPA, 1999a). Many sources contribute to ambient $\text{PM}_{2.5}$, and these sources and their relative contribution to ambient $\text{PM}_{2.5}$ can be identified on the basis of the chemical species present. The OC fraction of DPM is increasingly being used to assist investigators in identifying the contribution of diesel engine emissions to ambient $\text{PM}_{2.5}$. In particular, hopane and sterane compounds (aromatic compounds, $>\text{C}_{30}$) have been used in addition to other polycyclic aromatic hydrocarbons (PAHs) and long-chain alkanes to distinguish DPM from other mobile source PM and from ambient PM (Schauer et al., 1996; Fujita et al., 1998). Although PAH compounds make up 1% or less of DPM mass, diesel emissions have been observed to have elevated concentrations of methylated naphthalenes and methylated phenanthrene isomers compared to other combustion aerosols (Benner et al., 1989; Lowenthal et al., 1994; Rogge et al., 1993). Enrichment of benzo[*a*]anthracene and benzo[*a*]pyrene (B[*a*]P) in DPM has also been

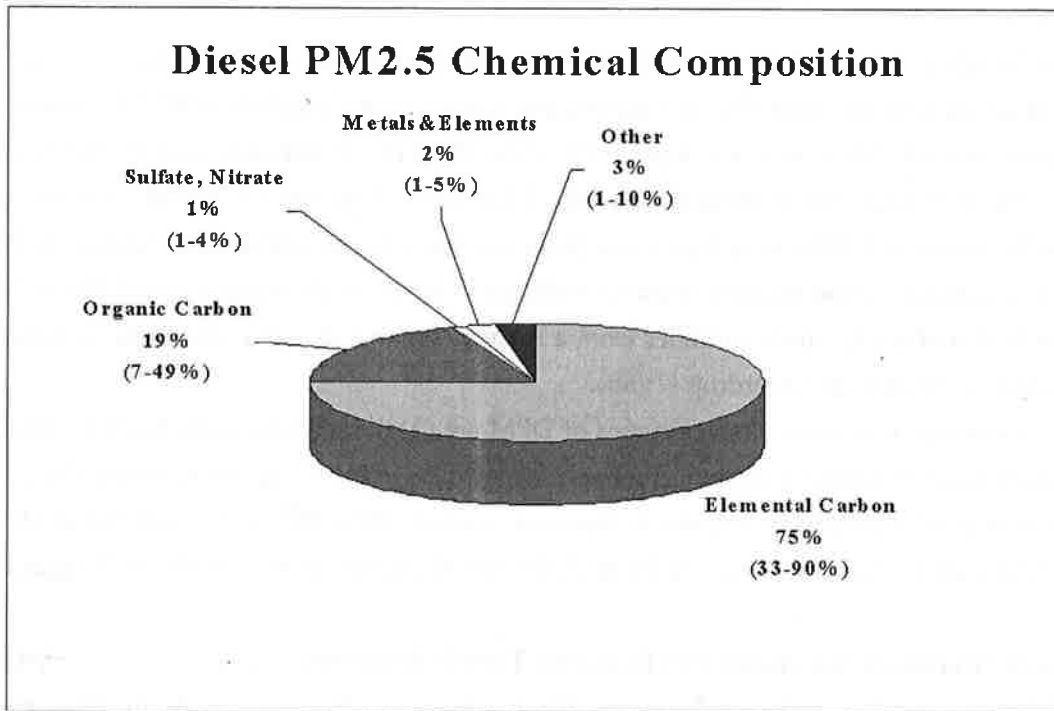


Figure 2-8. Typical chemical composition for diesel particulate matter (PM_{2.5}) from new (post-1990) HD diesel vehicle exhaust.

Table 2-3. Typical chemical composition of fine particulate matter

	Eastern U.S.	Western U.S.	Diesel PM _{2.5}
Elemental carbon	4%	15%	75%
OC	21%	39%	19%
Sulfate, nitrate, ammonium	48%	35%	1%
Minerals	4%	15%	2%
Unknown	23%	—	3%

Source: U.S. EPA, 1999a.

observed under some conditions and has been used to assess the relative contribution of DE to ambient PM.

Although specific OC species are being used to help distinguish DPM aerosols from other combustion aerosols, up to 90% of the organic fraction associated with DPM is currently classified as unresolvable complex material. Ultrafine DPM (5–50 nm) accounts for the majority (50% to 90%) of the number of particles but only 1% to 20% of the mass of DPM. A study conducted by Gertler (1999) in the Tuscarora Mountain tunnel demonstrated an increase in 20 nm diameter particles as the fraction of diesel vehicles in the tunnel increased from 13% to 78%. The contribution of nuclei-mode particles from a freeway on an ambient aerosol size distribution was reported by Whitby and Sverdrup (1980).

In summary, four main characteristics of DPM are (1) the high proportion of EC, (2) the large surface area associated with the carbonaceous particles in the 0.2 μm size range, (3) enrichment of certain polycyclic organic compounds, and (4) 50%–90% of the number of DPM particles in diesel engine exhaust are in the nuclei-mode size range, with a mode of 20 nm.

2.2.3. Diesel Emission Standards and Emission Trends Inventory

EPA set a smoke standard for on-road HD diesel engines beginning with the 1970 model year and added a carbon monoxide (CO) standard and a combined hydrocarbon (HC) and NO_x standard for the 1974 model year (Table 2-4). Beginning in the 1979 model year, EPA added a HC standard while retaining the combined HC and NO_x standard. All of the testing for HC, CO, and NO_x was completed using a steady-state test procedure. Beginning in the 1985 model year, EPA added a NO_x standard (10.7 g/bhp-hr), dropped the combined HC and NO_x standard, and converted from steady-state to transient testing for HC, CO, and NO_x emissions. EPA introduced a particulate standard for 1988 model year diesel engines using the transient test (0.6 g/bhp-hr). Transient testing involves running an engine on a dynamometer over a range of load and speed set points.

Since the 1985 model year, only the NO_x and particulate standards have been tightened for on-road diesel engines. For truck and bus engines, the particulate standard was reduced to 0.25 g/bhp-hr in 1991, and it was reduced again in 1994 for truck engines to 0.1 g/bhp-hr. For urban bus engines, the particulate standard was reduced in 1994 to 0.07 g/bhp-hr and again in 1996 to 0.05 g/bhp-hr. The NO_x standard was reduced to 4.0 g/bhp-hr in 1998 for all on-road diesel engines (bus and truck engines). The standards for nonmethane hydrocarbon (NMHC) and NO_x combined were further lowered in a 1997 rulemaking, to take effect in 2004. EPA has recently finalized a regulation that will further reduce NO_x , NMHC, and PM emissions from diesel engines starting in 2007.