Assessment of Highway Particulate Impacts: Phase I, Task A—Problem Evaluation, Final Report

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FOREWORD

The highway system and its traffic have been found to be a source of particulate matter suspended in air, which can contaminate ambient air.

The national ambient air quality standard for the size of particulate matter when this investigation was underway was $\leq 10\ \mu m$ (PM10) (24-h average of 150 $\mu g/m^3$; annual average of 50 $\mu g/m^3$). This standard replaced a standard for total suspended particulate matter. Since 1997, there has been a new standard for ambient air quality for the size of particulate matter of $\leq 2.5\ \mu m$ (24-h average of 65 $\mu g/m^3$; annual average of 15 $\mu g/m^3$).

Phase I reports are *An Evaluation of the Highway Particulate Impacts* and *Field Sampling and Evaluation Program*. Phase II of the study evaluated procedures for assessing PM10 "emission" and dispersion rates from highways, mitigation measures, and perspectives.

Related studies have been done over many years. Three pertinent efforts recently started under the National Cooperative Highway Research Program (NCHRP) are:

- 25-14, Heavy-Duty Vehicle Emissions.
- 25-17, Transportation Effects of the 1997 Changes to the National Ambient Air Quality Standards.
- 25-18, Particulate Matter (PM2.5 and PM10) Apportionment for On-Road Mobile Sources.

These NCHRP investigations will build on and supplement this FHWA study. Reports are expected in approximately 3 years. The two FHWA Phase I reports, which are being submitted to the National Technical Information Service (NTIS), will be of interest to State and local government personnel, and academic and consulting specialists concerned with the effect of mobile sources on ambient air quality.

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Office of Infrastructure
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NOTICE

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16. Abstract

This work was performed during the first phase of the above-referenced contract. This report provides an assessment of highway particulate impacts. It evaluates the existing state of knowledge related to: (1) what the sources of the particulates are, especially those specific to highways; (2) methods of assessing particulate impacts from highway sources; (3) relationships between highway derived particulates and other particulate sources; (4) particulate instrumentation, measurement, and monitoring methods; (5) assessment potential public health risks; and (6) control strategies for mitigating highway particulate impacts. Sources evaluated included diesel and spark ignition engine emissions, tire and brake wear, resuspensions from road sanding and salting operations, and unpaved roads. Models evaluated included dispersion, source/receptor, and emissions modeling. Based on this information, four sources of highway particulates were determined to need additional study in order to more completely understand the highway particulate source problem. These areas were: paved road resuspension, dust suspension resulting from road sanding and salting operations, diesel engine emissions, and unpaved roads. Although unpaved roads were considered outside of the scope of the work of this contract, the other three areas were targeted for additional field work as part of Phase I. Details of the field work appear in the second report, namely, Report No. FHWA-RD-99-181, Assessment of Highway Particulate Impacts: Phase I, Tasks B and C--Field Sampling and Evaluation Program, Final Report.

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SI* (MODERN METRIC) CONVERSION FACTORS

APPROXIMATE CONVERSIONS TO SI UNITS

Symbol	When You Know	Multiply By	To Find	Symbol
		LENGTH		
in	inches	25.4	millimeters	mm
ft	feet	0.305	meters	m
yd	yards	0.914	meters	m
mi	miles	1.61	kilometers	km
		AREA		
in^2	square inches	645.2	square millimeters	mm^2
ft^2	square feet	0.093	square meters	m^2
yd^2	square yards	0.836	square meters	m^2
ac	acres	0.405	hectares	ha
mi^2	square miles	2.59	square kilometers	km^2
		VOLUME		
fl oz	fluid ounces	29.57	milliliters	mL
gal	gallons	3.785	liters	L
ft^3	cubic feet	0.028	cubic meters	m^3
yd^3	cubic yards	0.765	cubic meters	m^3

NOTE: Volumes greater than 1000 l shall be shown in m^3.

Symbol	When You Know	Multiply By	To Find	Symbol
		MASS		
oz	ounces	28.35	grams	g
lb	pounds	0.454	kilograms	kg
T	short tons (2000 lb)	0.907	megagrams (or "metric ton")	Mg (or "t")
		TEMPERATURE (exact)		
°F	Fahrenheit temperature	5(F-32)/9 or (F-32)/1.8	Celcius temperature	°C
		ILLUMINATION		
fc	foot-candles	10.76	lux	lx
fl	foot-Lamberts	3.426	candela/m^2	cd/m^2
		FORCE and PRESSURE or STRESS		
lbf	poundforce	4.45	newtons	N
lbf/in^2	poundforce per square inch	6.89	kilopascals	kPa

APPROXIMATE CONVERSIONS FROM SI UNITS

Symbol	When You Know	Multiply By	To Find	Symbol
		LENGTH		
mm	millimeters	0.039	inches	in
m	meters	3.28	feet	ft
m	meters	1.09	yards	yd
km	kilometers	0.621	miles	mi
		AREA		
mm^2	square millimeters	0.0016	square inches	in^2
m^2	square meters	10.764	square feet	ft^2
m^2	square meters	1.195	square yards	yd^2
ha	hectares	2.47	acres	ac
km^2	square kilometers	0.386	square miles	mi^2
		VOLUME		
mL	milliliters	0.034	fluid ounces	fl oz
L	liters	0.264	gallons	gal
m^3	cubic meters	35.71	cubic feet	ft^3
m^3	cubic meters	1.307	cubic yards	yd^3
		MASS		
g	grams	0.035	ounces	oz
kg	kilograms	2.202	pounds	lb
Mg (or "t")	megagrams (or "metric ton")	1.103	short tons (2000 lb)	T
		TEMPERATURE (exact)		
°C	Celcius temperature	1.8C + 32	Fahrenheit temperature	°F
		ILLUMINATION		
lx	lux	0.0929	foot-candles	fc
cd/m^2	candela/m^2	0.2919	foot-Lamberts	fl
		FORCE and PRESSURE or STRESS		
N	newtons	0.225	poundforce	lbf
kPa	kilopascals	0.145	poundforce per square inch	lbf/in^2

* SI is the symbol for the International System of Units. Appropriate rounding should be made to comply with Section 4 of ASTM E380.

(Revised September 1993)

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

LIST OF ABBREVIATIONS AND SYMBOLS

ADT	average daily traffic
ADTV	average daily traffic volume
Al	aluminum
APCC	Arizona Portland Cement Company
ADEQ	Arizona Department of Environmental Quality
Ba	barium
Br	bromine
CAA	Clean Air Act
CAAA	Clean Air Act Amendments of 1990
Ca	calcium
CARB	California Air Resources Board
CEB	chemical element balance
CEQA	California Environmental Quality Act
CH_4	methane
Cl	chlorine
CMB	chemical mass balance
CO	carbon monoxide
CO_2	carbon dioxide
CRC	Coordinating Research Council
CSMCS	Carbonaceous Species Methods Comparison Study
Cu	copper
CVSA	Coachella Valley Study Area
DOT	Department of Transportation
EC	elemental carbon
EEM	Energy & Environmental Management, Inc.
EF	emission factor
EGR	exhaust gas recirculation
EPA	U.S. Environmental Protection Agency
ERT	Environmental Research and Technology
EV	effective variance
FDM	fugitive dust model
Fe	iron
FHWA	Federal Highway Administration
FRG	Federal reformulated gasoline
FTA	Federal Transit Authority
ft^3	cubic feet
ft^3/min	cubic feet per minute
g/mi/s	grams per mile per second
g/bhp/h	grams per brake horsepower-hour
g/mi	grams per mile
GMR	General Motors Research Laboratory
GVWR	Gross Vehicle Weight Rating
HC	hycrocarbon
HCHO	formaldehyde
HDE	heavy-duty engine
HDV	heavy-duty vehicle
IC	Ion Chromatography
I/M	inspection and maintenance

IMPROVE	Interagency Monitoring of Protected Visual Environments
ISCST	industrial source complex model short-term
ISWS	Illinois State Water Survey
K	potassium
km	kilometer
LDT	light-duty truck
LDV	light-duty vehicle
LEV	low-emission vehicle
LIPM	Laser Integrating Plate Method
L/min	liters per minute
µm	micrometer
µg/m^3	micrograms per cubic meter
mg/km	milligrams per kilometer
mi/h	miles per hour
Mg	magnesium
MLR	multiple linear regression
Mo	molybdenum
MPO	Metropolitan Planning Organization
MRI	Midwest Research Institute
NAAQS	National Ambient Air Quality Standard
NAPAP	National Acid Precipitation Assessment Program
NEPA	National Environmental Policy Act
NH_3	ammonia
NLEV	National low-emission vehicle
NMHC	nonmethane hydrocarbon
NMOG	nonmethane organic gases
NO_2	nitrogen dioxide
NO_x	nitrogen oxides
OBD	on-board diagnostic
OC	organic carbon
ODEQ	Oregon Department of Environmental Quality
OGC	Oregon Graduate Center
OTR	Ozone Transport Region
PACS	Portland Aerosol Characterization Study
PAG	Pima Association of Governments
Pb	lead
PCA	principal component analysis
PESA	Proton Elastic Scattering Analysis
PIXE	Proton Induced X-ray Emission
PLS	partial least squares
PM	particulate matter
PM-10	particles with an aerodynamic diameter less than or equal to a nominal 10 micrometers
PM-2.5	particles with an aerodynamic diameter less than or equal to a nominal 2.5 micrometers
RESOLVE	Research on Operations Limiting Visual Extinction
RFP	reasonable further progress
RH	relative humidity
RP	respirable particulates
S	sulfur

SBC	Santa Barbara County
SBR	styrene-butadiene rubber
SCAQMD	South Coast Air Quality Management District
SCAQS	Southern California Air Quality Study
SCENIC	Study Cooperative for Emissions and Impact Characterization
SEDAB	Southeast Desert Air Basin
SFS	sequential filter sampler
SJV	San Joaquin Valley
SI	silicon
SIP	State Implementation Plan
Sn	tin
SO_2	sulfur dioxide
SOA	secondary organic aerosols
SVD	singular value decomposition
SVPA	Searles Valley Planning Area
TCMs	Transportation Control Measures
Ti	titanium
TIP	Transportation Improvement Program
TIUS	Truck Inventory and Use Survey
TLEV	transitional low-emission vehicle
TOR	thermal optical reflectance
TSP	total suspended particulate
ULEV	ultra-low emission vehicle
U.S.	United States
USS	United States Steel Corporation
VAQS	Valley Air Quality Study
VIEW	Visibility Investigative Experiment in the West
VMT	vehicle miles traveled
VOC	volatile organic compounds
WFPN	Western Fine Particle Network
ZEV	zero-emission vehicle
Zn	zinc

CHAPTER 1: INTRODUCTION

On July 1, 1987, the United States (U.S.) Environmental Protection Agency (EPA) published a final rule embodying an ambient air quality standard for particulate matter, size less than or equal to 10 micrometers (µm [PM-10]) designed to replace the existing particulate standard commonly referred to as total suspended particulate matter (TSP). The Federal Highway Administration (FHWA) of the Department of Transportation (DOT) needed an assessment of the potential impacts that highway sources have on the PM-10 standard. In order to assess highway particulate impacts, a two-phase, multi-task approach was initiated in 1991. Phase I consisted of: (A) an evaluation of the problem; (B) development of a field sampling and evaluation plan; (C) field sampling; and (D) reporting. Phase II, consisted of: (A) development of concentration prediction mechanisms; (B) proposal of mitigation guidance; (C) development of perspectives and rationales for assessing highway particulate impacts; and (D) reporting. Phase II followed the successful completion of Phase I.

This paper represents the effort conducted for Phase I, Task A above.

GOALS AND OBJECTIVES

The overall goal of this working paper is to provide input to the FHWA as part of an evaluation of the problem of highway particulate impacts (Phase I, Task A). An adequate assessment of highway particulate impacts requires information related to several distinct areas. Specific to this goal, the following areas are discussed:

- **Knowledge of what the sources of particulates are, especially those specific to highways.** This includes primary particulates derived from tailpipe emissions or from brake and tire wear as well as secondary particulates which result from condensation, chemical reactions, or resuspension of materials either blown or carried onto the road surface from other sources.

- **Methods of assessing particulate impacts from highway sources.** This can include utilization of dispersion models, source-receptor models or emission rates/emission models.

- **Relationships between highway derived particulates and other particulate sources.** This includes an understanding of the relative magnitude of emissions from various highway sources, the relative magnitude of emissions of highway sources and nonhighway sources, and those sources that can be considered exceptional or nonrecurring, but may impact monitoring sites. Highway construction maintenance is an example of the latter. Results from other studies are helpful in evaluating these relationships.

- **Particulate instrumentation, measurement, and monitoring methods.**

- **Assessment of potential public health risks.** This includes an assessment of the chemistry of the particulates emitted from gasoline and diesel vehicles, determination of the speciation of particulate emissions, and potential mutagenetic effects.

- **Control strategies for highway particulate impacts.** This includes an assessment of both new regulations as well as an understanding of the control strategies that are currently being implemented in States that have PM-10 nonattainment areas.

In all these areas, emphasis is placed on the evaluation of PM-10. However, in certain cases, TSP will also be discussed, since much of the earlier work on highway vehicle particulates was performed when the existing National Ambient Air Quality Standard (NAAQS) for particulates was for TSP rather than PM-10.

This report is organized into eight chapters. Chapter 2 explores the sources of particulates. Chapter 3 examines methods used to evaluate the impacts of highway particulates sources. Chapter 4 assesses the relationship between highway particulate sources and other particulate sources. Chapter 5 details particulate instrumentation, measurement programs and strategies, and monitoring methods. Chapter 6 examines the potential public health risks of highway particulate emissions. Chapter 7 provides information related to the control of highway particulates. Chapter 8 provides references for chapters 1 through 7.

CHAPTER 2: SOURCES OF HIGHWAY PARTICULATES

Highway particulate sources can be broken into two major categories: primary particulate emissions and secondary particulate emissions. Primary emissions are broadly defined as those emissions directly emitted from the vehicle itself either as the consequence of the engine combustion process or as the result of the wear and degradation of the vehicle itself. Secondary particulate emissions encompass all other particulate emissions from highway sources. This can include chemical transformations, reactions resulting in particulates, resuspension of material carried or tracked onto the road surface, or pavement wear. This section examines information related to both primary and secondary particulate sources and includes the results of various studies designed to assess the particulate impacts for these two major highway particulate source categories.

DIESEL PARTICLE EMISSIONS

General Characteristics

Diesel engines manufacture soot particles and emit them into the air. This process has always been the main limitation to available power from a particular engine design; for many years, peak power was determined by the fueling rate at first visible smoke, usually Ringelmann No. 2. Therefore, diesel particle emissions have always played an important role in engine design.

In the early 1970's, interest in the influence of fuel and fuel additives on emissions, particularly of toxic substances, extended to heavy-duty truck and bus engines. Early studies under this aegis produced sampling and analytical methods useful in defining these particle emissions. When diesel power plants for U.S. passenger cars became a major production strategy in the late 70's and early 80's, EPA began a significant research program to define the extent of any new environmental problem this change might cause. New studies of emissions, atmospheric chemistry, and health effects were quickly pursued. However, the diesel engine for passenger cars was a major market failure, at least in the domestic fleet. Sales, which were designed to exceed 20 percent of the market by the mid-80's, never topped 8 percent and are now well below 0.5 percent of the U.S. market. Environmental studies from this period are still relevant and these make up the major body of evidence available to judge the potential for environmental harm from diesel particles.

The intense research of the early 80's resulted in a number of special reports summarizing major research conclusions and results. In this compendium, reference is frequently made to these compendia and overview studies, including the National Research Council study on diesel car impacts (National Research Council, 1982), the very recent draft EPA study on diesel exhaust toxicity (EPA, 1991a) and the IARC monograph, Volume 46, on Diesel and Gasoline Engine Exhaust and some nitroarenes resulting from these exhausts (IARC, 1989). The general philosophy will be to summarize the information contained within these compendia and to update the information, particularly with evidence from direct measurements of engine and vehicle emissions. To this point, studies characterizing the direct emissions from diesel vehicles have received surprisingly little attention in the analysis of particle impacts. To understand the future significance of diesel particle control measures, it will be necessary to consider the nature and quantity of emissions measured from engines of the past. Then, both the degree of success and impact of recently developed diesel particle emissions controls will be much better understood.

Diesel exhaust particles are made up of four major components, a carbon black matrix supporting an oily, organic film, minor amounts of alkaline earth metal oxides, and the anions sulfate and nitrate. These may vary with conditions, lubricant, and fuel.

Diesel Exhaust Particle Size

Most direct emissions determinations have found three size ranges or modes of directly emitted particles. Figure 1 demonstrates the three modes and their approximate contribution to emitted particle mass (National Research Council, 1982). Usually a very small proportion of diesel particles are in the nuclei mode, roughly 0.01 µm in diameter. These particles are considered to have been freshly condensed from the gas phase and not yet agglomerated to the next larger size. The major portion of the particle mass or volume is in the accumulation mode, with approximate diameter of 0.2 µm. Smaller particles agglomerate to form quite stable particles of this characteristic size. Because of relatively small diffusion rates, these larger particles undergo collisions only very infrequently and consequently grow no larger. The third size range shown is the coarse mode, generally in the range of 2 to 15 µm. These particles consist mainly of materials previously deposited within the engine or exhaust system and subsequently ablated from the deposit layer by mechanical or temperature stress (National Research Council, 1982). Engine or vehicle emissions measurements frequently contain varying amounts of ablated deposits, particularly in measurements associated with cold starts.

More recently, Kittelson et al. (1988) have reported on an extensive set of field measurements for Coordinating Research Council (CRC). These authors measured the physical properties of diesel particles in the air on roadways by following moving vehicles in a vehicle which sampled and analyzed particle composition. Since no starting operations or steep accelerations could be sampled in this way, these measurements contained little evidence of ablated particles. On the other hand, there was a wide vehicle-to-vehicle range in the nuclei mode, ultrafine particles found in air. The authors note that the amount of this ultrafine material correlated very well with the solvent extractable organic fraction of the sampled bulk particles. Therefore, they concluded that this ultrafine material was mainly composed of organic material condensed from the gas phase near the end of the exhaust pipe. It is plausible, then, that the variability in nuclei mode particulate material and in organics generally is associated with engine-to-engine variability in engine oil consumption.

These authors report that 85 percent of the particulate volume and mass has a characteristic centroid dimension of about 0.3 µm and is the accumulation mode (Kittelson et al., 1988). Therefore, both direct emissions and air measurements behind diesel vehicles confirm that about 85 percent of diesel particle mass is in the submicronic size range and would be incorporated in a PM-10 classification. These studies also contain some evidence that a component of the organic material associated with diesel emissions can be volatilized in the air. This may lead to some variability in the estimation of effective particle mass emitted to the air amounting to about 13 percent of the mass. This volatilization matter will be discussed at more length later.

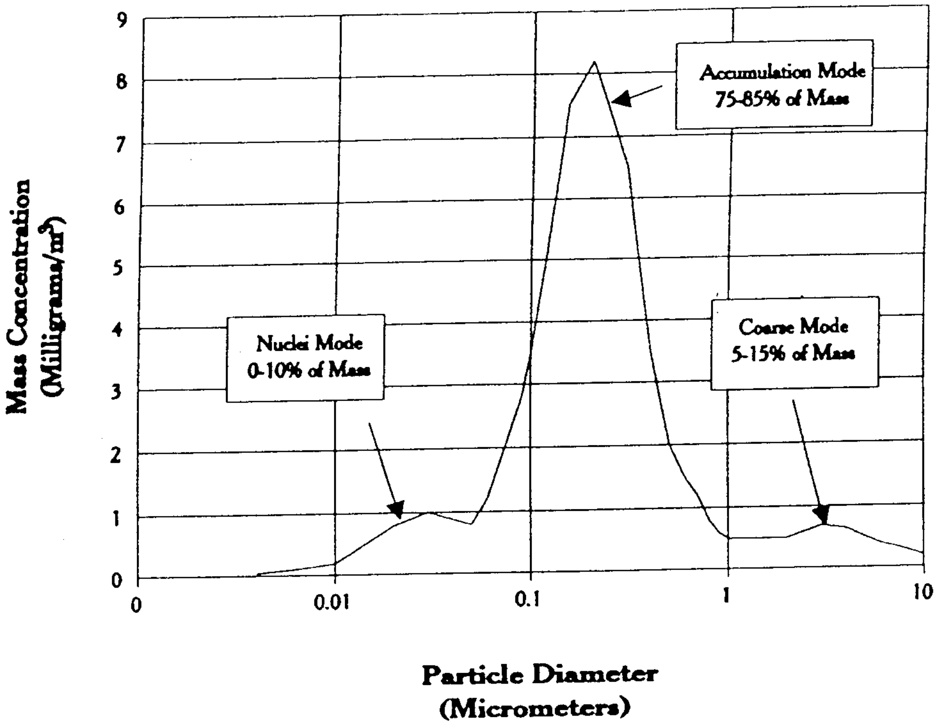


Figure 1. Particle size distribution for diesel particle emissions by mass.

EMISSION OF PARTICLES FROM SPARK-IGNITION ENGINES

Habibi (1970) pioneered studies of particle emissions from passenger cars fueled with leaded gasoline. In this work, air-dilution tunnel sampling was introduced for the first time to simulate the dilution processes immediately behind a moving automobile. Habibi found that the major emitted material was lead chlorobromide, accounting for about half the particulate mass and also for about half the lead in gasoline. The balance of the material consisted of organic compounds, lubricating oil, and heavy ends of gasoline primarily.

Black et al. (1985) and Dietzmann, Parness, and Bradow (1981) have reported similar results for heavy-duty trucks fueled with leaded gasoline. Generally the results for both passenger cars and trucks are complicated by retention of particulate material in engine and exhaust system deposits. Relatively low emissions rates of very fine particulate material are experienced with low mileage vehicles while at high mileage, very much higher emission rates are experienced primarily of coarse particulate material ablated from engine and exhaust deposits. For example, Black et al. (1985) have reported fine fraction (diameter < 2.0 μm) emissions ranging from 12 percent of the mass at high emissions to 85 percent of the mass at relatively low emission rates. At all emission rates, about half the emitted particles were organic. These issues are of less concern in the United States than was previously the case, since lead compounds are no longer used in gasoline formulations. However, older cars, which were previously fueled with leaded gasoline, are still emitting coarse particles which are deposited by roadsides; moreover, roadside dust is still heavily contaminated with lead as a result of previously deposited material and these substances are commonly entrained in fugitive dust.

With the advent of catalytic control systems for motor vehicles, particle emissions from SI engine passenger cars changed abruptly. Pierson et al. (1974), found that minor amounts of sulfur in gasoline could be oxidized to sulfuric acid by noble metal oxidation catalysts. While only about 2 percent of the sulfur was emitted as sulfate in noncatalyst vehicles, for oxidation catalyst equipped vehicles the emission rate was limited only by thermodynamics; the conversion rates typically were 20 to 30 percent of the fuel sulfur and could conceivably have been as high as 70 percent. For steady state high-speed driving, the emission rates could have been as high as 100 milligrams per mile (mg/mi) of sulfate as sulfuric acid.

An extensive industry-government cooperative program was conducted in the mid-1970's to further investigate this potential problem area. A final summary report of the findings of that program was published in 1979 as a special report to Congress which had been required by section 403G of the 1977 Clean Air Act (CAA) Amendments (Kawecki, Bradow, and Duffield, 1979). This report covered results from emissions measurements of 100 in-use passenger cars in California, where the more stringent emissions standards should have produced higher sulfuric acid emission rates and from about 100 in-use 49 state passenger cars tested by Gibbs and his group at New York State DEC. Emission rates were found to be about 10 mg/mi in California and considerably less than that in New York. Review of the dispersion models used to assess sulfuric acid exposure revealed considerable overprediction (factors of 3-4) by EPA guideline roadway models under parallel wind conditions. This combination of corrections produced estimates of maximum sulfuric acid exposure due to automotive sources in the range of a few tenths of a micrograms per cubic meter (μg/m^3), only a minor fraction of those actually experienced in city environments or of those considered safe.

Subsequent adoption of three-way technology reduced particle emissions from passenger cars still further (Lang et al., 1981) and similar adoption of this technology in new gasoline truck engines should eventually eliminate these sources of atmospheric particles. There have been, however, no follow-up studies to confirm these suppositions, nor are any planned as far as is currently known.

ON-ROAD MEASUREMENTS OF MOBILE SOURCE PARTICULATE EMISSIONS

On-road measurements of mobile source emissions are critical if one is to accurately determine the overall contribution from mobile sources. Basing estimates for use in emissions inventories on currently existing models fails since estimates used by both the EPA and State and local agencies for developing air pollution abatement strategies may be underestimated by as much as a factor of 2 or more for both hydrocarbons (HC) and carbon monoxide (CO) (Pierson et al., 1990). Since motor vehicles are the dominant source of these species and other pollutants (e.g., particulate matter) in many nonattainment areas, such large errors make determination of appropriate abatement strategies difficult and prone to error. In addition, current mobile source emission factor models were not originally designed to evaluate the impact of reformulated fuels, new technologies, and new control strategies on HC, CO, nitrogen oxides (NO_x), PM-10, and speciated organics.

There are two main sources of **primary** particulate emissions from on-road sources: tailpipe emissions and tire wear. Note, however, that secondary reactions can occur leading to particulate formation. The mixing of mobile source emissions with both each other and those from other sources followed by chemical transformation to yield ambient aerosols is a significant source of ambient particulate matter.

Results from Tunnel Studies

Roadway tunnels have been successfully employed for determining on-road emission rates of gaseous (Pierson et al., 1990, Gertler et al., 1991, Ingalls et al., 1989) and particulate species (Ingalls et al., 1989, Pierson and Brachaczek, 1983).

The calculation of emission rates is straightforward. The concentration at the tunnel exit, less the concentration already existing in the ambient air coming in the entrance, represents the emissions into a known volume of air from a known number of vehicles traversing the tunnel. Thus:

$$\frac{mg}{mi} = \frac{(C - C_o)V}{LN}$$

where: C = concentration (mg/m^3) at the exit portal
C_o = concentration (mg/m^3) in the intake or ambient air
V = air displacement (m^3) through the tunnel during the sampling period
N = vehicle count during the sampling period
L = tunnel length (mi)

Additional advantages of tunnel studies are that the formation of secondary particles is minimized due to the short residence time and lack of sunlight and the importance of resuspended road dust is minimized inside a tunnel.

Few studies have been performed which quantify particulate emissions in tunnels. The most comprehensive assessment of particulate matter associated with primary motor vehicle emissions was reported by Pierson and Brachaczek (1984). They reported results of experiments at the Allegheny and Tuscarora Mountain Tunnels of the Pennsylvania Turnpike in 1975-1979. Because of strong variations in vehicle mix (i.e., day/night and weekend/weekday) they were able to resolve emission rates into both light-duty gasoline and heavy-duty diesel categories. For Tuscarora in 1977 emissions rates were 51 ± 30 mg/kilometer (km) and 865 ± 161 mg/km for gasoline and diesel vehicles, respectively. For Allegheny in 1979 the results were quite similar with gasoline powered vehicles being statistically insignificant (6 ± 11 mg/km) and diesels reported as 860 ± 22 mg/km. Per vehicle-km diesels, which comprised 10-15 percent of the total vehicle fleet, were the major source of emissions. Overall, approximately 80 percent of the fine particulate matter mass observed was from carbon (elemental and organic), 5 percent from sulfate (due to the oxidation of S in diesel fuel), 10 percent from fuels and motor oils, 1 percent from tire wear, 10 percent from soil dust and 3.5 percent unknown. They concluded the particulate mass due to tire wear was not significant when compared with the remaining sources. This is consistent with an earlier paper in which they reported tire debris comprised 1 to 4 percent of the total particulate matter generated by vehicles (Pierson and Brachaczek, 1974).

Using observed Pb concentrations in the ambient air, Pierson and Brachazek (1984) further concluded the contribution of vehicle primary particulate emissions was approximately 11 percent in an urban area and 2 percent in a rural area in the Northeast.

As part of the Southern California Air Quality Study (SCAQS), Ingalls et al. (1989) measured on-road vehicle emission factors in the Van Nuys Tunnel. For the high speed (average 41 miles per hour [mi/h]) runs PM-10 emission factors ranged from 0.07 to 0.19 grams (g)/mi for all vehicles. The average ratio of measured vs. EMFAC7C emission factors was 1.7. As with CO and HC the on-road result was significantly higher than the model prediction. The average ratios of tunnel emissions factors to EMFAC7C emissions factors were: 1) 2.7 ± 0.7 for CO; 2) 4.0 ± 1.5 for HC; and 3) 1.02 ± 0.21 for NO_x (Pierson et al., 1990). These differences between measured and modeled emissions rates raised substantial concern regarding the validity of the *in-situ* measurement, the vehicle emissions modeling procedure, the model inputs, and the current vehicle emissions inventories, and automotive pollutant abatement strategies.

Gertler et al. (1991a,b) discussed a number of potential shortcomings in the models which may have led to the observed discrepancies between observations and model predictions. A number of liabilities or potential liabilities in the tunnel experiment were reviewed including: 1) biases in the chemical measurements; 2) inaccuracies in the tunnel flow measurements; 3) backflow of air at the tunnel exit; 4) correlations between tunnel flow and emissions; 5) calculational errors and uncertainties; 6) differences in vehicle operating conditions between model inputs and on-road conditions; 7) differences in vehicle distributions between model inputs and experimental values; and 8) representativeness of the samples taken. It was determined that none of these (or even all taken together) were sufficient to account for the observed differences. Furthermore, the impact of any experimental errors became irrelevant when one considers concentration ratios or emissions-rate ratios. These too, however, were found to differ by factors of close to 3 or more. The evaluation of the differences in the ratios of emitted species implied that the limitations in EMFAC and MOBILE owe less to deficiencies in the models than to limitations in the data base used to construct the emissions rate model input.

Based on review of the models and their development and comparison with historical data, they postulated a number of groupings of questions about the input assumptions. These included:

- Representativeness of the volunteer fleets for estimating maintenance/ deterioration.
- Representativeness of the volunteer fleets for estimating emitter level distributions.
- Treatment of engine load.

While these questions were primarily aimed at the differences in the HC and CO predictions, they also apply to estimates for particulate emissions. For example, one of the primary potential differences may lie in the fact that neither EMFAC nor MOBILE take into account the impact of grade on emissions. This effect is noted in a report by Schlaug and Carlin (1979) in which they mention unpublished results from a Belgium study showing diesel buses travelling at 40 km/h up a 6 percent grade produced 14.5 times more "smoke" relative to a 0 percent grade. Impacts of this magnitude cannot be ignored.

Ambient Air Studies

A number of studies have been performed to quantify the sources and impacts of ambient particulate matter. This has been especially true in the western United States where PM-2.5 has played an important role in visibility degradation. Studies have recently been completed in Denver, Reno, Phoenix, and Tucson with the objective of apportioning the sources of ambient PM-10 and PM-2.5.

Following the collection of ambient data, receptor models have proven to be the primary tool for apportioning the sources of ambient particular matter. The application of this methodology is discussed in chapter 3. Results of a number of studies apportioning the contribution of mobile sources to different particulate size fractions are described below.

Scheff et al. (1984) applied the chemical element balance receptor model (CEB, this is the same as the chemical mass balance [CMB] receptor model) to Chicago for a 7-month period from July, 1981 to January, 1982. Measurements were made for TSP and respirable particulates (RP - particles with diameters less than 2 μm). Mobile sources contributed 3.6 $\mu g/m^3$ out of a total TSP loading of 75.7 $\mu g/m^3$ or 4.8 percent. An additional 33.5 $\mu g/m^3$ or 44.3 percent was attributed to soil, much of which is due to resuspended road dust. In the size segregated fraction, mobile sources contributed 1.7 $\mu g/m^3$ out of a total loading of 36.7 $\mu g/m^3$ or 4.6 percent. Soil contributed 7.3 $\mu g/m^3$ or 19.9 percent.

Principal component analysis (PCA) was applied by Thurston and Spengler (1985) to metropolitan Boston over a 2-year period. Motor vehicles were found to contribute 14.4 percent (2.5 µg/m^3 out of 17.3 µg/m^3) of the fine (PM-2.5) particle mass and 7.0 percent (0.6 µg/m^3 out of 8.5 µg/m^3) of the coarse (2.5 µm to 15 µm) particle mass.

Watson and Chow have recently completed studies in Reno, Denver, Phoenix, and Tucson with the objective of apportioning the sources of ambient PM-10 and PM-2.5. Emphasis will be placed on the PM-10 results.

Chow et al. (1990), in a study completed in Reno, NV during the winter of 1986 to 1987, evaluated the causes of high PM-10 levels and the impact of street sweeping on the ambient particulate levels in the Reno area. Mobile sources contributed 44 percent of the PM-10 during the months of February and March. An additional 50 percent was attributed to geological material. Street sweeping was found to have no effect on the ambient loadings.

As part of the 1989 to 1990 Phoenix PM-10 Study, Chow et al. (1991a) concluded:

- PM-10 was generally higher at the West Phoenix and Central Phoenix sites than at the South Scottsdale site. Average PM-10 concentrations for the sampling period were 46 µg/m^3 at West Phoenix, 50 µg/m^3 at Central Phoenix, and 37 µg/m^3 at South Scottsdale. The Estrella Park nonurban site, which is normally downwind of the urban area, experienced average PM-10 similar to South Scottsdale at 37 µg/m^3.

- PM-10 was much lower at the nonurban sampling sites which are generally upwind of the urban area. Average PM-10 for the study period was 23 µg/m^3 at Gunnery Range and 15 µg/m^3 at Pinnacle Peak.

- The PM-10 was approximately evenly divided between PM-2.5 and coarse material at the urban sites. Coarse particle concentrations were approximately twice PM-2.5 concentrations at nonurban sites.

- Primary geological material was a major contributor to PM-10 at every sampling site. Geological material typically contributed from 40 to 60 percent of the PM-10 at the three urban sites on the days with highest concentrations. The spatial and temporal nature of the geological contributions at the urban sites indicates they are almost entirely resuspended road dust.

- Primary motor vehicle exhaust typically contributed from 30 to 50 percent of the PM-10 at the three urban sites on the days with highest concentrations. The source profiles which best explained the ambient measurements contained emissions which were at least 50 percent from diesel-fueled vehicles. This is consistent with estimates from vehicle counts, vehicle registration, and fuel consumption information taken during the study. No evidence of species from brake and clutch wear was found in the ambient data or in measurements taken beside a heavily traveled street. Given the chemical composition of these brake- and tire-wear emissions, they could not have contributed more than a few µg/m^3 to ambient PM-10 at any of the sites.

Similar observations were made in Tucson (Chow et al., 1991b) where primary geological material accounted for 50 percent of the PM-10 and primary motor vehicles accounted for 30 to 40 percent of the PM-10.

In Denver, during the winter of 1987 to 1988, Watson et al. (1989) found primary particulate emissions from motor vehicles accounted for 22 percent of the PM-2.5 mass. Of this 50 to 85 percent was attributed to diesel vehicles and cold transient catalyst vehicles, 5 to 30 percent from cold stabilized vehicles, and 15 to 20 percent from leaded vehicles.

SOURCES OF SECONDARY PARTICULATES

Particles derived from sources of secondary particulates include those derived from condensation of exhaust gases, chemical transformations of gases and particles emitted from tailpipes and from resuspension of material from the road surface. Far and away the largest source of secondary particulate emissions is the latter category. Particle emissions from resuspension processes are generated from two major sources: paved roadways and unpaved roads. Evaluation of the emissions of particles from these two sources is detailed in chapter 3 in the discussion of emission rates and emission models. The relative magnitude of emissions attributable to these sources is discussed in chapter 4.

CHAPTER 3: METHODS OF ASSESSING PARTICULATE IMPACTS FROM HIGHWAY SOURCES DISPERSION MODELS

BACKGROUND

There are many sources of airborne particulate matter that contribute to PM-10 loadings, including:

- Primary PM-10 emissions from highway segments.
- Primary PM-10 emissions from other mobile sources.
- Primary PM-10 emissions from major point sources.
- Primary PM-10 emissions from area sources.
- Secondary PM-10 formation from highway emissions.
- Secondary PM-10 formation from other mobile sources.
- Secondary PM-10 formation from point sources.
- Secondary PM-10 formation from other area sources.

Contributions of vehicle emissions from highways to PM-10 concentrations can occur at the microscale and regional scales of analysis. This is a complex problem, one which requires the consideration of direct vehicle emissions, as well as particles formed in the atmosphere from highway emissions. There also is a need to place total highway impacts (primary and secondary) in context by evaluating their importance relative to other source categories that contribute to PM-10.

Multiple modeling approaches may be needed to evaluate and assess the various contributions to highway vehicle impacts, because neither dispersion modeling nor receptor modeling alone can deal with microscale and regional scale analyses. Together, however, these two modeling techniques can effectively address these scales.

Dispersion modeling is well suited to evaluating current conditions, and the expected air quality benefits of alternative mitigation strategies to reduce "hot spot" and regional PM-10 incremental contributions from highway sources. Receptor modeling, on the other hand, can be used to estimate, for current conditions, the highway related PM-10 impacts (direct and secondary) relative to other source categories, including fugitive dust, and secondary particle formation.

This section presents a survey and evaluation of applicable dispersion modeling techniques that are appropriate to evaluate particulate impacts. The following factors are addressed: entrainment, suspension, transport, transformation, reactions, and deposition processes. A discussion also is provided concerning how well the reviewed models performed in previous studies. Dispersion models that may have particular importance to the Phase I, Task B and C efforts are also discussed.

SURVEY AND EVALUATION OF APPLICABLE DISPERSION MODELING METHODS

The scope of the survey and evaluation needs to be clearly specified. The objective of the dispersion modeling survey and evaluation was to characterize the ability of the models analyzed relative to their ability to evaluate the impacts of both primary and secondary particulate emissions from highway sources. The following helps define the scope of the dispersion modeling component:

- Only direct emissions from highway sources were evaluated in the dispersion modeling analyses. This includes tailpipe and resuspended particulates.

- Only highest and second highest 24-hour average PM-10 concentrations, and annual average PM-10 concentrations, were required to be modeled, since that is the current criteria for determining attainment of the PM-10 standards

- The capability of evaluating specific highway links, which would need to be evaluated in a microscale ("hot spot") analysis, were examined.

- The capability of the models to assess a full highway system for a regional-scale dispersion modeling analysis was also addressed.

There are numerous dispersion models that could have been evaluated. Pechan has focused its review on models that have been technically reviewed by the EPA, and listed in the Guideline on Air Quality Modeling (EPA, 1986). The advantage of this set of models is that: (1) the choice of models is broad enough to meet the objectives of this project, and (2) these models have undergone technical review that includes the evaluation of the logic of the algorithms, as well as the adequacy of the computer programs.

EPA's evaluation process of air quality models, before they are listed as preferred models (Appendix A of the Guideline), consists of a determination of technical merit in accordance with the following six requirements (EPA, 1986):

(1) The model must be written in common Fortran language and be suitable for use on a variety of computer systems.

(2) "The model must be documented in a user's guide which identifies the mathematics of the model, data requirements and program operating characteristics at a level of detail comparable to that available for currently recommended models..."

(3) "The model must be accompanied by a complete test data set including input parameters and output results."

(4) "The model must be useful to typical users, e.g., State air pollution control agencies, for specific air quality control problems."

(5) "The model documentation must include a comparison with air quality data or with other well-established analytical techniques."

(6) The model must be made available for public access (not proprietary).

Each model under review is subjected to a performance evaluation for an appropriate data base and to a peer scientific review. Those models that are found to perform better, in comparison to the results to the list of preferred models listed in Appendix A of the Guideline on Air Quality Modeling, are then considered for inclusion as preferred models in future guideline revisions. These preferred models "may be used without a formal demonstration of applicability as long as they are used as indicated in each model summary..." (EPA, 1986).

The EPA Guideline on Air Quality Models (the Guideline) makes general recommendations for mobile source models in chapter 6 of that publication (EPA, 1986). The following are some key aspects of the Guideline, as it relates to this study:

- The Guideline specifically mentions CALINE4 for CO, and indirectly recommends CALINE4 for nitrogen dioxide (NO_2) when it indicates to use an appropriate Appendix A model. An Appendix A model is a dispersion model that is preferred by EPA. Appendix B models are alternative dispersion models that can be considered on a case-by-case basis. CALINE4 is the only Appendix A model that is specific to mobile sources. While the Guideline does not specifically indicate a preferred model for evaluating particulates released by mobile sources, it appears that CALINE4 would be the preferred model for short-term impacts from specific roadway segments.

- The Guideline indicates that situations that require the use of refined techniques for urban-scale analysis should be considered on a case-by-case basis.

- Section 7.2.3 of the Guideline mentions the use of the ISC line source treatment when deposition is of interest, which is an issue for this project.

- Section 7.2.2 indicates that no dispersion model is recommended at this time for modeling secondary particulate formation.

The survey and evaluation of alternative modeling techniques for assessing microscale and regional scale impacts are described separately.

Microscale Dispersion Models Suitable for Short-Term Averaging Periods

Six dispersion models were reviewed to consider their applicability to this project. table 1 summarizes the pertinent features of the models reviewed:

Table 1. Summary of Dispersion Models Capabilities

Feature	CALINE-4	APRAC-3	FDM	HIWAY-2	GMLINE	ISCST
Gravitational Settling	*		*			*
Dry Deposition	*		*			
Wet Deposition						
Mech. Turb. from vehicles	*			*	*	*[1]
Therm. Turb. from vehicles	*				*	
Elevated Sections	*		*			*
Depressed Sections	*			*		
Urban-scale Features		*				*
Annual Averages			*			*

[1] Can be simulated through volume source treatment in ISCST.

CALINE-4 (Benson, 1984) is a steady-state Gaussian plume model that can be used to estimate concentrations of nonreactive pollutants emitted from highway traffic. The model is applicable for any wind direction, highway orientation, and receptor location for various highway situations such as elevated, depressed, and at grade sections for relatively simple (rolling) terrain regions. The model can also address gravitational settling velocity and dry deposition of particulates, which is important for this project. CALINE-4 does not calculate long-term (annual) average concentrations.

APRAC-3 (Simon et al., 1981) calculates hourly average concentrations for urban traffic situations including freeway, arterial, feeder street, and local street canyons. Particulates may not be addressed very well by this model since it does not contain gravitational settling and dry deposition features. APRAC-3 does not calculate long-term (annual) average concentrations.

The street canyon treatment in APRAC-3 is based on the street canyon/box model approach. A box model is a convenient, although very conservative, tool to provide general screening analyses for air pollutant concentrations over urban areas. As the name implies, this model assumes a three-dimensional shape, a box, is placed over the area of interest. Air pollutant emission levels are estimated for the "enclosed" area and, because the volume of the box is known, average pollutant concentrations within the urban area can be calculated. The assumptions that cause these models to be conservative include those related to emission levels and distribution, wind speed, wind direction, meteorology, existing air quality, and general dispersion characteristics. These assumptions, coupled with others to further simplify the numerical calculations, allow a straightforward, but highly simplified, initial calculation of pollutant concentrations.

The concept of urban areas as boxes for the purpose of pollutant concentration calculation is a widely used device in air quality modeling analyses. The principal is logically extended to smaller scale areas within cities such as the streets, where the buildings on either side of the streets create "canyons," as illustrated in figure 2. The calculation of air pollutant concentrations in these street canyons requires the addition of pollutant concentrations from sources within the canyon, motor vehicles, to the background pollutant concentrations from the urban area in general.

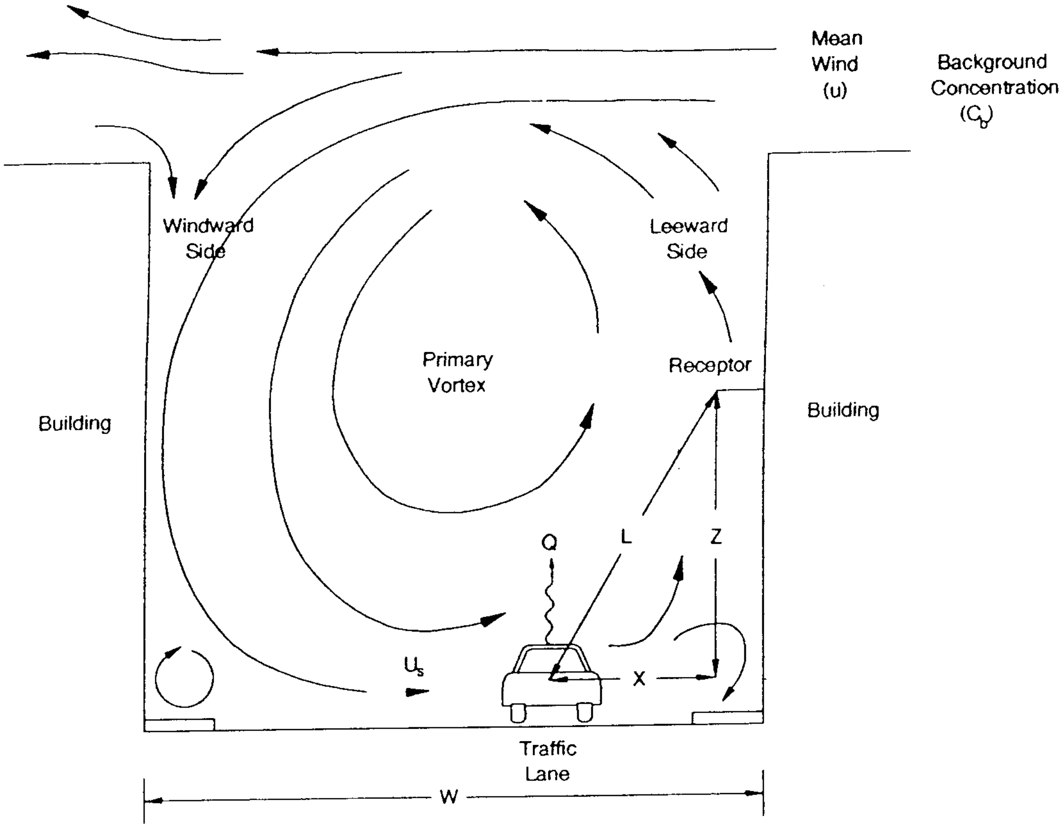


Figure 2. Schematic diagram for parameters included in the street canyon/box model.

The calculation of the effects of street-level sources on air pollution in urban areas is important for two reasons. First, these sources contribute significantly and immediately to the air pollutant concentrations experienced by pedestrians on the street. Second, because most urban monitoring stations are located close to these source areas, these source areas are major contributors to the monitored data. The effects from these source areas must be understood to verify and interpret correctly the monitored data.

The circulation pattern experienced within the street canyon, as indicated in figure 2, leads to two distinct concentrations that can be calculated - the concentration on the leeward side of the street, C_l, and the concentration on the windward side of the street, C_w - when the mean wind flow is perpendicular to the street. (The resulting wind flow patterns shown in figure 2 is a two-dimensional representation - in reality the three-dimensional wind flow pattern is more complex.) When the wind flow is not perpendicular to the street, an average of the two concentration equations is used and the resulting two- and three dimensional wind flow patterns are far more complex.

Research and field work in the early 1970's showed that even though the box model, as applied to street canyon situations, was very conservative and involved simplifying assumptions, there was a reasonable good fit between the predicted pollutant (CO) concentrations from the model and the observed pollutant concentrations from street-side monitors. Better correlation between predicted and observed concentrations occurred when the mean wind flows were perpendicular to the streets. This early work has been incorporated into APRAC-3.

The Fugitive Dust Model (FDM) (Winges, 1990) is specifically designed to calculate concentration and deposition impacts from fugitive dust sources. The model is based on the Gaussian plume formulation that has been specifically adapted to incorporate an improved gradient-transfer deposition algorithm. Gravitational settling and dry deposition of particulates are calculated for a series of particle size classes identified by the user. The model can address short-term (≤ 24 hours) and long-term (annual) averaging periods. The model does not address thermal turbulence factors associated with mobile source releases.

HIWAY-2 (Petersen, 1980) is another steady-state Gaussian plume model that can be used to estimate concentrations of nonreactive pollutants from highway traffic. It can account for mechanical turbulence generated by the vehicles on the roadway. Only short-term (≤ 24 hours) averaging periods can be addressed. Gravitational settling or dry deposition for particulates are not considered.

GMLINE (Chock, 1978) is a simple line-source model using the Gaussian plume algorithm to calculate downwind dispersion of pollutants near roadways. The model does account for thermal turbulence and plume rise due to heated exhaust as well as mechanical turbulence created the vehicles. The predicted impact is limited to the short-term (≤ 24 hours) time averages and does not contain gravitational settling or dry deposition considerations.

The Industrial Source Complex Model Short-Term (ISCST) (EPA, 1987a) is a steady-state Gaussian plume model that can be used to address pollutant concentrations from a wide variety of sources (area, line, and volume) associated with an industrial source complex. This model can account for gravitational settling of particulates. ISCST is a preferred model for regulatory applications in simple (rolling) terrain for rural or urban conditions. The model can be used for both short-term (≤ 24 hours) and long-term (annual) averaging periods.

CALINE4 is recommended for modeling microscale impacts from highways because of the three major reasons listed below:

1. The EPA Guideline on Air Quality Models (EPA, 1986) lists CALINE4 as a preferred model for mobile source impacts.
2. CALINE4 contains many desirable features for this specific project, such as a detailed treatment of the initial mixing zone in the immediate vicinity of the highway segments, consideration of thermal effects from vehicle exhaust on dispersion coefficients, the features to address at-grade, elevated and depressed highway segments. These features, and other model features that are specific to highway sources, are especially important for meeting the microscale goals of this project.
3. CALINE4 is reasonably well validated based on field studies (Cadle et al., 1976; Benson and Squires, 1979; Noll and Claggett, 1979; Rodes and Holland, 1979).

Other dispersion models could be used to evaluate microscale impacts from highway sources since CALINE4 is not the only viable alternative. In Pechan's judgment, however, CALINE4 could be used, in conjunction with representative model inputs, to effectively analyze highway vehicle impacts at this scale.

Microscale Models - Annual Average Periods

The most significant complication of the scope of work for dispersion modeling is the need to evaluate annual average impacts from highway sources. Mobile source models generally are best suited for short-term averaging periods (e.g., equal to, or less than, 24 hours). More general purpose dispersion models, such as the EPA models RAM, CDM, and ISCST, which can be used to estimate annual average concentrations, were not designed to directly simulate many of the special mobile source features that are contained in mobile source models, such as CALINE4. The primary challenge of utilizing dispersion modeling for analyses at this scale, therefore, would be to develop a modeling strategy that overcomes the limitations in either mobile source or urban-scale models. The modeling strategy would need to produce estimates of 24-hour and annual average contributions of highway sources to microscale ("hot spot" receptors) and regional scale PM-10 concentrations.

The preferred model for microscale analysis would be CALINE4, which can directly address peak 24-hour concentrations from the highway segments under review. As previously mentioned, however, CALINE4 cannot address annual average impacts. While annual average analyses could be represented by a series of model runs that conceivably could include each day of the year, such a brute force approach may not be practical. An alternative approach that could be used instead is one that: (1) matches the ISCST model formulation to be as consistent as possible with CALINE4, and (2) then uses ISCST to screen annual average impacts.

Regional Scale Models Suitable for Short-Term and Annual Averages

There are several dispersion models that commonly are used for regional-scale modeling applications (e.g., the EPA RAM [Turner and Novak, 1978] and CDM [Irwin et al., 1985] models). For most urban-scale model applications, it is important to address urban-scale area sources, such as from mobile sources, emissions from numerous small industrial facilities in a metropolitan area, dry cleaning emissions, emissions from degreasing, and so forth. The area source component, however, may not be as important in this case, because the focus in dispersion modeling will be on the highway system that is in place for the selected area. This simplification is important because the most versatile EPA model, the Industrial Source Complex Model/Short-Term Mode (i.e., ISCST), can be used to address the regional scale, as long as area source evaluations are not required. The main objectives when using ISCST in this manner are to: (1) preserve the geometry of the highway segments, and (2) to avoid excessive separation of the volume sources used to represent the highway segments — where the appropriate separation is a function of the distance from each highway segment to its' closest receptor.

The primary benefit of using ISCST is that it can simulate specific highway segments or the general highway system through a series of volume sources. Additionally, ISCST can be set up to simulate the mixed zone used in the CALINE4 model. This provides a means of estimating short-term and annual average concentrations at the microscale and regional scale.

As the most widely used dispersion model, ISCST has most likely undergone more user checkout than any alternative EPA preferred model. Although it has not been tested specifically for the use suggested here, ISCST has been tested and generally found to be an acceptable model (EPA, 1981). Since the ISCST modeling would be tailored to match as closely as possible the mobile source model, CALINE4, ISCST could be expected to adequately address microscale and regional scale impacts at the regional scale.

The following summarizes the features of the models most highly recommended for use in assessing highway particulate impacts (CALINE4 and ISCST), for key topic areas of this study:

Entrainment - Entrainment in the context of this study refers to the resuspension of particulate matter from the roadway surface into the ambient air. Neither CALINE4 nor ISCST have specific algorithms to directly compute entrainment rates, therefore, emission factors need to be used to make such estimates prior to executing these models. The rate of entrainment is a function of vehicles per hour, type of vehicle (i.e., wake size), vehicle speed, the state of the road surface (i.e., number of lanes, road surface condition), and other factors. This term will need to be simplified, however, into average entrainment rates. These emission rates could then be modeled through the line source treatment (with initial mixing volume) available through CALINE4, and the volume source treatment in ISCST.

Suspension - The suspension of particulate matter into the ambient air occurs through direct exhaust emissions and through reentrainment of particulate matter from the roadway surface. Once suspended, CALINE4 and ISCST account for the transport, dispersion, and gravitational settling of these emissions. With the exception of the treatment of gravitational settling, these models treat suspended particulate emissions in the same manner as gaseous pollutants. Impaction is not treated by either model.

Transformation - Particulate matter can be formed in the atmosphere from the exhaust of mobile sources (i.e., secondary particulate matter). Section 7.2.2 of the EPA Guideline on Air Quality Models (EPA, 1986) indicates that there is no EPA recommended dispersion model for modeling secondary particle formation. Neither CALINE4 nor ISCST can be used to model secondary particle formation. Utilization of receptor modeling would be the preferred method of accounting for secondary particles formed from gaseous pollutants emitted from highway sources.

Reactions - Reactions of gas-phase pollutants can produce secondary particulate matter. Refer to the above description concerning modeling of transformation processes.

Deposition - Deposition can be an important factor for estimating ambient concentration because deposition mechanisms can deplete pollutants from the atmosphere. Deposition can occur in three forms: (1) gravitational settling caused by the weight of the particles, (2) dry deposition, which deposits pollutants on the surface such as through the filtering action of the surface or surface reactions, and (3) wet deposition caused by precipitation. CALINE4 can be used to account for the first two terms, although there generally would be significant uncertainty in the nongravitational settling component. ISCST accounts for gravitational settling. Neither model accounts for wet deposition, which can be of equal magnitude with the first two terms. This simplification will introduce some conservatism into an analysis, which is likely to be of greater significance for annual averages than for maximum 24-hour periods.

INTERPRETATION OF MODEL PERFORMANCE FROM OTHER STUDIES

The following summarizes model performance evaluation results based on available data from other applications. While strictly speaking, model performance evaluations are only directly applicable to the locations and time periods tested, a series of successful tests enhances the confidence in a model. This section is subdivided into two components: (1) CALINE4 and (2) ISCST.

CALINE4

CALINE4 has undergone reasonably extensive evaluation relative to many other dispersion models. It has been tested for five data bases, three of which were highway segments, and two intersection data bases. Highway tests are focused on in this summary, because they are most applicable to the project objectives.

The three highway-related data bases used to evaluate CALINE4 are as follows (Benson and Squires, 1979; Cadle et al., 1976; Noll and Claggett, 1979):

- CALTRANS Highway 99 Study
- General Motors Sulfate Dispersion Experiment
- Illinois EPA Study

The General Motors and CALTRANS 99 studies used a tracer, sulfur hexafluoride, as the basis for model performance testing. This results in the emission rate being relatively well known, and provides a firmer basis to assess the physical characteristics of the model in comparison with tests based on emission factors. Even though a gaseous tracer was used in these tests, these evaluations are appropriate to assess the general physical aspects of the model. The gravitational settling term for solid-phase pollutants has been tested in other applications, including the testing of ISCST, which is described below.

Statistical tests were done to evaluate correlation and bias. The following summarizes the results of the regression analyses:

Test Data Set	**n**	**R^2**	**Slope**	**Int.**
General Motors	599	0.76	1.015	0.185
CALTRANS 99	163	0.26	0.54	0.31
Illinois EPA (N)	995	0.45	1.00	0.77
Illinois EPA (S)	195	0.61	0.86	1.46

The Illinois data set was gathered for 2 urban sites. These results show, based on relatively large data sets, that the CALINE4 would be expected to explain approximately half of the variance in the observed concentrations (the range was 26-76 percent). This is acceptable model performance relative to expected performance for alternative models. These data also show that in some locations, such as CALTRANS 99, that relative poor model performance can occur.

Based on these three data bases, CALINE4 was found to have ideal bias (i.e., the model was found to be slightly biased to overestimate concentrations relative to measured data). In each of these three data bases, CALINE4 was found to show 75 percent or more of the paired measured and modeled data points to be within a factor of 2. For the two studies with tracer data, the cases where the model was not within a factor of 2 showed the following bias (Benson, 1984):

- 12-15 percent of the paired data were above the factor of 2 envelope.
- 1-7 percent of the paired data were below the factor of 2 envelope.

While poorer correlation and bias may occur in actual applications because of substantially greater uncertainty in the emission term, the available testing of CALINE4 suggests that the physical terms in this model are generally acceptable to meet the objectives of this study.

ISCST

The ISCST dispersion model also has undergone extensive model performance testing and has generally been found to predict concentrations within a factor of 2. This discussion will concentrate on the gravitational settling option. The depletion effect of deposition could be considered in modeling analyses involving ISCST (and CALINE4).

The conclusion of a detailed evaluation of the ISC model (EPA, 1981) stated: "The gravitational settling/dry deposition option adds capabilities lacking in most current models, and the accuracy of this option for particulates with appreciable gravitational settling velocities appears to correspond to the approximate factor of 2 accuracy generally attributed to the results of short-term dispersion model calculations in the absence of complicating factors (AMS, 1978)."

The evaluation study of the gravitational settling option of the ISC model was based on comparison with three deposition experiments (Walker, 1965; Stewart, 1968; and Boyle et al., 1975). The Walker (1965) and Stewart (1968) experiments consisted of continuous releases (≤ 60 minutes) of solid glass spheres from elevated point sources (> 7 meters high) with no buoyancy and minimum momentum. The Boyle et al. (1975) experiment consisted of releases of oil spray from a low-flying aircraft and is not directly representative of this study.

Table 2 presents the results of the statistical regression analyses of the comparison of modeled to measured data for both the Walker and Stewart experiments.

Table 2. Results of the Walker and Stewart Experiments to Evaluate Gravitational Settling Option of ISC Model

Test Data Set	n	R^2	Slope	Int.	Bias*
Walker	12	0.45	0.31	3.69	0.71
Stewart	6	0.74	1.16	-0.01	1.12

* Ratio of predicted to observed

This again shows reasonable model performance relative to expected performance to other alternative models. In the specific case of the Walker experiment, the best overall model performance was observed when the mean layer wind speed (between emission height and surface) was used in the deposition calculations with the surface reflection coefficient set equal to zero (no reflection [i.e., all particles that come in contact with ground remain there]). In general, the ISC model tended to be slightly biased in underestimating the amount of deposition. The model performance for the Stewart experiment showed a slight overestimate of the deposition with better correlation compared to the performance found with the Walker data. However, the measured data set for the Stewart experiment was not as extensive as the Walker data set.

The above tests of the ISCST model were not specific to mobile sources, however, it would be expected that if the initial source dimensions of the highway sources could be adequately characterized, that model performance should be similar to the results shown in this section. The treatment of the initial source dimensions in ISCST would need to be matched as closely as possible to those of the CALINE4 model (see above), which has been specifically tested for mobile sources. Therefore, adequate model performance could be expected for the ISCST model treatments.

SOURCE-RECEPTOR MODELS

Aerosol Source Apportionment Using Receptor Models

The first part of this section describes commonly used receptor models for aerosol source apportionment. Uncertainties inherent in applications of the models and the application of receptor modeling to on-road sources is discussed in the remaining subsections.

Receptor Models for Apportioning Atmospheric Constituents to Sources

Both source-oriented dispersion models and receptor models have been applied to the apportionment of atmospheric constituents to sources. Current scientific beliefs are that both models are necessary, either operating independently (EPA, 1987b) to engender the reconciliation of model results, or operating in concert with each other (Chow, 1985; Watson et al., 1987b; Freeman et al., 1987) so that the strengths of one type of model can compensate for the weaknesses of another model.

This section is centered on the use of receptor models to apportion ambient particulate matter and will focus on solely these models. Nevertheless, the meteorological and day-specific emissions data bases generally acquired for these studies are well-suited for the application and verification of source-oriented models.

Receptor models use the chemical and physical characteristics of gases and particles measured at source and receptor to both identify the presence of and to quantify source contributions to the receptor. The particle characteristics must be such that: 1) they are present in different proportions in different source emissions; 2) these proportions remain relatively constant for each source type; and 3) changes in these proportions between source and receptor are negligible or can be approximated.

Types of commonly applied receptor models include: 1) CMB; 2) PCA (otherwise known as factor analysis); and 3) multiple linear regression (MLR). Extensive explanations of each of these models, operating separately and together, are given by Watson (1984), Chow (1985), Hopke (1985), and Watson et al. (1987b; 1987c). All of these receptor models have been applied in previous Denver source apportionment studies (Heisler et al., 1980a, 1980b; Lewis et al., 1986), and the source apportionments, on the average, have been similar.

The PCA, CMB, and MLR have been combined with a dispersion model in a PM-10 assessment package prepared for the California Air Resources Board (CARB [Watson et al., 1987b; Freeman et al., 1987]) which provides interfaces among data bases and modeling software. An extensive protocol for CMB model application and validation has been developed for PM-10 source assessment (Pace and Watson, 1987), and elements of that protocol need to be applied to each model used in the source apportionment process.

The following subsections examine the CMB, PCA, and MLR receptor models, their assumptions, their input data requirements, and their applicability to this study.

Chemical Mass Balance

The CMB is the fundamental receptor model, and the derivation of the PCA and MLR models from fundamental physical principles begins with the CMB. The chemical mass balance consists of a least squares solution to a set of linear equations which expresses each receptor concentration of a chemical species as a linear sum of products of source profile species and source contributions.

The source profile species (i.e., the fractional amount of the species in the emissions from each source type) and the receptor concentrations, with appropriate uncertainty estimates, serve as input data to the CMB model. The output consists of the amount contributed by each source type to each chemical species.

The model calculates values for the contributions from each source and the uncertainties of those values. Input data uncertainties are used both to weight the importance of input data values in the solution and to calculate the uncertainties of the source contributions.

The CMB model assumptions (Watson, 1979) are: 1) compositions of source emissions are constant over the period of ambient and source sampling; 2) chemical species do not react with each other (i.e., they add linearly); 3) all sources with a potential for significantly contributing to the receptor have been identified and have had their emissions characterized; 4) the number of sources or source categories is less than or equal to the number of species; 5) the source profiles are linearly independent of each other; and 6) measurement uncertainties are random, uncorrelated, and normally distributed.

The degree to which these assumptions are met in applications depends to a large extent on the characteristics which are determined at source and receptor.

Solutions to the CMB equations consist of: 1) a tracer solution; 2) a linear programming solution; 3) an ordinary weighted least squares solution with or without an intercept; 4) a ridge regression weighted least squares solution with or without an intercept; and 5) an effective variance least squares solution with or without an intercept. An estimate of the uncertainty associated with the source contributions is an integral part of several of these solution methods.

Weighted linear least squares solutions are preferable to the tracer and linear programming solutions because: 1) theoretically they yield the most likely solution to the CMB equations providing model assumptions are met; 2) they can make use of all available chemical measurements, not just the so-called tracer species; 3) they are capable of analytically estimating the uncertainty of the source contributions; and 4) there is, in practice, no such thing as a "tracer."

CMB software in current use applies the effective variance solution developed and tested by Watson et al. (1984) because this solution: 1) provides realistic estimates of the uncertainties of the source contributions (owing to its incorporation of both source profile and receptor data uncertainties); and 2) chemical species with higher precisions in both the source and receptor measurements are given greater influence than are species with lower precisions.

Watson (1979) observed that individual sources with similar source profiles, such as different soils and road dusts, would yield unreliable values if included in the same CMB. Henry (1982) proposed a quantitative method of identifying this interference between similar source compositions, which is known as "collinearity."

Henry's (1982) "singular value decomposition" defines an "estimable space into which resolvable sources should lie." The source types which do not fall into this estimable space are collinear, or too similar to be resolved from a combination of one or more of the source types which do lie within the estimable space. Henry (1984) further proposed that linear combinations of source contributions resulting from collinear source compositions would be more representative of the summed contributions of these sources. Analytical measures of collinearity, and Henry's linear combination method, are available in Version 6.0 of the CMB model, though they have not been documented for the general user community.

The CMB modeling procedure requires: 1) identification of the contributing sources types; 2) selection of chemical species to be included; 3) estimation of the fraction of each of the chemical species which is contained in each source type (i.e., the source compositions); 4) estimation of the uncertainty in both ambient concentrations and source compositions; and 5) solution of the CMB equations. Pace and Watson (1987) emphasize the need for evaluating the effects of deviations from model assumptions. Several studies have addressed the effect of these deviations.

Assumptions 1 through 6 stated above are fairly restrictive and they will never be totally complied with in actual practice. Fortunately, the CMB model can tolerate reasonable deviations from these assumptions, though these deviations increase the stated uncertainties of the source contribution estimates.

The CMB model has been subjected to a number of tests to determine its abilities to tolerate deviations from model assumptions (Watson, 1979; Gordon et al., 1981; Henry, 1982; Currie et al., 1984; Dzubay et al., 1984; Watson and Robinson, 1984; DeCesar et al., 1985, 1986; Watson et al., 1987c; Javitz and Watson, 1988; and Watson et al., 1988b). These studies all point to the same basic conclusions regarding deviations from the above-stated assumptions.

With regard to assumption 1, source compositions are known to vary substantially among sources, and even within a single source over an extended period of time. These variations are both systematic and random and are caused by three phenomena: 1) transformation and deposition between the emissions point and the receptor; 2) differences in fuel type and operating processes between similar sources or the same source in time; and 3) uncertainties or differences between the source profile measurement methods.

Evaluation studies have generally compared CMB results from several tests using randomly perturbed input data or substitutions of different source profiles for the same source type. The general conclusions drawn from these tests are: 1) the error in the estimated source contributions due to biases in all of the elements of a source profile is in direct proportion to the magnitude of the biases; and 2) for random errors, the magnitude of the source contribution errors decreases as the number of chemical species measured at sources and receptors increases.

With regard to the nonlinear summation of species, assumption 2, no studies have been performed to evaluate deviations from this assumption. While these deviations are generally assumed to be small, conversion of gases to particles and reactions between particles are not inherently linear processes. This assumption is especially applicable to the end products of photochemical reactions and their apportionment to the sources of the precursors. Further model evaluation is necessary to determine the tolerance of the CMB model to deviations from this assumption.

A major challenge to the application of the CMB is the identification of the primary contributing sources for inclusion in the model, assumption 3. Watson (1979) systematically increased the number of sources contributing to his simulated data from four to eight contributors while solving the CMB equations assuming only four sources. He also included more sources in the least squares solutions than those which were actually contributors, with the following results:

- Underestimating the number of sources had little effect on the calculated source contributions if the prominent species contributed by the missing source were excluded from the solution.
- When the number of sources was underestimated, and when prominent species of the omitted sources were included in the calculation of source contributions, the contributions of sources with properties in common with the omitted sources were overestimated.
- When source types actually present were excluded from the solution, ratios of calculated to measured concentrations were often outside of the 0.5 to 2.0 range, and the sum of the source contributions was much less than the total measured mass. The low calculated/measured ratios indicated which source compositions should be included.
- When the number of sources was overestimated, the sources not actually present yielded contributions less than their standard errors if their source profiles were significantly distinct from those of other sources. The over-specification of sources decreased the standard errors of the source contribution estimates.

Recent research suggests that assumption 3 should be restated to specify that source contributions above detection limits should be included in the CMB. At this time, however, it is not yet possible to determine the "detection limit" of a source contribution at a receptor since this is a complicated and unknown function of the other source contributions, the source composition uncertainties, and the uncertainties of the receptor measurements. Additional model testing is needed to define this "detection limit."

The linear independence of source compositions required by assumption 4 has become a subject of considerable interest since the publication of Henry's (1982) singular value decomposition (SVD) analysis. As previously noted, this analysis provides quantitative measures of collinearity and the sensitivity of CMB results to specific receptor concentrations, which can be calculated analytically in each application. Henry (1982) also proposed an optimal linear combination of source contributions that have been determined to be collinear.

Other "regression diagnostics" have been summarized by Belsley et al. (1980) and have been applied to the CMB by DeCesar et al. (1985; 1986). At this point, the exact values of these diagnostics that correspond to intolerable degrees of collinearity are unknown, though this is an area of active research.

Tests performed on simulated data with obviously collinear source compositions typically result in positive and negative values for the collinear source types. Unless the source compositions are nearly identical, the sum of these large positive and negative values very closely approximates the sum of the true contributions.

With most commonly measured species (e.g., ions, elements, and carbon) and source types (e.g., motor vehicle, geological, residual oil sea salt, steel production, wood burning, and various industrial processes), from five to seven sources are linearly independent of each other.

Gordon et al. (1981) found instabilities in the ordinary weighted least square solutions to the CMB equations when species presumed to be "unique" to a certain source type were removed from the solution. Using simulated data with known perturbations ranging from 0 to 20 percent, Watson (1979) found: "In the presence of likely uncertainties, sources such as urban dust and continental background dust cannot be adequately resolved by least squares fitting, even though their compositions are not identical. Several nearly unique ratios must exist for good separation." It is widely recognized at this time that a substantial amount of research is needed to minimize the effects of source composition collinearity in the CMB. The most important finding of current research is that no single species controls the source apportionment and that reducing variability in the source profile input data is more important than identifying a set of so-called "tracer" species.

With regard to assumption 5, it is very unlikely that the true number of individual sources contributing to receptor concentrations is much larger than the number of species that can be measured. It is therefore necessary to group sources into source types of similar compositions so that this assumption is met. For the most commonly measured species, meeting assumption 5 practically defines these groupings.

With respect to assumption 6, the randomness, normality, and the uncorrelated nature of measurement uncertainties, there are no results available from verification or evaluation studies. Every least squares solution to the CMB equations requires this assumption, as demonstrated by the derivation of Watson et al. (1984). In reality, very little is known about the distribution of errors for the source compositions and the ambient concentrations. If anything, the distribution probably follows a log-normal rather than a normal distribution. Ambient concentrations can never be negative, and a normal distribution allows for a substantial proportion of negative values while a log-normal distribution allows no negative values. For small errors (e.g., less than 20 percent), the actual distribution may not be important; but for large errors, it probably is important. A symmetric distribution becomes less probable as the coefficient of variation of the measurement increases. This is one of the most important assumptions of the solution method that requires testing.

Principal Component Analysis

PCA has been proposed and applied (Henry and Hidy, 1979, 1982; Lioy et al., 1982; Thurston, 1983; Wolff and Korsog, 1985; Malm, 1986; Chow, 1985; Chow and Spengler, 1986) as an objective method of removing the highly intercorrelated nature of variations in atmospheric measurements. The variability in an aerosol concentration which arises from the variability of a number of observables is apportioned among the various principal components.

The PCA receptor model is meant to classify variables into groups which can then be associated with factors which contribute to chemical concentrations measured at receptors. These factors can be identified as emissions sources, chemical interactions, or meteorological phenomena, depending on the data sets which have been submitted to the PCA. Many of these variables are indicative of more than one causative factor. By themselves, they cannot be used to distinguish the influence of one factor from another.

Several causative factors have been identified in air pollution studies. Photochemical components were found for Los Angeles, CA; New York, NY (Henry and Hidy, 1979); St. Louis, MO (Henry and Hidy, 1982); and Lewisburg, WV (Lioy et al., 1982). These photochemical factors were consistently associated with daily average and maximum ozone, maximum temperatures, and absolute humidity.

Relative humidity (RH) components were found for Los Angeles and were highly correlated with daily maximum and minimum RH measurements. Local source components were found for Salt Lake City and Los Angeles and were highly correlated with sulfur dioxide (SO_2) and the wind frequency distributions from various quadrants. Dispersion/stagnation components were found for St.Louis, Salt Lake City, and Lewisburg. The variables correlated with this factor were NO, NO_2, wind speed at midnight and noon, average wind speed, morning mixing height, maximum hourly precipitation, and average precipitation. Each of these factors resulted from the application of PCA to gaseous, particle chemistry, and meteorological measurements.

The PCA model consists of the following steps: 1) select variables and measurement cases to be included; 2) calculate the correlation coefficients between the selected variables; 3) calculate the eigenvectors and eigenvalues of the correlation matrix; 4) select eigenvectors to be retained; 5) rotate these eigenvectors into a more physically meaningful space; and 6) interpret the rotated vectors as causative factors (some of which may be source surrogates) based on the variables with which they are highly correlated.

The PCA model assumptions as applied to source apportionment include those of the CMB plus the following: 1) the case-to-case variability of actual source contributions is much larger than the variability due to other causes such as measurement uncertainty or changes in source profiles at the emissions point or during transport; 2) causes of variability that affect all sources equally (such as atmospheric dispersion) have much smaller effects than causes of variability for individual source types (such as wind direction or emission rate changes); 3) the number of cases is much larger than the number of variables in the PCA (this number has been empirically shown to be approximately 70); and 4) eigenvector rotations are physically meaningful.

The mathematical structure implied by this verbal procedure is explained by Henry and Hidy (1979) and has been derived from first principles through step 4 by Watson (1979). Henry (1986) has recently shown that known physical constraints do not yield a unique solution to step 5, though the mathematical rotations in common use yield empirically logical and comparable results in most cases. Henry and Kim (1988) have derived a method which may result in a reduction in range of possible PCA solutions and a quantitative estimate of model uncertainty.

When PCA models are used for source apportionment, the input data consists of chemical species concentrations measured at a receptor for a time series. The model output consists of the number of contributing sources, the chemical characteristics associated with those sources, and the degree to which those sources are influential for each sample in the time series. The elements of each eigenvector can be converted to source profiles with certain assumptions (Henry, 1977b) which are not likely to be met in practical situations.

The only method of propagating uncertainties consists of randomizing model input data in proportion to measurement uncertainties and determining the standard deviation of source profiles derived from many model applications to the same randomized input data. This process is very tedious and is seldom performed in practice. Malinowski (1977a; 1977b) proposed a theory of error propagation in factor analysis, but it has never been verified in practical applications nor used in air quality PCA models.

PCA can be applied in an infinite number of variations, and because a sound physical basis for selection of factors and rotation methods is lacking, it seems at times that: 1) the conclusions drawn from PCA are the same as those which would be derived from an examination of the input data by themselves; and 2) the solution which fits preconceived ideas is generally selected while those solutions which do not are ignored. Only a few objective tests of PCA applied to air quality data have been performed.

Watson et al. (1987c) and Javitz et al. (1988) have examined real and simulated data with respect to selection of variables, randomly selected subsets, different factor selection criteria, different rotations, correlation among variables, and random variability. The generalized findings of these tests are presented below.

All available air quality variables should not be submitted to the PCA with the assumption that they will automatically migrate to the appropriate mathematical factor. There are several reasons why it may be wiser to limit the number of variables: 1) very highly correlated variables (greater than r= 0.98) can produce a singular correlation matrix; 2) highly correlated (r= 0.8 to r= 0.98) variables of similar quantities are redundant and may result in mathematical factors which are not related to physical factors; and 3) if one causative factor is represented by many more variables than another factor, the factor associated with the greater number of variables will always appear to explain a greater amount of the total variance. The "amount of variance explained" which is usually ranked to determine the "most important factors" is totally dependent on the variables included in the PCA.

Since PCA operates on complete data sets, a larger number of variables increases the likelihood that one or more measurements will be missing and cases will be eliminated. The statistical reliability of the eigenvectors, eigenvalues, factor loadings, factor scores, and regression coefficients decreases with the difference between the number of cases and the number of variables. Henry et al. (1984) recommend that the number of cases exceed half the number of variables included in the PCA by at least 32. PCA performed on random numbers by Preisendorfer (1977) indicates that in excess of 100 cases are required for 10 to 15 variables in order to obtain eigenvalues typical of random variations.

Other results of PCA model testing are:

- Mathematical factors derived from PCA are not necessarily associated with physical factors. The selection of variables is the most important determinant of the factors derived from PCA.
- A minimum of 75 to 100 cases is required to extract clearly defined and reliable mathematical factors that can clearly be associated with physical factors. These cases must include a variety of intensities in physical factors.
- An association of mathematical factors with physical factors provides a better method of selecting the number of eigenvectors for rotation than nonempirical mathematical criteria.

Javitz et al. (1988) applied PCA to simulated data sets that introduced deviations from assumptions with respect to source contribution variability, source profile uncertainty, and correlation among source contributions. They found that even when all uncertainties and intersource correlations were equal to zero, the usual criterion for selecting the number of sources (i.e., the number of eigenvectors with eigenvalues greater than one) resulted in three instead of the actual four sources. Intersource correlations of up to 0.9 could be tolerated by the model, however.

Javitz et al. (1988) also showed that when all assumptions were met, the calculated factor loadings could indeed approximate true source profiles, though they were unable to reproduce them exactly. With higher uncertainties associated with the source profiles used to generate the simulated data, however, the PCA did not adequately identify the correct number or the correct chemical character of the source types known to be present.

Ito et al. (1986) introduced random errors of up to 30 percent of their model input data and determined that while the factor loadings shifted somewhat, the selection of independent predictor variables for the MLR model was unaffected. They also selected subsets of data from their original 138 cases. While the 70-case subsets did not result in significantly different factors, qualitatively different solutions were observed in the 35-case subset. This finding is in agreement with those of Henry (1984) and Watson et al. (1988b).

PCA models have been widely used to study visibility and chemical constituents found in pristine areas of the western United States which include the Denver metropolitan area. Ashbaugh et al. (1984), Flocchini et al. (1981c), Pitchford et al. (1981) and Malm et al. (1985) applied PCA to various subsets of the Western Fine Particle Network (WFPN) and the Visibility Investigative Experiment in the West (VIEW) fine particle data bases.

Flocchini et al. (1981a; 1981b; 1981c) calculated principal components for elemental species measured at 40 sites located in eight western states for the period of October 1979 to May 1980 and found factors for soil, fine particle sulfur, smelters, and motor vehicle exhaust. Pitchford et al. (1981) combined fine particle chemical concentration data with meteorological and visibility observables at five sites distributed around the Four Corners area. Fine particle sulfur, coarse particle mass, and visibility reduction were associated with the same factor.

Ashbaugh et al. (1984) applied PCA to the spatially distributed sulfur concentrations at the 40 WFPN sites for a period of September 1979 to September 1980. They identified one spatial factor including all sites in the southern part of the network, which they attributed to copper smelter emissions in Arizona and New Mexico. The second factor included sites in the Northern Great Plains and was attributed to episodic incursions of sulfur from the East. The third, fourth and fifth factors were attributed to locally important conditions.

Malm et al. (1985) performed PCA on WFPN data taken from October 1979 to August 1981 at the Grand Canyon; the extinction coefficient, sulfur mass, soil mass, fine copper, fine zinc, and other fine mass concentrations were included in the analysis. They also included the residence time of air masses over eight geographical zones in the Southwest calculated with the ATAD trajectory model (Heffter, 1983). The first factor was found to be associated with air masses from the north or south; those from the south correlated most highly with visibility reduction. The second factor contained sulfur and air masses from southern Arizona and the Mojave Desert. The third factor contained significant loadings on visibility extinction, sulfur, soil, zinc, and air masses from southern California. The fourth factor was associated with air masses from other zones, and factor five was identified as a smelter factor.

While somewhat consistent in their findings, the results of the PCA applications in the western United States are inferential rather than causal, and other interpretations of the same PCA output can be drawn. PCA has always been applied as an afterthought to existing data, and the necessary receptor measurements to differentiate among source regions, major point sources, and other causes of co-variability have not been defined in most areas.

Watson et al. (1988b) recommend PCA applied to chemical and meteorological data to help identify source contributions as a first step in a CMB source apportionment. As noted by Lewis et al. (1986) in the source apportionment of Denver fine and coarse particulate matter measured in 1982, PCA has not yet been highly enough developed to allow the direct apportionment of fine particles to sources.

In spite of these recognized limitations, PCA serves a useful function in that it might identify associations which help to select source types and chemical species for inclusion in the CMB. Chow (1985), for example, noticed that SO_2 was loaded on the soil factor in Portage, WI. She derived a source profile for a nearby coal-fired power plant which contained SO_2 as one of its components, and this allowed her to apportion a fraction of that power plant's emissions to fine particles in that area. Recent applications of CARB's PM-10 assessment package show associations among elements (especially zinc from tire and brake wear) which indicate that road dust, rather than other forms of geological material, may be major contributors. This information can be used to select an appropriate source profile for inclusion in the CMB source apportionment.

Multiple Linear Regression

MLR applied to the apportionment of sources expresses a dependent variable (usually fine particle aerosol mass or a secondary particulate species such as sulfate or nitrates) as a linear sum of products of influence coefficients and source contribution surrogates. The source contribution surrogate can be a chemical species which is present only in the emissions from a source (including an injected tracer species) (e.g., Kleinman et al., 1980; Iyer et al., 1987), a source contribution calculated from the CMB, or principal components derived from a time series of chemical measurements taken at a receptor (e.g., Bowen et al., 1986; Watson et al., 1988b). Lewis et al. (1986), for example, applied this model in Denver using fine mass concentration as the dependent variable. The independent variables were nitrate and sulfate concentrations, silicon concentrations to represent soil, lead concentrations to represent motor vehicle exhaust, and noncrustal potassium to represent wood combustion.

The dependent variables and the source surrogates, with their uncertainties, determined for a time series at a receptor, serve as input data to the MLR. The influence coefficients are calculated by the model. The product of the influence coefficient and the source surrogate for an individual sample yields the contribution of the corresponding source to the dependent variable. The uncertainty of model outputs can be calculated analytically from uncertainties associated with the model input data (Watson et al., 1984).

The assumptions of the MLR models are the same as those of the CMB and PCA models with the addition of the following: 1) independent variables (e.g., source surrogates) are uncorrelated with one another; and 2) the independent variables (often termed "tracers") originate only in a single source type.

The MLR model has undergone very little testing. Watson et al. (1987c) showed with simulated data that the model provides a good source apportionment when the species used to represent a source is indeed a unique "tracer" and when its variability is low with respect to other chemical species and fine particle mass in the source profile. Javitz and Watson (unpublished simulation results) have shown that if there is substantial variability of the tracer species in the source profile, then the source apportionment is significantly worse than that of the CMB. This is because the MLR solution is similar to the tracer solution to the CMB, and any bias in the tracer is translated directly to the source apportionment. In the least squares CMB solution, random deviations in a positive direction by one species are offset by similar deviations in the opposite direction by another species. This tends to lower the overall uncertainty of the apportionment (Watson, 1979). Javitz and Watson also found that when the species used as "tracers" were not totally unique to a source profile, the MLR did not reproduce the source contributions which were used to generate the simulated data.

Ito et al. (1986) examined the effects of sample size and random error on the stability of source apportionment to particle mass concentrations using the MLR method. They found that the effect of a reduction in sample size had a significant effect on the stability of the calculated regression coefficients. Random errors on the input data of up to 30 percent did not appreciably change the source apportionment when the full data set of 138 cases was used. When less than 70 cases were included, however, the reproduction of true source contributions was much less reliable. Many cases are needed for the MLR to function effectively.

Uncertainty and Validity Analysis

The CMB receptor model has proven to be the one generally applied to particulate source apportionment; however for any given study, it is not possible to determine a priori whether or not a set of source profiles can resolve contributions from different sources. This "resolvability" is a function of: 1) the degree to which species are unique to certain sources; 2) the quantity of each species in each source profile; 3) the variability of each species in the source profile; and 4) the relative contributions from different mixtures of source types. The CMB software returns performance measures which identify potential difficulties in the resolution of source contributions from one another. These measures are only useful after source and receptor measurements are taken, however. Recent studies such as RESOLVE (Trijonis et al., 1988) and the Western States Acid Rain (WESTAR) project (Watson et al., 1987c) have made a priori estimates of feasibility using the methods developed by Watson (1979) and Javitz and Watson (1988).

Four types of uncertainty and validity tests have been found useful:

- Basic tests include running the CMB model with equal average source strengths for up to nine source types and introducing various uncertainty levels for source profiles and receptor concentrations. Input data uncertainties are related to uncertainties of the source contribution estimates.
- Collinearity tests involve simulations with source profiles containing similar chemical species. These can be actual source profiles (e.g., different types of soils) or ones which have been constructed from linear combinations of other sources in the CMB solution. Actual or expected variabilities in source profiles are used in these tests. Unique species can be added to source profiles at different concentration and uncertainty levels to determine the point at which the collinearity is minimized.
- Detection limit and dominance tests consist of simulations in which one or more sources have average source strengths which are significantly lower than those of other sources. This is the only way to verify the minimum source contribution which can be resolved by the CMB in the presence of other sources. Many source profiles will be determined to be collinear with other profiles by Version 6 software when, in reality, their contributions are so small that their species concentrations are within the variability caused by more dominant source contributions.
- Mis-specification tests address systematic biases in the source compositions. One or more elements in a source profile are changed with respect to the solution source profiles. These differences are increased until the differences between calculated and true source contributions exceed the calculated uncertainties of the source contribution estimates.

Application of Receptor Models to Apportionment of On-Road Contributions

As described above, applying receptor models to source apportionment requires information about the chemical characteristics of the emissions sources which are likely to affect pollutant concentrations at a receptor. For the CMB receptor model the required information is the fractional composition of each chemical species in the source emissions and an estimate of the variability of those compositions.

In a study to evaluate the feasibility of source apportionment of utility emissions, Javitz and Watson (1988) concluded that the major weakness of all receptor models is inadequate source composition data. They identified the following deficiencies in currently available source profiles: 1) the species measured are more often those which are convenient rather than those which differentiate among sources; 2) the types of species and size fractions measured are not the same for different source types and are not equivalent to the types of measurements made at receptors; 3) measurement methods are nonstandard and do not give equivalent results for the same species; 4) source characteristics, fuels, and operating parameters are inadequately documented; 5) data are of poor or unknown quality; 6) source profile uncertainties are not reported; 7) source samples are not representative of source profiles as they appear at the receptor; and 8) data are not available in formats which can be conveniently interfaced to modeling software. Javitz and Watson (1988) recommended the development of a standardized approach to sampling and analyzing particulate and gaseous emissions which would minimize these concerns with respect to future source profile measurements and this approach has generally been adopted.

Source Type

The potential source types which contribute to primary particulates in the a mobile source dominated area (i.e., an area without major industrial sources) are: 1) geological material (fugitive dust from agricultural tilling, paved and unpaved roads, vacant land and construction; 2) motor vehicle exhaust (from cars, trucks, and buses fueled with diesel, leaded gasoline, and unleaded gasoline); 3) vegetative burning (forest fires, prescribed fires, residential fireplaces, and residential wood stoves); 4) background aerosol (primary and secondary particles which advect into the airshed from outside the urbanized area); and 5) secondary particles. (Note that fugitive dust from paved and unpaved roads is lumped together under geological material.)

A "source type" does not necessarily correspond to a particular emitter. For the primary contributors, several source sub-types may exist within some of these categories which are not easily distinguishable from each other by receptor models using commonly measured chemical species. The source type perceived at a receptor may be a mixture of these different sub-types, and the profile which best explains the receptor measurement should represent this mixture.

One problem which arises with the apportionment of resuspended geological material from roads is the similarity in composition with that of geological material. Because of this similarity, profile mixtures, or "composites" are constructed for use in modeling. Among geological material, attempts may be made to distinguish between agricultural fugitive dust, resuspension from vacant areas, unpaved road dust, and paved road dust. As described by Chow et al. (1991) an attempt can also be made to distinguish between gasoline and diesel-fueled vehicles.

Geological Source Profiles

Examples of geological source profiles for Phoenix are presented in figure 3. In each panel of this figure, the height of each bar indicates the percent of the corresponding chemical species as emitted coarse particles. The percent of mass emissions indicated by this bar height is the average of the fractional compositions for the samples included in the composite. The position of the star shows the variability in the percent composition, which is estimated as the standard deviation of the fractional compositions or the analytical precision, whichever is greater. For example, silicon (Si) in the PHPVRD paved road dust profile has a bar height of 15 percent and a star height of 5 percent. This means that the fractional composition of silicon in paved road dust is 15+5 percent.

When the percent composition for a species is several times larger than its uncertainty, then that species is a good marker for that source type. When few other source profiles contain this species, then it is very probable that the source types which these profiles represent can be distinguished from each other by CMB modeling.

In each geological profile presented in figure 3, organic carbon (OC), aluminum (Al), Si, potassium (K), calcium (Ca), titanium (Ti), and iron (Fe) are present in abundances which exceed 1 percent. Al, Si, K, Ti, and Fe are present in all of the geological sub-types in the same abundance, within stated uncertainty intervals. The measurement of these species in ambient samples allows the geological material source type to be distinguished from other source types, but it does not allow the subtypes to be separated from each other.

The abundance of total potassium (K) is approximately 10 times that of soluble potassium (K+) in all of the geological profiles. This is an important difference, since soluble potassium is one of the key markers for vegetative burning. It is partly because soluble potassium is not very abundant in geological material that vegetative burning can be distinguished from other sources.

There are some significant differences among the geological source subtypes which might allow their contributions to be separated from each other in receptor samples. Dust on a paved road surface builds up by being tracked out from unpaved areas such as construction sites, unpaved roads, parking lots, and shoulders; by spills from trucks carrying dirt and other particulate materials; by transport of dirt collected on vehicle undercarriages; by wear of vehicle components such as tires, brakes, clutches, and exhaust system components; by wear of the pavement surface; by deposition of suspended particles from many emissions sources; and by water and wind erosion from adjacent areas. Axetell and Zell (1977) estimated typical deposition rates of 240 lb/mile of curb/day for particles of all sizes from the following sources: 1) 42 percent from mud and dirt carryout; 2) 17 percent from litter; 3) 8 percent from biological debris; 4) 8 percent from ice control compounds (in areas with cold winters); 5) 8 percent from erosion of shoulders and adjacent areas; 6) 7 percent from motor vehicles; 7) 4 percent from atmospheric dustfall; 8) 4 percent from pavement wear; and 9) less than 1 percent from spills. These proportions are highly uncertain because they apply to the TSP size fraction and because these investigators did not consider all of the sources cited above.

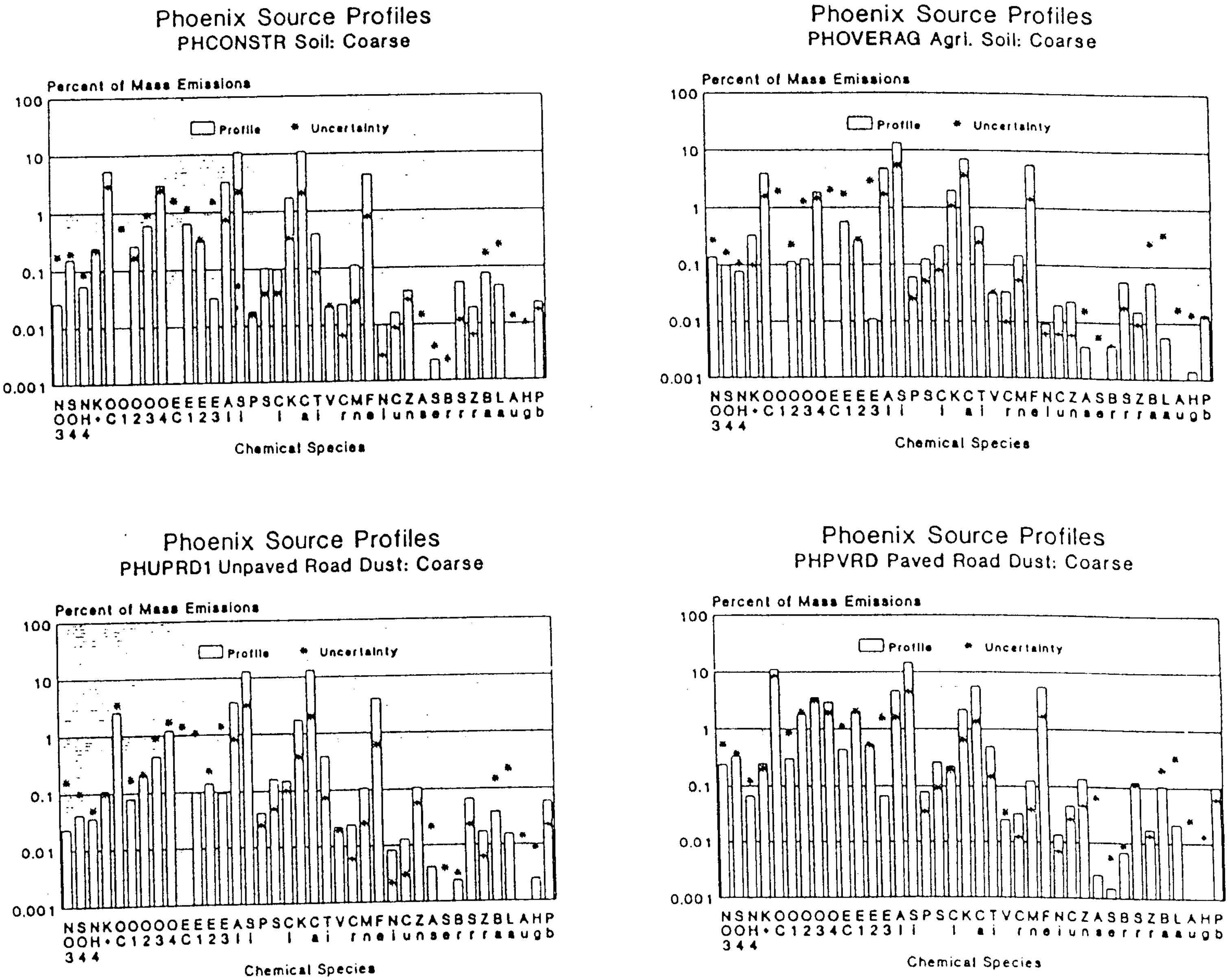


Figure 3. Geological source profiles for Phoenix.

Paved road dust is, therefore, much like an ambient PM-10 sample. It is a complex combination of particulate matter from a wide variety of sources, especially other geological source types. This complexity is evident in the comparison of the paved road dust profile (PHPVRD) with the other coarse particle geological profiles in figure 3. The first feature is the abundance of OC, which at 11±9 percent is larger and more variable than its abundance in the other profiles. The presence of tire wear, detritus, and engine oils can account for this enrichment. This OC content also places an effective upper limit on the contribution from tire wear to suspendable paved road dust, since OC is the major component of tires, with approximately 25 percent being a single compound, styrene-butadiene rubber (SBR) (Pierson and Brachaczek, 1974). Ondov (1974) measured elemental components of tire material and found minor concentrations for most species, with sulfur (S, ~2 percent), chlorine (Cl, ~1.5 percent), and zinc (Zn, ~1 percent) being the most abundant components.

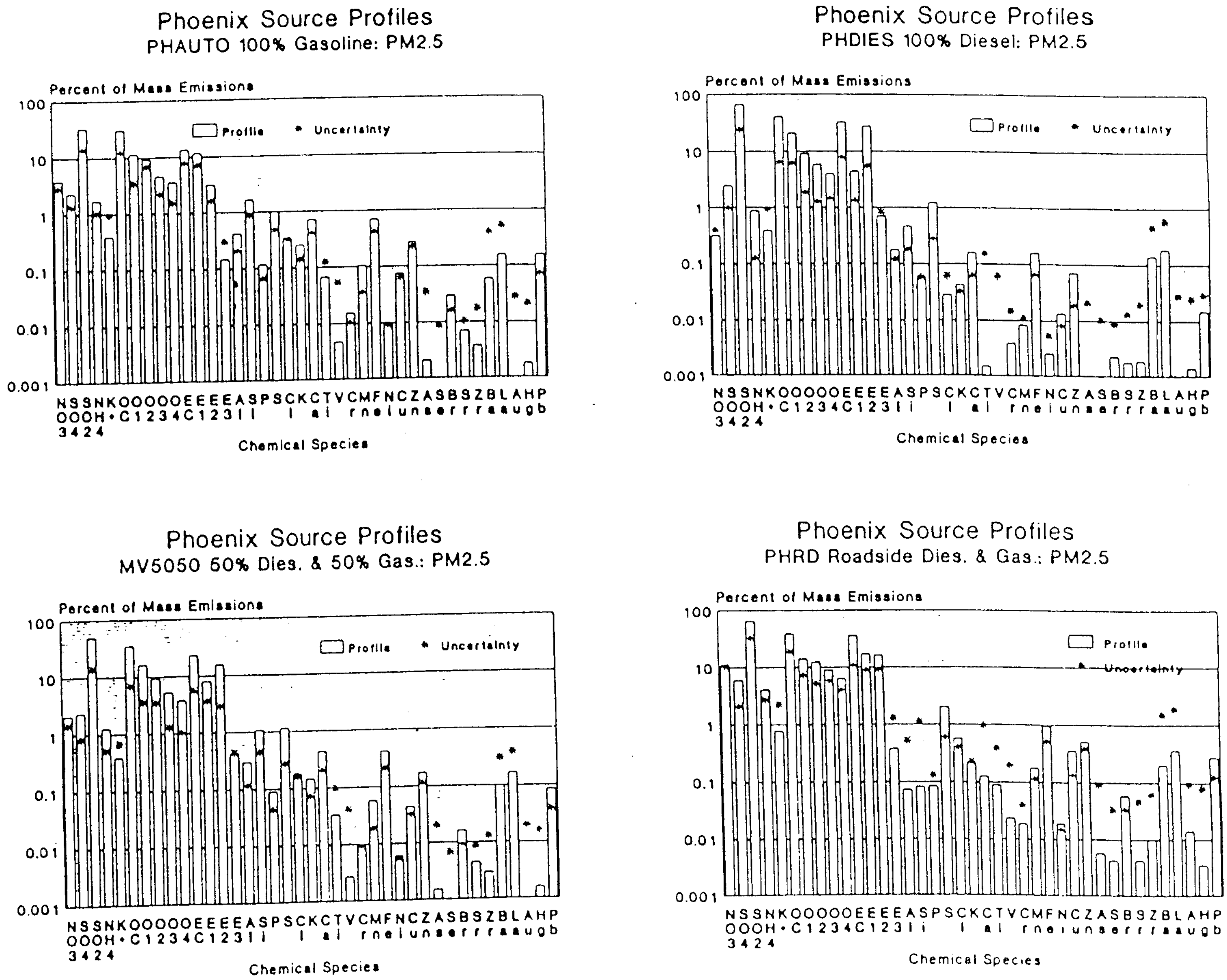
The abundances of lead (Pb) and bromine (Br) in paved road dust are also more than double the concentrations in the other geological profiles, which demonstrates the presence of tailpipe exhaust from vehicles burning leaded fuels. Enrichments in species from clutch and brake wear are absent in the paved road dust profiles. These are often composed of asbestos and/or semi-metal carbon composites. Ondov (1974) measured abundances of ~14 percent magnesium (Mg), ~2 percent Ca, ~4 percent Fe, and ~1 percent barium (Ba) in asbestos brake shoes, while Anderson et al. (1973) reported Si abundances of ~10 percent. Cooper et al. (1987) examined the elemental composition of semi-metal brake shoes and found abundances of ~45 percent iron (Fe), ~2 percent copper (Cu), ~0.5 percent tin (Sn), ~3 percent Ba, and ~0.5 percent molybdenum (Mo). None of these species is found in the Phoenix paved road dust profiles at levels which are significantly in excess of their abundances in the other geological sub-types.

The unpaved road dust profile in figure 3 contains OC at an abundance similar to the other unpaved geological sub-types, which is less than half of the abundance in the paved road profile. There is evidence of deposition from leaded-gasoline motor vehicle exhaust in the enrichment of lead and bromine with respect to the nonroad profiles. The notable feature of unpaved road dust profiles is the large abundance of calcium (~13 percent). Indeed, Barnard et al. (1986) showed that unpaved roads may have a calcium chemistry that can be as high as 20-30 percent by weight. In addition, Barnard et al. (1986) suggested that many of the limestone profiles identified in previous receptor modeling studies may actually represent emissions of dust from unpaved roads especially in areas where unpaved roads are known to utilize limestone aggregate as a surfacing material.

Motor Vehicle Exhaust Profiles

Composite source profiles for receptor modeling can be constructed from either dynamometer samples, inspection and maintenance (I/M) sampling, or roadside samples. The dynamometer and I/M samples are grouped into two categories: 1) diesel-fueled vehicles; and 2) gasoline-fueled vehicles.

Figure 4 shows profiles developed for Phoenix from I/M samples. In the PHAUTO example, the most abundant (>1 percent composition) species are OC (1.5+1 percent). There is substantial variability among the individual samples, as indicated by the large standard deviations associated with each abundance. The mass of SO_2 emitted by



Phoenix Source Profiles
PHAUTO 100% Gasoline: PM2.5

Phoenix Source Profiles
PHDIES 100% Diesel: PM2.5

Phoenix Source Profiles
MV5050 50% Dies. & 50% Gas.: PM2.5

Phoenix Source Profiles
PHRD Roadside Dies. & Gas.: PM2.5

Figure 4. Motor vehicle source profiles for Phoenix.

gasoline-fueled vehicles is approximately one-third of the fine particle mass emissions. The most abundant components in the diesel emissions are the same as those of the gasoline-fueled emissions, but in different proportions of fine particle mass. The OC abundance is 40+7 percent for diesel exhaust and much less variable than it is for the PHAUTO profile. The EC abundance in PHDIES is twice that of the PHAUTO profile. Other striking differences are that the 01 abundance in PHDIES is twice that in PHAUTO, the El fraction in PHAUTO is twice that in PHDIES, and the E2 fraction in PHDIES is nearly 10 times its abundance in PHAUTO.

For profiles created from the roadside samples, the samples may be affected by vehicle-related resuspended road dust and from a small quantity of pollutants from the surrounding air. Contributions to the mass and chemical species concentrations can be estimated and the geological contribution subtracted out.

EMISSION RATES/EMISSION MODELS

Particle Emission Rates

The official emission rate estimates of heavy duty diesel particulate matter were published by EPA in the Notice of Proposed Rule Making for Heavy Duty NO_x and Particulate Matter, initially for 1987 and later engines and finally for 1990 and later engines (EPA, 1984). This document contained a conversion factor for engine dynamometer grams per brake horsepower-hour (g/bhp/h) emission rates to roadway g/mi emission rates. Average emission rates listed are given in table 3.

Table 3. OMS Emission Factors for Heavy Duty Diesel Engines

Uncontrolled Vehicles	0.70 gm/bhp/h	2.18 g/mi
1987 controlled level	0.60 gm/bhp/h	1.87 g/mi
1990 controlled level	0.25 gm/bhp/h	0.90 g/mi

Later, the Office of Mobile Sources developed an emissions factor estimation computer program based on Mobile 2 which produced overall heavy duty emissions factors in the range 1.7 to 1.9 g/mi for vehicle speeds from 25 to 35 mi/h (EEA, 1985). This computer program produced some irrational emission rates for leaded fuel gasoline vehicles and, apparently, it was never widely distributed.

There are a few published studies which compare reasonably well with these estimates (Dietzmann, Parness, and Bradow, 1980; Dietzmann, Parness, and Bradow, 1981; Warner-Selph and Dietzmann, 1985; Black et al., 1985; Warner-Selph, 1991). In total, about 35 heavy duty vehicles were tested using a chassis dynamometer version of the engine test procedure (Urban, 1984). In the most extensive study, an average emission factor for heavy duty tandem axle tractors of 1.47 g/km (2.37 g/mi) was measured (17 vehicles). Estimated particle rate for 6 buses was 2.1 g/km (3.36 g/mi) and for 5 single axle tractors the estimate was 1.08 g/km (1.74 g/mi). These values seem generally consistent with those estimated by OMS.

The distribution of emission values for major vehicle categories is very wide. Figure 5 demonstrates the available data for all diesel and gasoline in-use trucks which have been tested thus far. There is a very wide range of emission rate for each category of vehicle, covering about 1 order of magnitude. A few of the better emission vehicles have come close to achieving the 0.25 g/bhp-hr standard with no emission controls, while worn engines with high lubricating oil consumption can emit at least 10 times as much material. It should be noted that the buses tested, most of which were equipped with DD 6 or 8V-71 engines, had substantially higher emission rates than comparably rated truck engines. However, this attribute was not shared by the DD 8V-92 series 2-stroke cycle engine with an emission rate similar to that of the 4-stroke cycle vehicles tested. Apparently the lubricant control problem referred to previously was limited to the 71 series engines.

Table 4 shows the current (1990) distribution of PM-10 emissions among the different vehicle types. While it may seem surprising that 70 percent of the PM-10 emissions are from gasoline-powered vehicles, in 1990, gasoline-powered vehicle miles traveled (VMT) outweighed diesel-powered VMT by more than 5 to 1.

Table 4. PM-10 Emissions for 1990 by Vehicle Type (Megagrams/year)

Gasoline Powered	**PM-10 Emissions**
Light-Duty Vehicles	596
Light-Duty Trucks	197
Heavy-Duty Trucks	47
Motorcycles	4
Diesel Powered	
Light-Duty Vehicles	11
Light-Duty Trucks	4
Heavy-Duty Trucks	369
Total	**1228**

Sulfur or sulfate emissions are of interest as a PM-10 precursor. Reductions in motor vehicle emitted sulfur are expected after 1990 as a result of motor vehicle diesel fuel being limited to 0.05 percent by weight. This limit is expected to produce about an 80 percent reduction in emissions per diesel-powered vehicle. On a total emissions basis, some of this reduction may be offset by the expected increase in diesel fuel consumption over the next decade.

Figure 6 shows how current PM emission rates by vehicle type are expected to be affected by the 1990 CAA Amendments (CAAA). Note that gasoline-powered vehicles will not have to install any additional controls in order to meet the standards. The same is

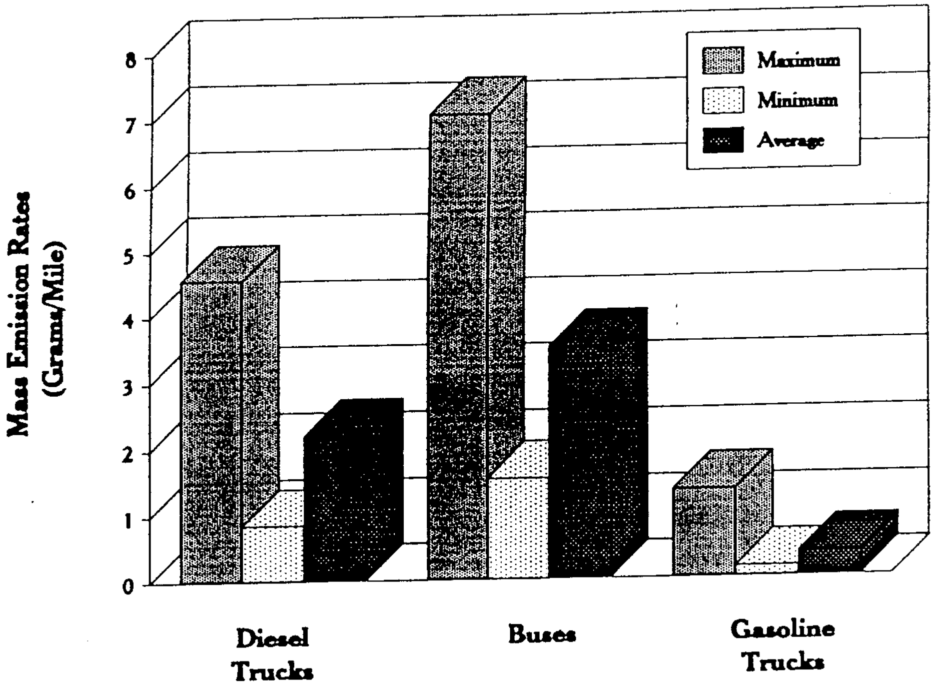


Figure 5. Measured emission rates from tailpipe emissions for various vehicle types.

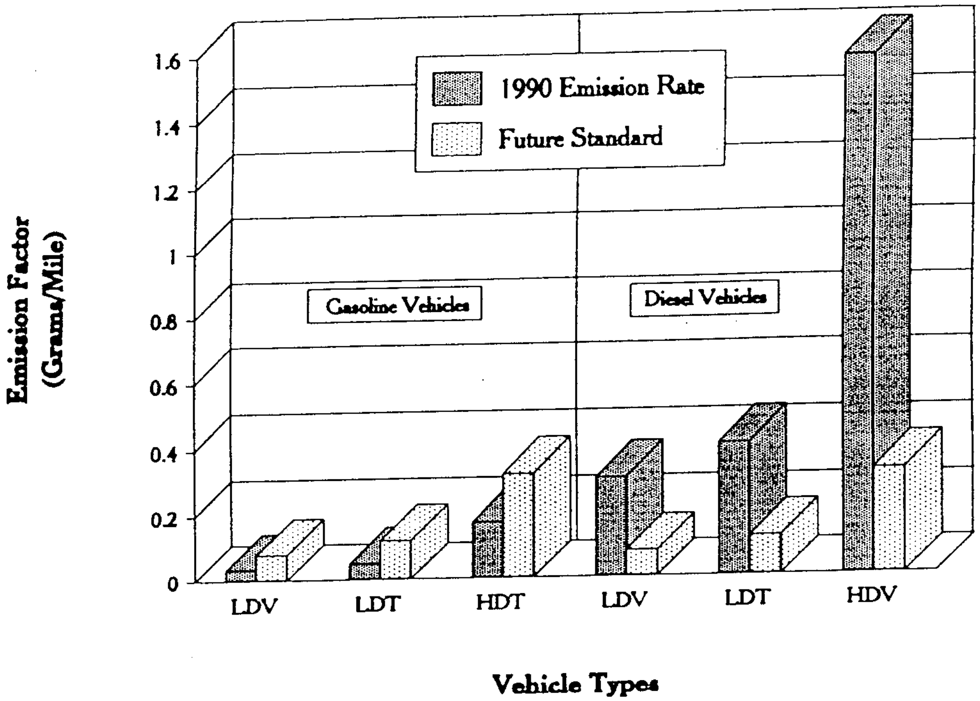


Figure 6. Current emission factors and future standards required by the 1990 CAAA for gasoline and diesel vehicles.

not true for diesels. As figure 6 shows, diesel emissions will have to be reduced by about 75 percent on average for the three vehicle types listed. Note that heavy-duty engine emission standards are in g/bhp. For the purposes of presentation in figure 6, the heavy-duty vehicle emission standard was converted to grams per mile by multiplying by 3.2. That factor may not be representative of all heavy-duty truck classes (or years).

EVALUATION OF THE IMPACT OF RESUSPENDED MATERIAL FROM PAVED ROADS

This section contains a discussion of the mechanisms and magnitude of particle resuspension from paved roads. As described in the previous section, resuspension of material from road surfaces can significantly affect ambient particulate loadings. There is an extensive body of knowledge on the subject of resuspension from unpaved roads which will be addressed in a later section.

Mechanism of Resuspension

Deposited material on a highway surface can be resuspended by either the mechanical action of vehicles passing over the surface or by the actions of nature (i.e., wind). Only studies pertaining to the actions of vehicles will be considered.

Nicholson and Branson (1990) report the results a study designed to evaluate the mechanism of resuspension from dry, paved roads. Fluorescent particles of known size were deposited on the road surface and the factors affecting their resuspension by vehicles travelling at known speeds were quantified. Particle diameters varied from a nominal diameter of 5 to 20 µm while speeds ranged from 5 to 40 mi/h. In all runs they observed that a large fraction of material was removed (resuspended) by the first passage of a vehicle. Subsequent passages had little effect. The amount suspended was strongly dependent on particle size with the larger particles being more readily removed. Vehicle speed also had a positive impact on resuspension. For 12 µm particles, 20 percent were resuspended by a vehicle travelling at 5 mi/h while 60 percent was resuspended by the same vehicle travelling at 40 mi/h. In addition, they observed that for the largest sizes (>9.5 µm) resuspension did not depend on the vehicle's tires passing through the material but rather on the induced turbulence of the vehicle as it passed over the material.

Sehmel (1973) performed a similar study using ZnS as a tracer. They observed the fraction resuspended to increase with vehicle speed, decrease with weathering (i.e., the longer the material was on the road surface, the lower the resuspension rate), and to be dependent on whether or not the vehicle passed through the material or in an adjacent lane.

Cowherd (1976) presents results on entrainment of fugitive material from paved roads. Emissions were observed to be dependent on traffic volume and surface loading. While obvious, this last result points out that for roads in areas where fugitive emissions are high (industrial or agricultural areas), resuspension from paved roads will play a more important role.

On-Road Studies

As mentioned above, results of on-road studies for paved roads are limited. Most of the attention has been focused on unpaved roads. Emission factors vary considerably depending on vehicle speed and weight, observed particle size, and material loading on the road's surface.

Sehmel (1973) defined the resuspension rate as the fraction of emitted mass of road material divided by the original mass of material. In this study ZnS was used as a tracer for the resuspended material. Either a car or a 3/4 ton truck was driven in the lane with the material or an adjacent lane. Vehicle speeds were 5, 15, 30, and 50 mi/h. The fraction resuspended increased with speed and ranged between 0.001 to 1 percent. Rates were greatest for vehicles travelling in the lane with the deposited material.

Under EPA sponsorship the Midwest Research Institute (MRI) performed a number of on-road studies quantifying emission factors (Cowherd, 1976; Cowherd et al., 1974, 1977, 1979, 1984; Bohn et al., 1978; Cuscino et al., 1983b). Cowherd (1976) observed emission factors to vary directly with traffic volume and surface loading (which was related to land use). For particles <30 µm diameter, the range of emission factors was from about 1 - 5 g/vehicle-mi. For TSP the mean values were: 8.2 g/vehicle-mi in a residential area, 2.0 g/vehicle-mi in a downtown commercial area, and 19 g/vehicle-mi in an industrial area. In a later report Cowherd et al. (1977) presented results for commercial/industrial/expressway measurements. Mean emission factors for $d < 30$ µm were 6.7/2,600/6.0 g/vehicle-mi. Obviously the degree of loading on the roads plays the key role in determining emission factors. Bohn et al. (1978) performed a study in an area dominated by iron and steel plants. The average emission factor was 430 g/vehicle-mi, consistent with the very high industrial result presented by Cowherd et al. (1977). Cowherd et al. (1979) and Cuscino et al. (1983b) further explored fugitive emissions from iron and steel plants. In the 1979 study, the average emission factor was 425 g/vehicle-mi for $d < 30$ µm. For $d < 15$ µm, the average result from the 1983 measurements was 267 g/vehicle-mi. PM-10 results were presented by Cowherd et al. (1984). For commercial/industrial, commercial/residential, expressway, and rural areas the mean values were 3.33, 2.35, 0.20, and 11.2 g/vehicle-mi, respectively.

In other studies, Axetell and Zell (1977) reported a TSP value of 3.3 g/vehicle-mi for streets in areas with differing land use.

Based on these studies, it should be emphasized that the important quantity which determines emission rates from paved roads is soil loading on the surface. That, combined with estimates of VMT, will be key in assessing the importance of highway particulate impacts.

Ambient Air Studies

As has been indicated above, within the context of tailpipe particulate emissions, a number of studies quantifying the contribution of geological material to the total particulate burden were reviewed. In urban areas much, if not all, of the observed ambient geological material has been attributed to resuspended road dust and this component is generally dominant over that contributed by tailpipe emissions.

In the 1981-82 Chicago study, Scheff et al. (1984) attributed 44.3 percent of the TSP to soil/geological material vs. 4.8 percent to mobile sources. In the smaller size fraction mobile sources accounted for 4.6 percent of the material while soil accounted for 19.9 percent. Since soil/road dust tends towards the larger size ranges the lower percentage contribution in the fine size range is to be expected.

Thurston and Spengler (1985) reported the soil contribution accounted for 11.0 percent of the PM-2.5 and 57.6 percent of the PM-15 in the Boston area.the to metropolitan Boston over a two year period. The results for motor vehicles were 14.4 percent of the PM-2.5 and 7.0 percent of the PM-15.

The series of studies performed in the Western U. S. by Watson and Chow similarly demonstrate the importance of contribution of geological material (presumably from resuspended road dust) on observed particulate levels. For the 1986-87 Reno study, Chow et al. (1990) reported mobile sources contributed 44 percent of the PM-10 during the months of February and March while 50 percent was attributed to geological material. Another interesting finding was that vacuum street sweeping did not reduce the geological contribution.

In the 1989-90 Phoenix PM-10 Study, Chow et al (1991a) concluded PM-10 was generally highest in the areas downwind of the urban areas. Primary geological material, which they attributed almost entirely to road dust, was a major contributor to PM-10 at every sampling site. Geological material typically contributed from 40 to 60 percent of the PM-10 at the three urban sites on the days with highest concentrations. Primary motor vehicle exhaust typically contributed from 30 to 50 percent of the PM-10 at the three urban sites on the same days.

Similar observations were made in Tucson (Chow et al., 1991b) where primary geological material accounted for 50 percent of the PM-10 and primary motor vehicles accounted for 30 to 40 percent of the PM-10.

UNPAVED ROADS

The EPA has expended considerable effort towards development of an emission factor for dust from vehicle travel on unpaved roads. Many field studies have been conducted to gain an understanding of the relationships between various field parameters and the emission factor. The purpose of this section is to provide a general understanding of the conclusions of these studies which relate to PM-10 impacts from unpaved roads. The major field studies in which empirical data has been gathered and analyzed are reviewed here. However, several issues must first be discussed.

Initially, field studies were focused on deriving factors for TSP. In 1987 the particulate NAAQS were modified to create a new particulate standard, particulate matter smaller than 10 μm in aerodynamic diameter, or PM-10, and, as a result, a better understanding of the relationships between PM-10 emission rates from unpaved roads and various field parameters was needed. Unfortunately, relatively little effort has gone into field research of these relationships since 1987. Although a few field studies have been conducted specifically for PM-10 emission factor development, the findings of the older, TSP studies must also be discussed in order to provide the fullest possible perspective on the impacts of both dust in general and PM-10 specifically from vehicle travel on unpaved roads. To apply the findings of the TSP emission factor studies, the relationship between TSP and PM-10 must be understood. Several issues may confound a clear understanding of this relationship.

The first is a very basic difference in the way TSP and PM-10 are operationally defined. TSP is defined as the filtrate mass divided by the air volume filtered by a high volume sampler. There is no definite upper bound on the size of particles collected in a TSP sample. On the other hand, PM-10 is defined as airborne dust particles smaller than a specific aerodynamic diameter, 10 µm. TSP is frequently considered as particles with Stokes diameter less than 30 µm (e.g., Bohn et al, 1978), although the upper bound on particle size varies, depending on wind speed and direction. For the purpose of this report, TSP will be assumed to consist of particles smaller than 30 µm Stokes diameter.

Herein lies a second caveat in the relationship between TSP and PM-10 emission factors: whereas the earliest field studies considered Stokes diameter to be the most appropriate measure of particle size, most later studies used aerodynamic diameter. The Stokes diameter of a given particle is "the diameter of a sphere having the same density and settling velocity as the particle in question" (Hesketh and El-Shobokshy, 1985). Aerodynamic diameter is the diameter of a sphere of unit density that has the same settling velocity as the particle of concern" (Hesketh and El-Shobokshy, 1985). Because the NAAQS for PM-10 is expressed in aerodynamic diameter, that is the size measurement of interest.

In developing fugitive dust emission factors for unpaved roads, the EPA has relied primarily on a single contractor, the MRI, to conduct the field studies and analyze the resulting data. MRI developed the exposure profiling equipment and methodology by which dust emissions from fugitive sources are directly measured under isokinetic conditions. Therefore, the emission factors for unpaved roads published by EPA are in large part the result of MRI's efforts. In fact, MRI was involved in eight field studies which generated data on emission rates and predictive field parameters for unpaved roads. After each field study, the new data was added to the database, and the predictive equation was revised using multiple regression analysis. In general, each successive field study either expanded the ranges of the predictive variables or added a new variable to the database of field tests, thus increasing the range of field conditions to which the estimated statistical relationships could be applied to estimate emissions. Some of the field studies have been conducted on industrial haul roads, including those in iron and steel plants and strip mines. Caution should be used in applying the findings of these studies to public unpaved roads, because significant differences may exist in the character of the road surface material and the traffic.

Although MRI has played a major role in the development of unpaved road emission factors, other organizations have also studied the relationships between various field conditions and the emission rate. The findings of these studies are also discussed in this review. Each field study, regardless of the organization which performed it, is discussed in chronological order.

Roberts (1973) sampled the dust plume created by vehicle travel on a gravel road using a towed rack to which a cascade impactor was attached. The rack was designed as a vertical grid oriented perpendicular to the car's path; the impactor was rotated among the various positions between tests so that, after a series of tests, the average concentration of dust in the plume could be determined. In order to derive an emission factor in lb/vehicle-mi, the average plume concentration was multiplied by the volume of air into which it was emitted. This volume was estimated in the following manner. First, the area of the plume behind the car was estimated by towing a grid/rack of open impaction plates and examining the dust pattern on the plates. Second, this area — 70 square feet — was multiplied by 5,280 feet (1 mile) to obtain the air volume (36,960 cubic feet [ft^3]) into which the dust was emitted after 1 mile of travel.

The impactor samples were also analyzed to determine the particle size distribution of the plume. This particle size breakdown was then used to estimate emission factors for various particle size categories based on the measured total particulate emission factors.

Seventeen sampling runs were conducted at 20 mi/h on a gravel road in the Duwamish Valley in Seattle. Roberts noted that the test road had been sprayed with oil two years before, and that emissions from the road were still effectively suppressed at the time of the study.

For these 17 runs, the average concentration of total particulates in the air sampled by the cascade impactor was .133 grains/ft^3. Following the procedure explained above, the emission factor was calculated at 7 lb/vehicle-mi.

Limited testing was also conducted with vehicle speeds of 10 and 30 mi/h. The cascade impactor collected dust samples at only two grid positions on the sampling rack (as compared to 6 positions at 20 mi/h). The following proportional relationship was assumed in estimating an emission factor for the 10 mi/h tests:

$$D_2/D_1 = X/Y$$

where: D_2 = average concentration at grid sampling point D at 20 mi/h
D_1 = average concentration at grid sampling point D at 10 mi/h
X = total average (of all grid sampling points) at 20 mi/h
Y = total average (of all grid sampling points) at 10 mi/h

The same procedure was used to estimate an emission factor for 30 mi/h.

The calculated factors for the various speeds are shown below in table 5. Roberts also presented emission factors for particles smaller than 10 µm and for particles smaller than 2 µm in diameter, though he did not explain how the fraction of the total particulate sample consisting of particles smaller than 10 µm was determined.

Table 5. Emission Factors for Various Speeds

Speed (mi/h)	Emission Factor (lb/vehicle-mi)		
	Total Particulates	PM-10	$d^* < 2\ \mu m$
10	3.5	.58	.10
20	7.0	1.9	.24
30	22.2	9.0	.77
20	7.3	2.0	nd

* particle diameter

Jutze and Axetell (1974) utilized a beta gauge to determine particulate concentrations at several heights and distances from an active unpaved road. These data points were substituted into the equation for a continuously emitting infinite line source (Turner, 1970), yielding an estimate for source strength (g/m/sec). This was then converted into an emission factor (lb/vehicle-mi) by dividing by the rate at which vehicles passed the sampling point (vehicle/minute).

Regression analysis was used to estimate the relationship between the measured emission factor and vehicle speed. Because the hi-vol was considered the standard equipment for measuring total suspended particulates, and it includes in its "catch" a much larger range of particle sizes than does the beta gauge, the average ratio of concentrations measured with the hi-vol sampler to those obtained with the beta gauge was used as a multiplier for the estimated relationship.

Simple wind erosion of particulates from unpaved roads was also considered. The researchers estimated emissions caused by wind using a model developed by Woodruff and Siddoway (1965). No field data on concentrations downwind from an unpaved road without traffic were collected; rather, assumptions were made for the values of predictor variables in the model. Emissions were estimated in tons/mile/year and divided by annual traffic volume (average daily traffic * 365) to derive a factor in lbs/vehicle-mi. This wind erosion factor was then added to the factor representing vehicle travel on unpaved roads to obtain a total emission factor for unpaved roads.

Although the investigators claimed to use the Guassian model for continuously emitting line sources, as found in the Workbook of Atmospheric Dispersion Estimates (Turner, 1970), their equation differs significantly from the model presented in that document. The equation as it appears in the workbook is:

$$\chi\ (x, y, O; H) = \frac{2\,q}{\sin \phi \sqrt{2\,\pi}\ \sigma_z\ u}\ \exp\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right]$$

The formula applied by the investigators was:

$$\chi\,(x,y,O;H) \;=\; \frac{2\,q}{\sqrt{2\pi}\;\sigma_z\,u}\;\sin\phi\;\exp\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right]$$

The ϕ represents the angle between the mean wind direction and the road.

Furthermore, contrary to Turner's (1970) recommendation, the investigators collected much of their data during periods in which ϕ was less than 45°.

The regression analysis mentioned above produced the following best fit line:

$$E = (0.16)(1.068)^x$$

where: E = dust emissions (lb/vehicle-mi)
x = vehicle speed (mi/h)

No upper bound on the particle size was given for this study. Evaluating this formula for x = 30 produces an emission factor of 1.15 lb/vehicle-mi. The vehicle speeds tested ranged from 15 to 40 mi/h. The silt content of the road surface material was not recorded.

The investigators found that plume concentration as measured by the standard hi-vol method averaged 1.68 times higher than the concentration estimated by the beta gauge, with a correlation of .87. Thus the coefficient in the above equation was multiplied by 1.68, and the equation was revised to be:

$$E=(0.27)(1.068)^x$$

In this case E is the emission factor for total suspended particulates. Evaluating the formula for x = 30 results in E = 1.94 lb/vehicle-mi.

The estimated emission factor for wind erosion on unpaved roads was 1.54 lb/vehicle-mi. The sum of these two factors is 3.7 lb/vehicle-mi.

Under a contract with the EPA the MRI developed and applied its exposure profiling methodology for measuring emissions for unpaved roads (Cowherd et al., 1974). The term exposure profiling refers to the measurement of the total horizontal flux of dust downwind from the source. This is done by isokinetically measuring exposure at several heights in the dust plume simultaneously. Integration of exposure with respect to height yields the dust mass flux per unit length of road, which is in turn divided by the number of vehicle passes to give an average emission factor for the test.

For each test the vehicle speed was either 30 or 40 mi/h. The mass percent of the loose road surface material in particles which can pass through a 200 mesh screen, typically referred to as "percent silt," was generally between 12 and 20; for one test the surface material was 68 percent silt.

Emission factors were calculated for four particle size categories, based on Stokes diameter: 1) particles of all sizes, 2) particles larger than 30 µm, 3) particles between 2 and 30 µm, and 4) particles smaller than 2 µm. Summing the latter two for each test yields an emission factor for particles smaller than 30 µm. For particles smaller than 30 µm in Stokes diameter, emission factors for the tests ranged from 3.9 to 8.1 lbs/vehicle-mi, except for the test that had factor of 31.9 lbs/vehicle-mi; the road used for this test had an unusually high percent silt, as noted earlier.

The investigators developed the following equation for estimating emissions of particles smaller than 100 µm in Stokes diameter from vehicle travel on unpaved roads, based on the data collected in this study:

$$e = 0.81\ s\ (S/30)$$

where: e = Emission factor (pounds per vehicle-mi)
s = Silt content of the road surface material (percent)
S = Average vehicle speed (mi/h)

This equation applies only to days with rainfall less than 0.01 inches. The predictions made using this equation for the six tests conducted were within 10 percent of the actual lb/vehicle-mi.

In a study performed by researchers at Pima County, Arizona Air Quality Control District, dust emissions from vehicle travel on unpaved roads were estimated using dispersion modeling (Lemon et al., 1975). Standard hi-vol samplers were used to measure upwind and downwind concentrations. The Gaussian equation for a continuously emitting line source (Turner, 1970) was used to derive emission rates from the calculated concentrations and other relevant field data. It should be noted that the investigators did not correctly apply the model: they made the same mistake as Jutze and Axetell (1974, see review of that study). The investigators calculated total suspended particulate emission factors of 8.14, 5.37, 6.81, and 3.25 lb/vehicle-mi. The average traffic speed was 30 mi/h. No measurements were taken for the road surface percent silt.

Dyck and Stukel (1976) utilized the upwind-downwind method to estimate emission factors for a truck traveling on unpaved roads in a construction access area. One upwind hi-vol sampler, 50 feet from the road, and four downwind hi-vol samplers, ranging from 50 to 250 feet from the road, were used to measure the road's contribution to dust concentration. This data was applied to a modification of the dispersion model for a continuously emitting, infinite line sources found in the Workbook of Atmospheric Dispersion Estimates (Turner, 1970) to yield estimated emission factors. The truck's weight and speed were varied by the investigators, as was the road surface type. Testing was conducted over several days. On each day, the choice for a test road was dictated by the prevailing wind direction. Tests were conducted with vehicle speeds of 10, 17.5, and 25 mi/h, and with vehicle weights of 8,580, 12,620, and 16,530 pounds. Each test lasted about one hour.

Measured emission factors and corresponding field parameters were not reported. The investigators used MLR to develop an equation for estimating the emission factor as a function of the experimental variables:

$$E = 5.286 - 3.599(R) + 0.00271(V)(W)(S)$$

subject to:

$$10 \leq V \leq 25$$

where: E = suspended particulate emission factor, lb/vehicle-mi
R = road type, silty-sand or clay
V = velocity, mi/h
W = weight, thousands of pounds
S = road surface percentage of silt particles

The dispersion model published in this article was an incorrect adaptation of the model cited in Turner, 1970. The model for a continuously emitting, infinite line source as it is presented by Turner (1970) and by the present study is

$$C(x,y,O,H) = \frac{2q}{\sin\phi \sqrt{2\pi}\,\sigma_z\, U} \exp\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right]$$

The investigators incorrectly solved this equation for source strength, q, as follows:

$$q = \frac{C \sin\phi \sqrt{2\pi}\ \sigma_z U}{2} \exp\left[-\frac{1}{2}\left(\frac{H}{\sigma_z}\right)^2\right]$$

The 'exp' term should be in the denominator in this equation. Because field data were not presented, it was not possible to determine whether this was a typographical error or a misapplication of the model.

Three different experiments were conducted by researchers from the U.S. Army Construction Engineering Research Laboratory (Struss and Mikucki, 1977). For the first phase a fully enclosed 25 foot diameter test track was used to model the effects of soil moisture potential, vehicle speed, and vehicle weight on emissions. Soil water potential is a measure of both the amount of moisture in the soil and how tightly that moisture is held in the soil. A cross-arm spanning the diameter of the track held one tire at each end and was supported by a vertical center pole, which could be rotated at various speeds. Emissions were measured by sampling the dusty air which was ducted from the enclosure. The measured particle size range (e.g., all particles less than 30 µm in aerodynamic diameter) was not documented for this phase or for any other part of this study. Gooselake clay was used to surface the test track.

For the second phase, upwind-downwind dispersion modeling was used to estimate emissions from a newly cleared unpaved road. The soil on the road surface was Drummer silty clay loam. Vehicle speed and weight were varied between tests. Soil water potential was also measured in this experiment. The emission rate was calculated using the Gaussian model for a continuously emitting, infinite line source.

For the third phase, soil from the outdoor test area was applied to the enclosed test track and tested in the same manner as described for the first phase. Thus, the only difference between this phase and the first was the soil type used for the road surface.

Data from the third and first phases were compared to determine the effect of the change in soil type. Data from the third and second phases were compared to evaluate the model, i.e., the test track, relative to the process being modeled, dust generation from outdoor haul roads. The authors felt that the outdoor test roads realistically represented construction haul roads.

Linear regression analysis of the data from the first phase yielded the following relationship, with an R^2 of 0.64:

$$\ln(D) = -5.37 + 0.12(T) + 0.21(S) + 0.90(W)$$

where: D = emission rate, grains/vehicle-mi
T = soil water potential, atmospheres of tension
S = vehicle speed, mi/h
W = vehicle weight, lb/tire

The emission rate was calculated in grains/tire-mi and then multiplied by six to get grains/vehicle-mi. Each of the independent variables were significant at the .95 level. Vehicle speed ranged from 5 to 15 mi/h, and weight ranged from 1,850 to 2,450 lbs/tire. The soil water potential ranged from about 10 to about 65 atmospheres of tension. The emission rate ranged from 0.99 to 136 grains/tire-mi.

The following equation was produced using linear regression analysis of the outdoor test road data:

$$\ln(D) = 5.28 + 0.01(T) + 0.06(S) + 0.50(W)$$

The R^2 value for this model was .66. In this experiment vehicle speed ranged from 7 to 15 mi/h, the weight per tire ranged from 1,370 to 2,500 lbs/tire, and the soil water potential ranged from 11.9 to 153.6 atmospheres of tension. The emission rate varied between 599 and 5,830 grains/tire-mi. With a six-wheel truck, this equates to an emission factor range of 0.5 to 5 lbs/vehicle-mi.

In the third phase of the study, soil plastic limit was considered as a potential predictor of TSP emissions, although no conclusion was drawn regarding its usefulness. Plastic limit, according to Struss and Mikucki, is "the percent moisture, by weight, contained in a soil when it passes from a plastic to a brittle state while drying." Of course, this variable would not be appropriate for gravel roads.

This is one of the few studies in which the affect of soil moisture on emission rate is systematically studied.

Data on upwind and downwind dust concentrations and on mean wind speed and direction were collected and applied to a dispersion model to estimate an emission factor for vehicle travel on unpaved roads by McCaldin (1977). Five pre-existing dirt roads in Pima County, Arizona were the test sites for this study.

Upwind and downwind concentrations were measured using standard hi-vol samplers and standard gravimetric methods. Sierra Impactors were used to measure particle size distribution. Vehicle speed was controlled by the investigator for most of the passes. The speed of the remainder were estimated visually. The number of passes was likewise controlled mostly by the investigator. Composite samples of road surface material were analyzed for percent silt using a 200 mesh screen. The Belfort Model 443 anemometer, held at 2 meters above the ground, was used to measure wind speed and direction.

Data were collected on 46 sampling runs. The measured emission factors, reported only in graphic form, varied roughly between 0.25 and 10 lbs/vehicle-mi. The road surface during testing ranged from 6 to 14 percent silt. Average traffic speed for the tests was between 10 and 50 mi/h. The average emission rate for vehicles traveling 30 mi/h on a road with a 10 percent silt surface was about 3 lb/vehicle-mi. Emission rates were found to vary directly with percent silt and exponentially with vehicle speed. An equation relating these three variables was estimated from the collected data as follows:

$$E = (s)\ 0.035\ (S)^2$$

where: E = TSP emission factor, lb/vehicle-mi
s = mass percent silt
S = traffic speed, mi/h

Exposure profiling, a technique originally developed by MRI in 1974, was used by Bohn et al. (1978) to measure emissions of particulates from vehicle travel on unpaved roads within integrated iron and steel plants. A total of nine sampling runs were conducted on unpaved roads, three on a road with fine slag cover and six on roads with a hard-based dirt cover.

Measured emission factors ranged from 4.9 to 29 lbs/vehicle-mi for suspended particles, nominally those with Stokes diameter less than 30 µm. The road surface material ranged from 4.1 to 8.7 percent silt, and the average vehicle speed ranged from 14 to 30 mi/h. The average vehicle weight ranged from 3 tons (light-duty vehicles) to 70 tons (loaded haul trucks).

The predictive equation which was published in this study and which is based on the cumulative database gathered by MRI is shown below:

$$e = 5.9(s/12)(S/30)(W/3)^{0.8}(d/365)$$

where: e = suspended particulate emissions, lb/vehicle-mi
s = road surface silt content, percent
S = average vehicle speed, mi/h
W = average vehicle weight, tons
d = dry days per year

Axetell utilized upwind-downwind dispersion modeling to measure TSP emissions from haul roads at two western surface coal mines. An effort was made to measure particle fallout rates. Concentrations at a series of downwind distances from the source were measured, and corresponding emission rates at the source were calculated using the above model, which assumes there is no particle fallout. Decreases in the "apparent emission rate" with increasing distance from the source would serve as a measure of fallout.

This theory was not supported by the empirical data. The apparent emission rate from haul roads did not generally decrease with downwind distance. For those tests in which concentrations were measured at downwind distances differing by ten meters, the concentration **increased** an average of 19 percent between the two sampling points.

Emission estimates based on concentration measurements taken 1.2 meters above the ground and between eight and ten meters downwind ranged from 1.7 to 21.1 lbs/vehicle-mi. Field parameters such as road surface percent silt and vehicle speed were not studied. It should be noted that the haul trucks weighed between 50 and 150 tons.

Exposure profiling was used by Cowherd et al. (1979) as the primary technique to estimate emissions of fugitive dust from two unpaved roads in iron and steel plants. One of the roads had a crushed slag surface and the other had a mixed dirt-slag surface.

Emission factors were presented for particles smaller than 30 µm and smaller than 5 µm in Stokes diameter for each test. Measured factors for the larger size category ranged from 1.7 to 16 lbs/vehicle-mi, and the factors for particles smaller than 10 µm ranged from 0.53 to 3.4 lbs/vehicle-mi. The average vehicle speed ranged between 15 and 29 mi/h, and the road surface percent silt ranged from 24 to 47. Some tests were conducted while the traffic was exclusively passenger cars, while others included a significant fraction of heavier haul truck. As a result, the average vehicle weight ranged from 3 to 30 tons. A new variable, the average number of wheels per vehicle traveling the road segment, was added to the equation. The addition of this predictive variable is indicative of the fact that the emissions database collected by MRI was expanded under this study to include a wider range of vehicle types. The average number of wheels varied between 4 and 13.

The revised equation is presented below. Data collected in this study of unpaved road emissions was added to MRI's database of field collected emissions data, and, using this larger database, a new predictive emission factor equation for particles smaller than 30 µm in Stokes diameter was developed.

$$e = 5.9(s/12)(S/48)(W/2.7)^{0.7}(w/4)^{0.5}(d/365)$$

where: e = Emission factor for particles < 30 µm in Stokes diameter, lbs/vehicle-mi
s = Silt content, percent
S = Average vehicle speed, mi/h
W = Average weight per vehicle, tons
w = Average number of wheels per vehicle
d = Dry days per year

According to the EPA (1985a), an equivalent equation for predicting PM-10 emissions is

$$e = 2.124(s/12)(S/48)(W/2.7)^{0.7}(w/4)^{0.5}(d/365)$$

where all variables are defined as above.

Emissions from six-wheeled haul trucks traveling on unpaved roads at a taconite mine were measured by Cuscino (1979). Exposure profiling was used to measure particulate emissions. Tests were conducted on two different road surface types, sand/gravel and crushed rock.

The plume's particle size distribution was measured using a high volume cascade impactor with a cyclone preseparator Standard hi-vol samplers measured total suspended particulate concentration at several downwind distances.

The measured emission factors ranged from 3.7 to 25.0 lbs/vehicle-mi for particles with Stokes diameter less than 30 µm. Silt content ranged between 4.7 and 6.1 percent, and the average vehicle speed was between 13.5 and 20 mi/h. The average vehicle weight for each test ran varied between 67 and 157 tons.

A regression analysis was conducted on the data from this study and previous MRI studies of unpaved road emissions (Cowherd et al., 1974; Bohn et al., 1978; and Cowherd et al., 1979). Although eight sampling runs were conducted, only three were included in this regression analysis; the remaining five runs were conducted under conditions considered unrepresentative of dry, uncontrolled, unpaved roads. The following equation was developed from this analysis:

$$e = 0.00380\ s\ SW^{0.7}w^{0.5}$$

where: e = Emission factor for particles with Stokes diameter < 30 µm, lb/vehicle-mi
s = Silt content, percent
S = Average vehicle speed, mi/h
W = Average vehicle weight, tons
w = Average number of wheels per vehicle

According to the method set forth in EPA, 1985a, this equation can be translated into a PM-10 emission factor:

$$e = 0.001368\ s\ SW^{0.7}w^{0.5}$$

where e is the emission factor for particles with aerodynamic diameter less than 10 µm, in lbs/vehicle-mi, and all other variables are defined as before.

Exposure profiling was used to measure emissions from unpaved rural roads and private roads in stone crushing, sand and gravel processing, and copper smelting facilities (Reider, 1983). The traffic on each of these road types is described in table 6.

Table 6. Description of Traffic for Industry Types

Industry	Traffic Type
Stone crushing	Medium Duty
Sand and gravel processing	Heavy Duty
Copper smelting	Light Duty
Public roads	
crushed limestone	Light Duty
dirt	Light Duty
gravel	Light Duty

Emission factors were reported for total particulates and for PM-10. The PM-10 factors ranged from 0.713 to 12.1 lbs/vehicle-mi. Interestingly, the highest emission factor was from a test on a public road with light duty traffic. Mean vehicle speed ranged from 5 to 35 mi/h, and mean vehicle weight ranged from 2 to 32 tons. The average number of wheels on vehicles in the rural road tests was always four; for the industrial roads, the average number of wheels per vehicle was as high as 16.6 for one sampling run. The silt content on the roads varied between 4.1 percent and 35.1 percent. No statistical analysis of the data was included in this report.

Exposure profiling was used to measure emissions of dust from vehicle travel on two gravel roads in iron and steel plants (Cuscino et al., 1983b). One road was used primarily by heavy-duty vehicles, and the other was predominantly light-duty traffic.

Emission factors were reported for three particle size categories: total particulates (i.e., no size limit), particles smaller than 15 µm in aerodynamic diameter, and particles smaller than 2.5 µm in aerodynamic diameter. Emission factors for the 15 µm category ranged from 1.05 to 4.25 lbs/vehicle-mi for light duty traffic and from 25.9 to 33.5 lbs/vehicle-mi for heavy duty traffic. Emission factors for particles smaller than 2.5 µm in aerodynamic diameter varied between 0.245 and 1.27 lbs/vehicle-mi for light duty traffic and between 7.74 and 8.84 lbs/vehicle-mi for heavy duty traffic. For runs with heavy duty traffic, the average vehicle weight ranged from 22 to 53 tons, whereas for the light duty traffic tests, the average vehicle weight was 3 tons.

A new predictive equation was not developed on the basis of the empirical data from this field research. The investigators interpolated from the three size categories noted above (assuming a log-normal distribution of particle sizes) to estimate an emission factor for particles less than 30 µm in Stokes diameter (the nominal cutpoint for TSP) for each of the sampling runs. Then they tested the predictive accuracy of an equation estimated previously by MRI:

$$e = 5.9(s/12)(S/30)(W/3)^{0.7}(w/4)^{0.5}(d/365)$$

where: e = Mass of particulates < 30 µm in Stokes diameter, lb/vehicle-mi
s = Silt content of road surface material, percent
S = Average vehicle speed, mi/h

W = Average vehicle weight, tons
w = Average number of vehicle, wheels
d = Number of dry days per year

The ratio of predicted to actual (interpolated) emission factors ranged from .34 to 1.21 for the seven sampling runs on uncontrolled, unpaved roads.

In a project conducted by the MRI (Axetell and Cowherd, 1984), emissions from unpaved roads in western surface coal mines were measured, using exposure profiling and upwind-downwind dispersion modeling. Profiles were created using both the isokinetic exposure profiler originally developed by the MRI and another profiler consisting of a vertical array of dichotomous samplers. Because the dichotomous sampler was found to provide the most consistent results, data collected using it were considered the closest to true emissions.

Emission factors were developed for particles with aerodynamic diameters less than 2.5 µm and for particles having aerodynamic diameters less than 15 µm. Emission rates for other particle size ranges were also measured, but these two are most relevant to an understanding of PM-10 emission rates. The dichotomous sampler provided cutpoints at 15 µm and 2.5 µm in aerodynamic diameter.

Traffic in approximately two-thirds of the sampling runs was dominated by haul trucks, and the remainder were dominated by light- and medium-duty trucks. Emissions of particles smaller than 15 µm ranged from 1.1 to 17.7 lbs/vehicle-mi for haul truck tests, and from 0.33 to 6.6 lbs/vehicle-mi for light and medium duty vehicles. Emissions of particles smaller than 2.5 µm in diameter ranged from 0.05 to 0.69 lbs/vehicle-mi for haul trucks, and from 0.03 to 1.5 lbs/vehicle-mi for light and medium duty traffic.

In the haul truck tests, the average vehicle weight varied between 24 and 125 tons, the average vehicle speed ranged from 14 to 35 mi/h, and the average number of wheels ranged from 4.9 to 10. The road surface percent silt ranged from 2.2 to 14.2. For test conducted with mostly light and medium duty traffic, the average vehicle weight was between 2 and 7 tons, and the average vehicle speed was between 24 and 43 mi/h. Nearly all vehicle had four wheels. The road surface silt content varied between 4.9 and 10.1 percent.

Multiple regression analysis was used to develop predictive equations relating emission factors for particles smaller than 15 µm in aerodynamic diameter to various parameters. It should be noted that, in addition to those tests of uncontrolled roads described above, the results of other sampling runs in which water was applied to the road as a dust control were included in this statistical analysis. As a part of this study to determine the effectiveness of water as a dust control, the percent moisture in the road surface was also monitored. The equations are shown below.

Traffic Type	Emission Factor (lbs/vehicle-mi)[a]
Light- and Medium-Duty Vehicles	$3.72/M^{4.3}$
Haul Trucks	$0.0051\ w^{3.5}$

a) variable definitions:
M = moisture content, mass percent;
w = average number of wheels;

In 1985 Cowherd and Englehart (1985) published a statistical analysis of qualifying data generated in earlier field studies of PM-10 emission factors. Only three studies directly measured PM-10 emissions: Axetell and Cowherd, 1981; Cuscino et al., 1983b; and Reider, 1983 (unpublished). The following variables were considered as candidate predictors of PM-10 emissions: mass percent silt, silt loading (mass/unit area), average vehicle weight, average number of wheels per vehicle, and average vehicle speed.

The initial regression analysis indicated the much of the variation in emissions from the coal mine haul roads (Axetell and Cowherd, 1981) was not explained be the candidate variables, whereas the emissions variability for the other studies was more adequately explained. Therefore, the data from Axetell and Cowherd, 1981 was removed from the database. From the remaining 26 records in the database, the following relationship was estimated:

$$PM_{10} = 0.766\left(\frac{sL}{400}\right)^{0.7}\left(\frac{W}{7}\right)^{0.4}\left(\frac{S}{24}\right)^{0.8}$$

where: PM-10 = PM-10 emission factor, kg/vehicle-km
sL = silt loading, g/m^2
W = mean vehicle weight, Mg
S = mean vehicle speed, kph

This equation is equivalent to the following equation in English units:

$$PM_{10} = 2.72\left(\frac{sL}{0.082}\right)^{0.7}\left(\frac{W}{7.714}\right)^{0.4}\left(\frac{S}{14.88}\right)^{0.8}$$

where: PM-10 = PM-10 emission factor, lbs/vehicle-mi
sL = silt loading, lbs/ft^2
W = mean vehicle weight, tons
S = mean vehicle speed, mi/h

This equation explains 73 percent of the variation in the PM-10 emission factor.

Southern Research Institute conducted a side-by-side field comparison of five organizations measuring emissions from vehicle travel on an unpaved road (Pyle and McCain, 1986). The organizations were Energy & Environmental Management, Inc. (EEM), MRI, PEI Associates, Inc. (PEI), TRC Environmental Consultants, Inc. (TRC), and United States Steel Corporation (USS). All five organizations used the same basic methodology: exposure profiling. However, each organization chose its own sampling equipment, equipment configuration, and analytical procedures. Consequently, each organization's implementation of profiling differed in some ways from the others. Therefore, this study provided the opportunity for evaluation of the ease of implementation of the various procedures, their accuracy, and the comparability of the results.

The test site was a paved slag haul road covered, for the purpose of this study, with 5 to 10 centimeters of aggregate. This aggregate was composed of clay, iron ore, and boiler ash, yielding an average silt content of 10 percent. The site was in a USS facility in Gary, Indiana, about 50 meters south of Lake Michigan. Five test positions were established on each side of the road. Traffic on the road was a mixture of service vehicles and dump trucks.

PM-10 emission factors were developed from data on exposure of total particulates and from measured particle size distributions of the dust plumes generated by the passing vehicles. The calculated emission factors ranged from 1.78 to 15.6 lbs/vehicle-mi. The road surface percent silt ranged from 3.9 to 21.0. The average vehicle speed varied between 16 and 28 mi/h. The average vehicle weight ranged from 6.3 to 35 tons, and the average number of wheels was between 4.6 and 16.

Two of the participating organizations developed equations for predicting emission factors based on field parameters. PEI used multiple regression analysis to derive the following relationship:

$$PM\text{-}10 = 0.20(W)^{0.59}$$

where: PM-10 = emission factor for PM-10, kg/vehicle-km
W = average vehicle weight, megagrams

The R^2 for this equation was 0.52.

EEM developed the following equation using regression analysis:

$$PM\text{-}10 = 0.557(s)^{0.139}(R)^{-0.203}(w)^{0.267}(W)^{0.395}$$

where: PM-10 = emission factor for PM-10, kg/vehicle-km
s = percent silt
R = ambient relative humidity, percent
w = average number of wheels
W = average vehicle weight, megagrams

The R^2 for this equation was 0.41.

Some of the most recent work involved with development of emission factors from unpaved roads has been carried out by researchers at the Illinois State Water Survey (ISWS) as part of the 1985 National Acid Precipitation Assessment Program (NAPAP) [Williams and Stensland, 1989; Williams et al., 1988; Barnard et al., 1988]). As part of that research, a total of 18 aerosol emissions measurements experiments were conducted. However, only 11 of these experiments resulted in data of sufficient quality to use in determining the emission factor developed for the NAPAP emission inventory. Several of these experiments were preliminary in nature (designed to test equipment configurations and to ascertain the viability of some of the experimental concepts) and were never intended to yield quality assured/quality controlled data. In addition, a few of the experiments were only partially completed due to changing meteorology (primarily shifting, nonconstant winds).

In general, unpaved road aerosol samples were collected next to several unpaved roads in Illinois using both dichotomous and high volume samplers. With the exception of the last four experiments, six dichotomous and four high volume samplers were used. One dichotomous and one high volume sampler were operated on the upwind side of the road to provide background correction values. Five dichotomous and three high volume samplers were used on the downwind side. Typically, samplers were located 10, 20, and 30 meters downwind. At the 20 meter location, three dichotomous samplers were employed in a vertical array as an exposure profiler. The inlet heights were at 1.5, 3.5, and 4.9 meters. A ground level high volume sampler was located at each downwind location. The high volume samplers were utilized to evaluate the PM-10/TSP ratio.

Experiments were normally conducted at a minimum of three vehicle speeds (typically 25, 35 and 45 mi/h) and a minimum of 100 vehicle passes were made for each speed. Only passenger cars were utilized to generate dust emissions from unpaved roads. Wind speed and direction were continuously monitored at a nearby meteorological station (approximately 2 miles away).

During the summer of 1988, the experimental design was modified slightly for the final four experiments. A fourth dichotomous sampler was added to the exposure profiler, increasing the height of the tower to 6.1 meters. An additional set of measurements was made at a vehicle speed of 55 mi/h, additional measurements of wind speed at the experimental site at a height of 10 meters were made and more extensive sampling to characterize the road silt content (see above section) was performed.

High volume and dichotomous samplers were calibrated prior to going out in the field to perform experimental measurements. The high volume samplers were calibrated using the Andersen variable orifice calibrator and standard calibration procedures detailed by Andersen's documentation. Dichotomous samplers were calibrated using a Kurz mass flow meter.

Filters used to obtain samples using both high volume and dichotomous samplers were conditioned and weighed in a constant humidity glove box. Filters were conditioned for 24 hours or more prior to weighing. All dichotomous filters were weighed on a microbalance. Some work was performed to try and determine the uncertainty due to adsorption of moisture onto the filters. Blank filters and control filters were weighed at regular intervals as was a nonadsorbing metallic wire. Calibration of the balance was performed prior to weighing sets of filters for the experiments. The blank filters were of two types. Filter blanks were blank filters that were never exposed to field conditions. Field blanks were blank filters taken out in the field and exposed to ambient conditions, but never had air filtered through them. Both types of blanks were maintained for the chemical analysis that was intended for the filter samples. However, only approximately 35 filter samples (including blanks) were analyzed using XRF and NAA.

During most of the field experiments, a Particle Measuring Systems optical particle size measurement system was utilized to determine the particle size distribution generated by vehicle traffic on unpaved roads. Readings were made over four channels (four different particle size ranges) approximately every 45 seconds during the experiments. Output was stored on floppy disks using a computer program to collect the data. Following collection of the raw data, the data were reduced in a spreadsheet program to determine average conditions during the experiment. While this data is important, some caveats must be made regarding the potential use of the information. First, since only one device was available, it was operated at different downwind positions during the various experiments. Second, the flow orifice has not been extensively characterized and there are some questions concerning orientation of the inlet and the use of such a device (whose basis for calibration is spherical particles) to measure platelet or irregular shaped particles. Still, this is one of the few times such a device has been operated next to a fugitive dust source, and the relative distribution of the particles would provide some interesting insights into the fraction of the total particulate amount (at least based on size) that is less than 10 μm.

The results of the field experiments were analyzed using both upwind/downwind techniques and exposure profiling techniques. This represents the only set of experiments that have tried to utilize both techniques to determine an emission factor. In addition, multiple regression analysis was performed on various functions of the silt content and vehicle speed. The results of this analysis suggested that two different models had the same statistical validity. The first model found that the emission factor was linear with respect to the silt content and vehicle speed. The second model found that the emission factor was related to silt content and vehicle speed by a power function. Each model only accounted for approximately 50 percent of the variability in the measured emissions.

Further investigation of the data following the field experiments conducted during the summer of 1988 indicated that other parameters measured during those experiments could be important in predicting the emissions from unpaved roads. ISWS investigators found that the predicted emissions could explain 92 and 93 percent of the variance if either H (the mass of material < 50 μm per unit area of the road surface) or P (the percent of millimeter sized particles in the tire track) were included in the predictive equations developed using multiple regression analysis. With respect to the equation containing P, the silt content was also required in the equation. However, with respect to H, only the velocity and H were required in the predictive equation. It is interesting to note that Cowherd and Englehart (1985) showed that the best model for predicting PM-10 emissions from unpaved roads is one that incorporates the silt loading (mass of material < 75 μm per unit area of the road surface), the vehicle weight and the vehicle speed rather than the current AP-42 emission factor that depends upon the silt content (percent). Indeed, Cowherd and Englehart stated:

> "Based on a comparison of precision factors, it is apparent that the model incorporating silt loading to characterize the amount of surface material available for entrainment, provides better estimates of IP and PM-10 emission factors than do the alternative models that use silt percent."

The ISWS H term and silt loading are fairly comparable with the only difference being in the particle size cutoff.

CHAPTER 4: RELATIONSHIPS BETWEEN HIGHWAY DERIVED PARTICULATES AND OTHER PARTICULATE SOURCES

ASSESSMENT OF THE RELATIVE MAGNITUDE OF EMISSIONS FROM HIGHWAY PARTICULATE SOURCES

One method of determining the relative contributions of the various highway particulate sources to the atmosphere is to examine either their emission rates (emission factors) or to examine their total emissions. This subsection examines the relative emission rates for tire and brake wear, road sanding and salting, unpaved road emissions, and paved road resuspension. In addition, total U.S. emissions estimates for the period 1985-1988 for these same sources of particles are examined and compared to the total U.S. emissions for the same period from point and process fugitive emissions and emissions from vehicle tailpipes.

Relative Importance Based on Emission Rates

It is possible to evaluate the relative importance of the various highway particulate sources listed above by examining their emission rates, as long as the emission rates are based on their activity data (i.e., VMT). Since VMT is the activity data required to produce emissions estimates from the highway particulate sources listed above, the relative magnitude of emissions can be assessed. Figure 7 presents information concerning the emission rates (emission factors) for tire wear, brake wear, sanding and salting, paved road resuspension, and unpaved roads. All values are given in lbs/VMT. It is evident from figure 7 that emissions from brake wear and tire wear would be 2-4 orders of magnitude lower than emissions from road sanding and salting or paved road resuspension given the same level of activity. Comparably, emissions from unpaved roads would be 1-2 orders of magnitude higher than road sanding and salting or paved road resuspension, and 4-5 orders of magnitude higher than emissions from tire or brake wear for the same activity levels.

Relative Importance Based on Emissions Estimates

Unfortunately, the activity levels for these sources are not always comparable, thus a more accurate reflection of the relative importance of these sources can be obtained by examining their emissions.

Each year the EPA develops emission estimates for the criteria pollutants and publishes them in what is commonly known as the "Emission Trends" report. The most recent of these reports contains emission estimates for various fugitive dust PM-10 sources including agricultural tilling, construction activities, wind erosion, unpaved roads, mining and quarrying activities, paved road resuspension, and burning. In addition, emissions estimates for PM-10 from point (i.e., stacks), area (i.e., roof vents or leaky valves) and mobile (i.e., tailpipe) sources are also included in the report (EPA, 1996).

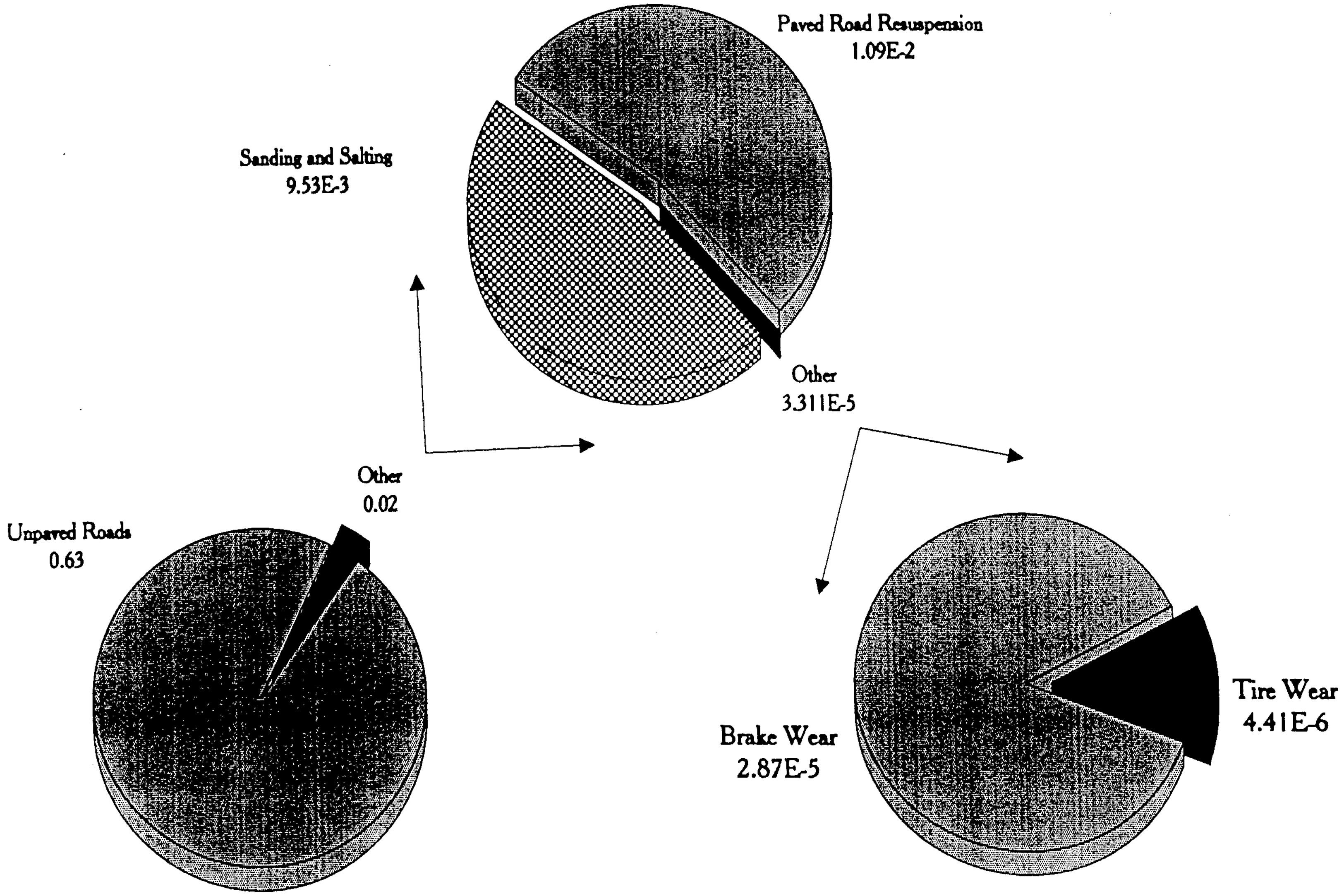


Figure 7. Emission factors for various highway particulate sources.

Barnard and Carlson (1990) evaluated the feasibility of developing PM-10 emissions estimates from fugitive dust sources for inclusion in the Emission Trends report. That study included emissions estimates from road sanding and salting which are not presently included in the Emission Trends report.

Figure 8 shows national emissions estimates of PM-10 from point and process fugitive emissions sources for the period 1985-1988. PM-10 emissions from these sources range between approximately 5 to 5.5 million short tons. Figure 9 shows emissions for the same time period from tailpipe, tire and brake wear (i.e. primary highway particulate emissions). The emissions from this source are between 1.2 and 1.3 million short tons per year. Thus emissions of primary highway particulates nationally are approximately a factor of 4 less than those from "facilities".

Figure 10 shows PM-10 emissions from paved road resuspension. This figure shows that PM-10 emissions from this highway source are slightly higher than those from point and process fugitive emissions sources and, as a consequence, are also approximately a factor of 4 to 5 higher than primary highway particulate emissions.

Figure 11 presents emissions from road sanding and salting for the period 1985-1988. These emissions are much lower than those from the other sources examined so far. Notice that the emissions scale is thousands of short tons rather than millions of short tons. This difference is primarily due to the seasonal nature of these emissions, however, the emission factor utilized to develop these emissions estimates has a large degree of uncertainty associated with it.

Figure 12 displays the PM-10 emissions from unpaved roads. These emissions range from approximately 14-16 million short tons per year, which is a factor of 10 higher than emissions from point and process fugitive sources and a factor of 3 higher than paved road resuspension.

Relative Importance Compared to Other Fugitive Dust PM-10 Sources

The U.S. EPA (1996) develops PM-10 emissions estimates for agricultural tilling, wind erosion, and other fugitive dust sources. National emissions from these sources are shown in table 7 for the 1985-1995 period.

Table 7. National PM-10 Emissions from Other Fugitive Dust Sources
(millions of short tons)

Source	1985	1986	1987	1988	1989	1990	1991	1992	1993	1994	1995
Wind Erosion	4.0	10.3	1.6	18.1	12.1	4.4	10.1	4.6	2.0	2.6	2.2
Agricultural Tilling	6.8	6.9	7.0	7.1	6.9	7.0	7.0	6.8	6.8	6.7	8.0
Other Fugitive Dust Sources	13.0	12.1	12.5	12.0	11.7	10.4	10.0	10.9	11.4	12.8	10.3

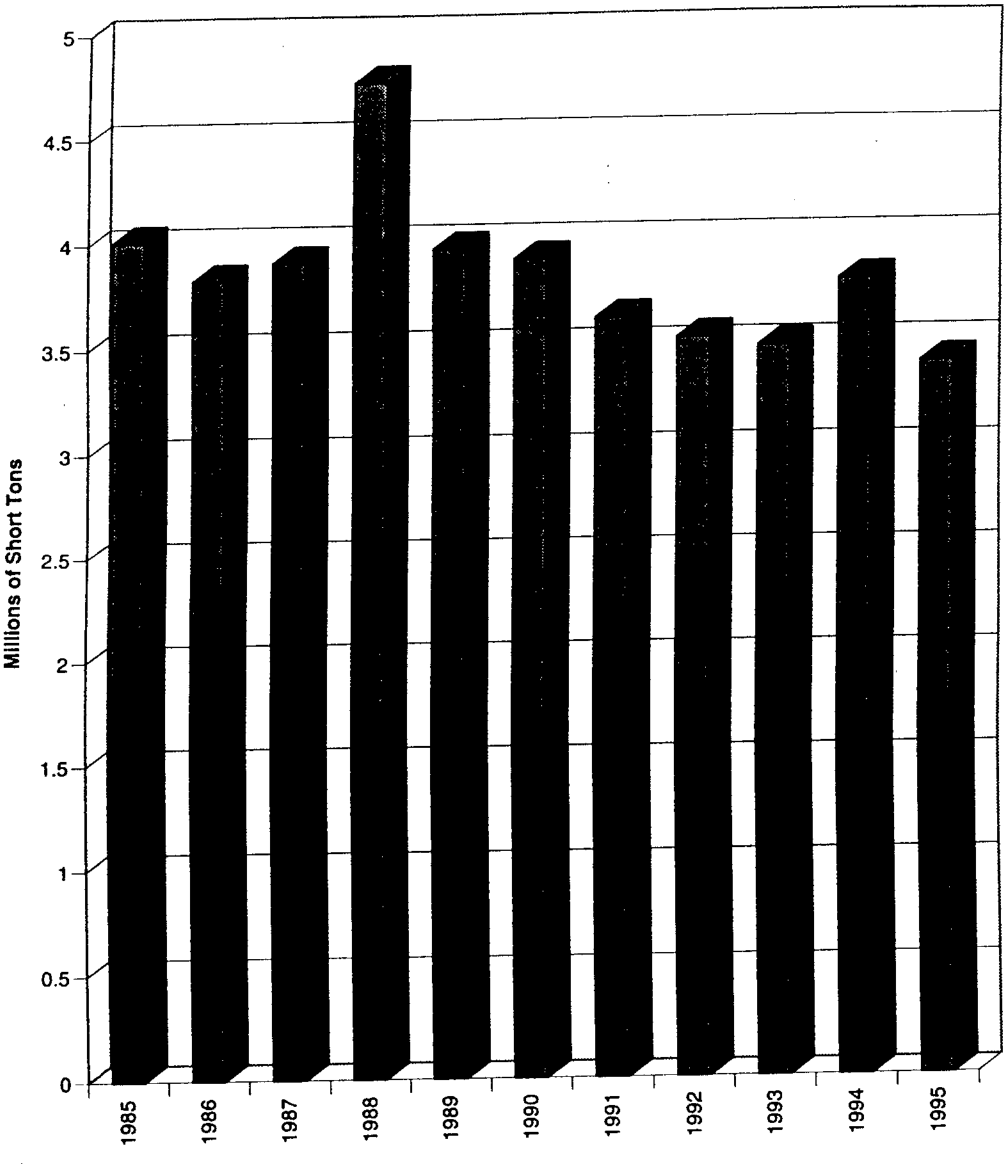


Figure 8. National estimates for point and non-highway area source PM-10 emissions for 1985-1995.

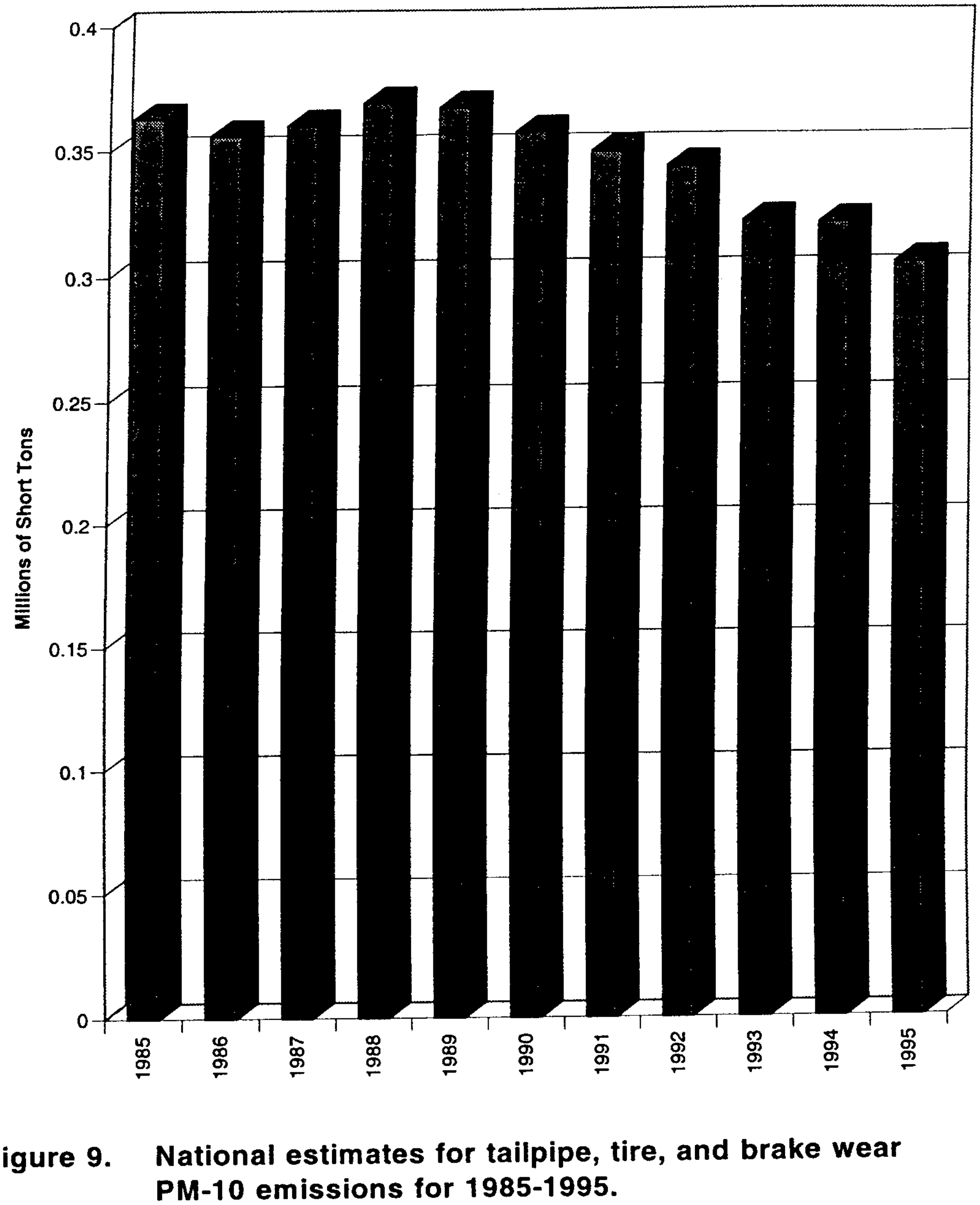


Figure 9. National estimates for tailpipe, tire, and brake wear PM-10 emissions for 1985-1995.

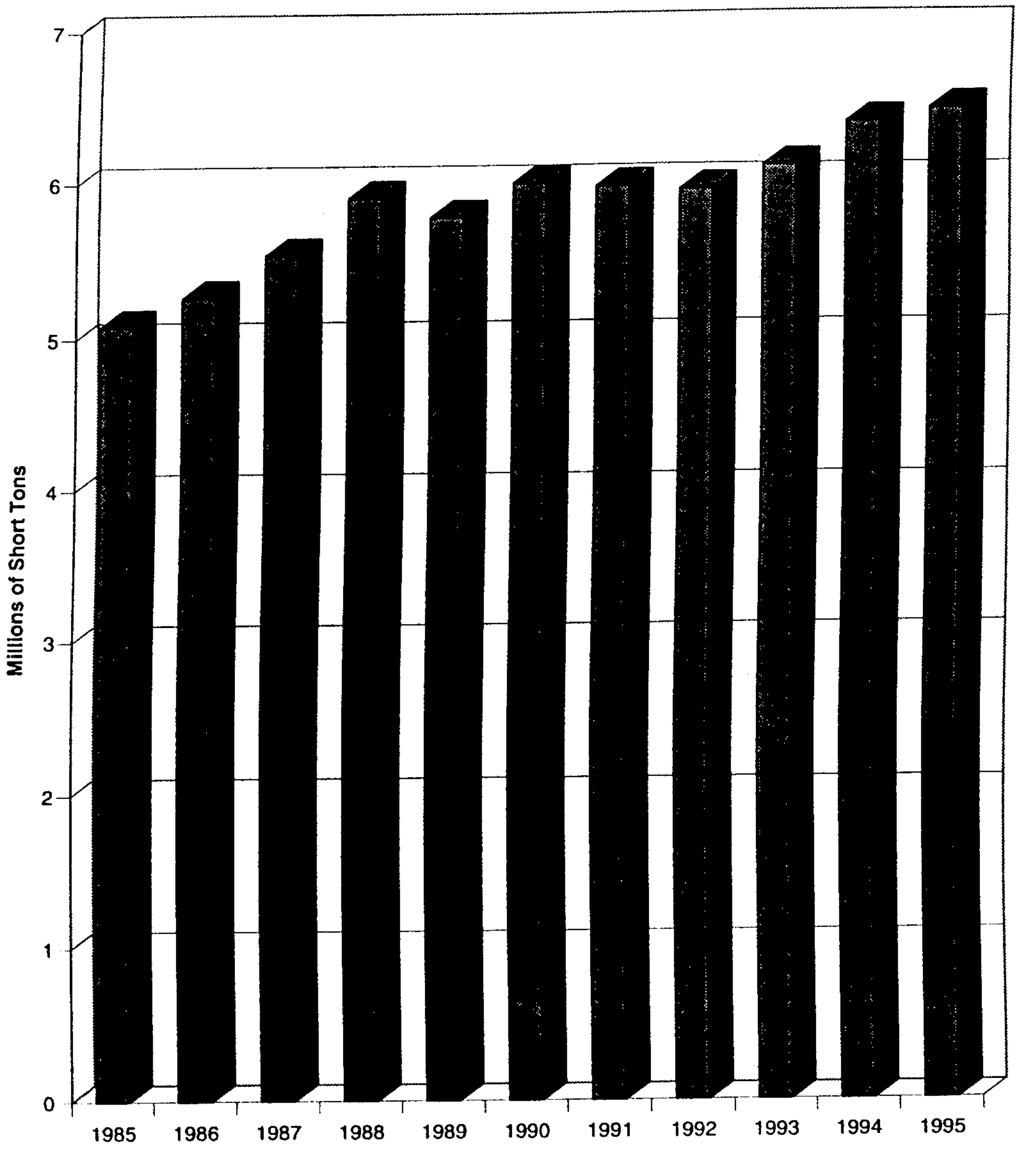


Figure 10. **National estimates for paved road resuspension PM-10 emissions for 1985-1995.**

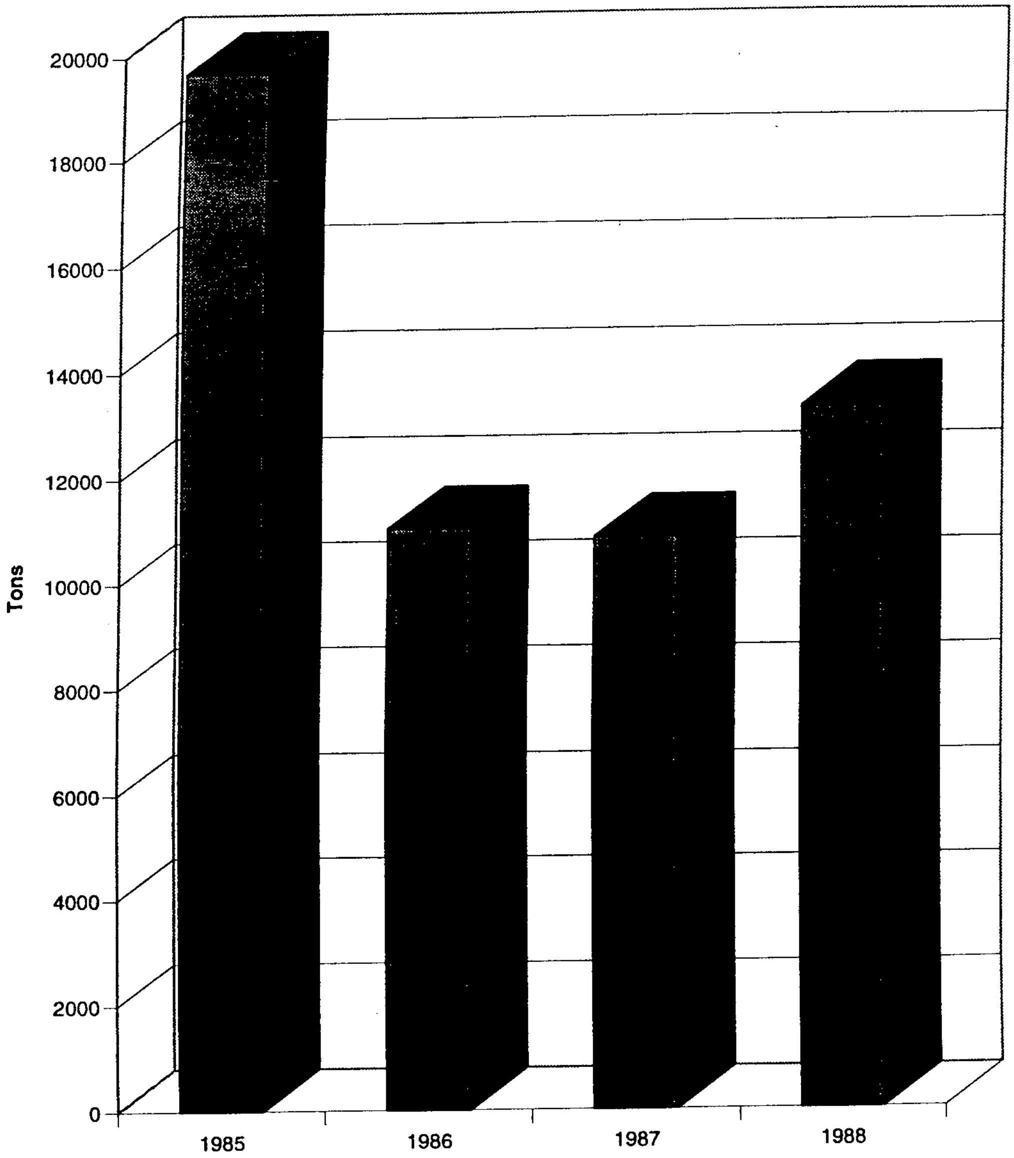


Figure 11. National estimates for road sanding and salting PM-10 emissions for 1985-1988.

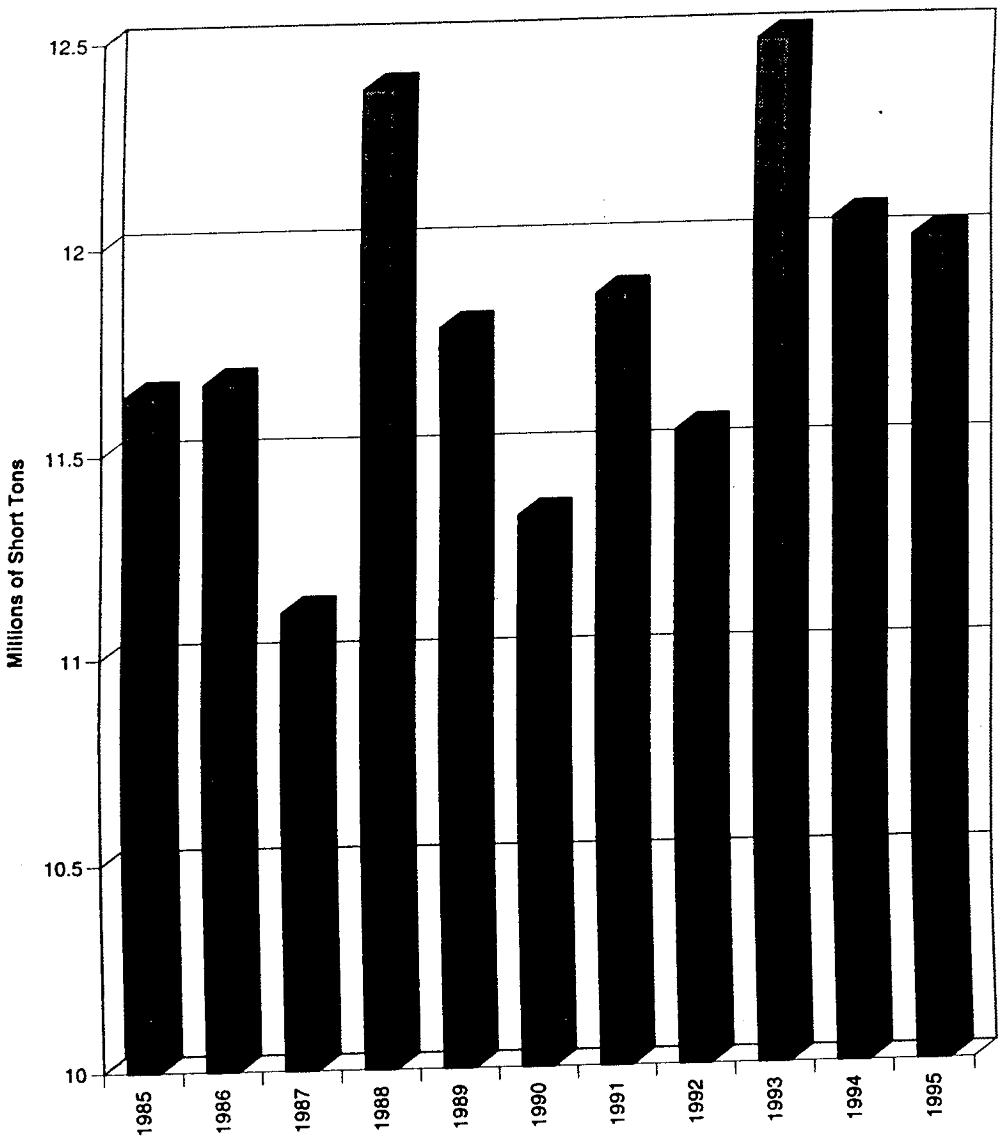


Figure 12. **National estimates for unpaved road PM-10 emissions for 1985-1995.**

Comparison of the values in table 7 with the levels indicated for the highway particulate sources in figures 9-12 clearly indicates that unpaved road emissions are equal to or greater than emissions from wind erosion and all other fugitive dust sources (primarily construction activities) and that emissions from paved road resuspension is of approximately the same level as agricultural tilling and approximately the same as the average wind erosion emissions for the 10-year period considered.

EXCEPTIONAL/NONRECURRING EVENTS AND HIGHWAY CONSTRUCTION MAINTENANCE

Exceptional or nonrecurring events have typically been dismissed from evaluation as to whether a measured exceedance of the PM-10 NAAQS should be included for averaging purposes. This is especially true in the southwestern United States, where dust devils or frontal dust storms occur on a seasonal basis, however, exceedances caused by such events are not counted towards the short-term or long-term averages. These events qualify as exceptional, even though they recur on an irregular schedule.

Construction activities may or may not be treated as nonrecurring events. It is conceivable that measurements at a receptor resulting in an exceedance value as the result of construction activities may be counted in the averaging process. However, construction emissions are truly temporary in nature. Typically, many regulatory agencies adopt a set of regulations to control the processes resulting in emissions from construction activities and write these regulations into a construction permit. Thus, they have effectively implemented a control strategy for these sources, despite the temporary nature of the emissions source.

CHAPTER 5: PARTICULATE INSTRUMENTATION, MEASUREMENT, AND MONITORING METHODS

AMBIENT SAMPLING METHODS

This section describes the methodology required to obtain the necessary chemical data required as input for the receptor modeling approaches detailed in the previous section. These techniques are based upon those that have been applied in recent particulate source apportionment studies performed throughout the United States. Different alternatives for sampling and analysis are examined in the sections which follow.

Sampling Frequency and Duration

Sampling frequency and duration result as much from technological and budgetary constraints as they do from scientific need. For the San Joaquin Valley Air Quality Study/Atmospheric Utilities Signatures-Predictions Experiment (SJVAQS/AUSPEX), Blumenthal et al. (1989) specify four periods per intensive sampling day of 7-hour duration for nighttime and 5-hour duration for daytime. Even for remote sites, the majority of summertime concentrations exceeded the minimum detectable concentrations which can be measured. In general, substantial flexibility with respect to sample duration must be built into the sampling system in order to reconcile field measurements with modeling needs.

Particle and Gas Sampling Methods

This subsection describes many of the current sampling methods for PM-10 and PM-2.5 aerosols and important trace gases which may lead to particulate formation. Sampling requirements for assessment of particulate impacts are: 1) well-defined size fractions (PM-2.5 and PM-10); 2) accurate measurements of species as they would appear in the atmosphere; 3) suitability for application in both urban and nonurban environments; 4) multiple substrates for a variety of analyses; 5) flow rate stability; 6) compatibility with analysis methods; and 7) availability of equipment and ease of operation. The alternatives for the major components which can be considered are outlined below.

Size-Selective Inlets

PM-2.5 consists of particles which are less than 2.5 µm in aerodynamic diameter while PM-10 consists of particles which are less than 10 µm in aerodynamic diameter. These size ranges are operationally defined by inertial separation inlets which transmit 50 percent of all suspended particles with 2.5 and 10 µm diameters. Smaller particles penetrate these inlets with higher sampling effectiveness and larger particles penetrate with lower effectiveness. Sampling effectiveness as a function of particle size is determined from wind tunnel tests (Federal Register, 1987b).

Six PM-10 size-selective inlets have been developed and tested (Watson and Rogers, 1986a; 1986b). These are:

- **The Wedding High Volume (hi-vol) Inlet**. Operating at 1132 L/min (40 ft^3/min), this inlet is cylindrically symmetrical to eliminate wind directional dependence (as are all of the PM-10 inlets described here) and incorporates an inner, coaxial cyclone as the main fractionating device; the cyclone's inner surface is lined with porous material impregnated with oil (Wedding and Weigand, 1985).

- **The Sierra-Andersen High Volume (hi-vol) Inlet** (retrofitted SA-321A or SA-1200). Operating at 1132 L/min, the SA-1200 inlet is similar to the earlier and discontinued Models SA-321 and SA-321A, which incorporated stages of opposing jets, to steepen the sampling effectiveness curve and to further reduce the transmission of particles 20 µm in size and larger (McFarland et al., 1984). The SA-1200 contains a greased impaction surface to mitigate against the re-entrainment of large particles. A retrofit kit is available for SA-321 and SA-321A inlets which inserts a greased plate below the impaction jets.

- **The Wedding Medium-Flow (med-vol) Inlet**. This omni-directional cyclonic inlet operates at 113 L/min (4 ft^3/min). The cyclonic particle removal mechanism is similar to that of the Wedding high volume inlet. Large particles deposit on an inner tube (Wedding and Carney, 1983).

- **The Sierra-Andersen Medium-Flow (med-vol) Inlet** (SA-254). This inlet is designed to operate at 113 L/min. Opposing jets are the primary fractionating device (Olin and Bohn, 1983). This inlet was used for PM-10 sampling in the ARB's SCAQS.

- **The Wedding Low Volume (low-vol) Inlet**. This inlet is designed to operate at 18 L/min and is similar in concept to the Wedding hi-vol inlet, except that the central, coaxial cyclone does not utilize an oiled surface in this case (Wedding and Weigand, 1982).

- **The Sierra-Andersen Low Volume (low-vol) Inlet** (SA-246). This inlet is designed to operate at 16.67 L/min, is cylindrically symmetrical, and utilizes an acceleration jet issuing into an impaction chamber as the main fractionating device (Andersen Samplers, Inc., 1985).

An intercomparison by Purdue et al. (1986) used various combinations of greased and ungreased impaction surfaces and cleaned and uncleaned inlets. The SA-321A yielded up to 15 percent higher PM-10 concentrations when compared to samples taken through the Wedding inlet, even when both samplers were clean and greased.

Several types of sampling inlets have been developed to sample fine particles in the zero to 2.5 µm size range (Watson and Chow, 1987). These devices differ from one another with respect to their specified flow rates, steepness of sampling effectiveness curves (Watson et al., 1983), degree of characterization, operating principle, and commercial availability. These size-selective sampling inlets and their relevant features are:

- **Sierra-Andersen Slotted Impactor**. This cascade impactor operates at 1132 L/min and has well defined size cuts, one of which is in the neighborhood of 2.5 µm (Willeke, 1975). It requires slotted fiber filters between stages, though it has been used in a single stage mode to sample fine particles on 8 x 10 inch filters (Watson, 1979). These impactors are commercially available, but they are expensive.

- **Bendix Unico 240 Cyclone**. This cyclone has been shown to have a sharp cutpoint at 2.5 µm (Chan and Lippman, 1977; Mueller and Hidy et al., 1983) in laboratory tests and operates at 113 L/min. It is the most widely used of fine particle cyclones, having seen service in the SURE, PACS, WRAQS, SCENES, SNAPS, SCAQS, 1978 Denver Winter Haze Study, the 1987-88 Metro Denver Brown Cloud Study, and many other aerosol studies. These cyclones are commercially available at a cost of a few hundred dollars apiece. Methods to coat these cyclones with PFA Teflon to minimize nitric acid losses have been perfected.

- **Bendix Unico 18 Cyclone**. This cyclone has a sharp cutpoint at 2.5 µm as measured by Chan and Lippman (1977). It operates at 18 L/min and is slightly less costly than the Bendix Unico 240.

- **Air Industrial Hygiene Laboratory (AIHL) Cyclone**. This cyclone was developed by Dr. Walter John of the Air Industrial Hygiene Laboratory to operate at 22 L/min (John and Reischl, 1978). It has a sharp cutpoint near 2.5 µm as shown in laboratory tests. It is not currently manufactured commercially, though it has been used in the RESOLVE visibility study, the IMPROVE visibility network, and the SCAQS. These cyclones can also be coated with PFA Teflon to minimize nitric acid losses.

- **Virtual Impactor**. The most commonly used virtual impactor is the Sierra-Andersen 241 (also in SA-244 and SA-245 models). This design has a sharp cutpoint at 2.5 µm with flow rates of 15 L/min through the fine particle filter (Mueller and Hidy et al., 1983). Virtual impactors are commercially available, but they are very expensive. Both fine and coarse samples have been measured with these units in many programs, including EPA's Inhalable Particulate Network (Watson et al., 1981), the Harvard Air Pollution Respiratory Health Study (Chow and Spengler, 1986), and the ARB dichotomous sampler network (Watson et al., 1987).

- **Stacked Filter Unit**. Large pore Nuclepore filters have been placed in series to eliminate particles larger than 2 or 3 µm in diameter (Cahill et al., 1977; Heidam, 1981). The sampling effectiveness curve is very broad at 10 L/min. These units have been used in numerous studies in California and in the Western Fine Particle Network by researchers at the University of California at Davis. This unit can be constructed from Nuclepore filters and Nuclepore filter holders.

- **University Glassware Impactor**. This inlet has been developed for annular denuder systems, though it could also be adapted to other applications (Baxter and Lane, 1987). It consists of a PTFE Teflon-coated glass jet containing a glass plate and operates at 16.7 L/min. The inlet was especially designed to minimize nitric acid removal.

An intercomparison study (Mathai et al., 1990) compared fine particle measurements from the virtual impactor, the AIHL Cyclone, and the Bendix Unico 240 cyclone. Concentrations measured with each inlet were found to be comparable for simultaneous samples. Sampling effectiveness curves for each of these inlets are also nearly identical to each other. For all practical purposes, these three PM-2.5 inlets yield the same particle size fractions.

Flow Rate Measurement and Control

At this time, four general methods of sampler air flow control are used in air quality samplers: manual volumetric, automatic mass, differential pressure volumetric, and critical orifice volumetric.

- **Manual Flow Control**. Manual control is accomplished when the operator initializes a setting, such as a valve adjustment, and then relies on the known and constant functioning of sampler components, such as pumps, to maintain volumetric flows within specifications. Manual flow settings generally change over a sampling period as the collection substrate loads up and presents a higher pressure drop.

- **Automatic Mass Flow Control**. Mass flow controllers utilize thermal anemometers which measure the heat transfer between two points in the gas stream. To a first approximation, the heat transfer is proportional to the flux of gas molecules between the two points. Hence the mass flow controller is able to sense the flux of mass. The feedback signal from the mass flow controller is then used to modulate the rotation rate of an air mover such as a hi-vol blower. Mass flow controllers require compensating circuitry to avoid errors due to absolute temperature variations of the gas itself as well as the controller sensing probe. Wedding (1985) observes that the size-selective properties of inlets require the consistency of volumetric, rather than mass, flow properties of the atmosphere. Wedding estimates potential differences in excess of 10 percent between mass and volumetric measurements of flow rates.

- **Differential Pressure Volumetric Flow Control**. Constant pressure is maintained across an orifice (usually a valve which can be adjusted for a specified flow rate) by a diaphragm-controlled valve located between the filter and the orifice. The diaphragm is controlled by the pressure between the orifice and the pump. When this pressure increases (as it does when filters load up), the diaphragm opens the valve and allows more air to pass. Variations on this type of flow control are used in the Sierra-Andersen 244 dichotomous sampler, the old MISCO high-volume samplers, and the RESOLVE particle sampler.

- **Critical Orifice**. A critical orifice consists of a small circular opening between the filter and the pump. When the downstream pressure at the minimum flow area downstream of the orifice is less than 53 percent of the upstream pressure, the air velocity attains the speed of sound and it will not increase, regardless of changes in pressure. Critical orifices provide very stable flow rates, but they require large pumps and low flow rates (typically less than 20 L/min with commonly available pumps) to maintain the high pressure differences. Wedding et al. (1986) report a diffuser arrangement which allows recovery of over

90 percent of the energy which is normally expended in back pressure behind a critical orifice. This design allows higher flow rates to be obtained with a given pump.

Filter Holders

Filter substrates must be protected from contamination prior to, during, and after sampling. The only way to ensure this in the CADMP dry deposition network is to load filters into holders in the laboratory. These holders must: 1) mate to the sampler and to the flow system without leaks; 2) be composed of inert materials (e.g., no metal) which do not absorb acidic gases; 3) allow a uniformly distributed deposit to be collected; 4) have a low pressure drop across the empty holder; 5) accommodate the sizes of commonly available air sampling filters (e.g., 37 or 47 mm); and 6) be reasonably priced.

Polycarbonate Nuclepore filter holders have been used in previous sampling programs and are currently being used in the SCENES visibility network. These holders must be modified by widening the outlet hole, gluing the retainer ring to the threaded extender section, using multiple extender sections for filter stacking, and replacing the rubber O-ring with a Viton O-ring to prevent artifact formation. Great care must still be taken to obtain a seal of the threaded extender section with the female receptacle on a sampler. The polycarbonate material has not been tested for inertness with respect to nitric acid.

The Savillex 47 mm filter holder is made of injection molded PFA Teflon, a substance which has shown the lowest inclination to absorb nitric acid (Mitchell, 1987; Hering et al., 1988). These filter holders have a tapered extender section (called a receptacle) which can be mated to a sampler plenum with an O-ring in a retainer ring. Several grids and grid rings can be stacked within the holder to obtain series filtration. The cost is reasonable. The major disadvantages of this holder are nonuniform manufacturing tolerances (diameters can be specified within a 0.01 inch tolerance) and nonuniform porosity of the support grid.

Specifications were given to Savillex for the extender section dimensions, and these were met within the tolerances allowed by a typical sampler retainer ring. The Savillex support grid was replaced with a grid manufactured by ATEC, Inc. which was especially designed for dry deposition monitoring. This new support grid has a more uniform porosity which results in a uniform filter deposit. It also reduces the flow resistance across the filter holder.

The Electric Power Research Institute (EPRI) has sponsored tests of this filter holder under its Operational Evaluation Network (OEN) program in which ENSR Consulting and Engineering (formerly ERT) is the prime contractor. Monodisperse fluorescent droplets of 9.2 µm diameter were submitted to a filter through an inlet which was longer and of smaller diameter than that used in the CADMP sampling system. Slightly over 6 percent of the 9.2 µm particles were found to be deposited in the inlet while less than 1 percent were found on the filter holder surfaces. Particle deposition in all parts of the holder summed to less than 1 percent of the test concentration for 5.2 µm particles. These tests also showed that the inline version of this filter holder causes particles to impact on the center of the sampling substrate, rather than being homogeneously distributed.

EPRI/OEN also performed tests of filter sealing at temperatures ranging from -20 to 25°C and found that any shrinkage of the PFA Teflon components in response to temperature changes did not affect the sealing of the filters within the holder. No leaks were detected as a result of temperature changes.

Sequential Filter Sampling for PM-2.5 PM-10 and Gases

The sequential filter sampler (SFS) combines all of the characteristics described in the previous sections. A schematic is depicted in figure 13. The SFS was originally developed for the SURE project in 1977, and some variation of it has been used in many aerosol studies over the past decade (e.g., Watson, 1979; Heisler et al., 1980a, 1980b; Mueller et al., 1983; Watson et al., 1988b, 1988d; Chow et al., 1988a, 1988b, 1989a). The SFS allows air to be drawn through a size-selective inlet and through different sets of filter media. Solenoid valves are controlled by a timer switch between up to six sets of filters at pre-set intervals. A vacuum pump draws air through these filters when the valves are open.

The flow rate is controlled by maintaining a constant pressure across a valve with a differential pressure regulator. This flow rate can be adjusted from 5 to approximately 40 L/min while maintaining a total flow rate of 113 L/min through the inlet. Flow rates of 20 L/min are generally selected because they provide adequate sample loadings for analysis without overloading the filters. This flow rate is drawn through each of the two filter packs simultaneously. The remaining flow rate needed by the inlet is drawn through a makeup air port. The sequential sampling makes it unnecessary to have a person present at every sample changing interval. The PM-10 size fraction is transmitted through a Sierra-Andersen SA-254I size-selective inlet and into a plenum. The PM-2.5 is transmitted through a Bendix 240 cyclone into a separate sampling unit.

Other sampling systems have been used in recent studies, but never with results as successful as the SFS. The SCAQS sampler (Fitz and Zwicker, 1987; Fitz et al., 1989), for example, has seriously compromised the aerosol data taken in that and several other programs owing to its inadequate design. Several of its flaws were identified by DRI before it was built, but these warnings were ignored during its construction in the rush to complete it in time for SCAQS field sampling.

Fujita and Collins (1989) show that the use of in-line metal filter holders in this sampler resulted in a large spot in the center of the filter. Measurements made on sections of the filter near its edge are several times lower than those made at its center. The filter holder has since been modified for recent samples taken with the SCAQS sampler in the VAQS (Valley Air Quality Study). Though the homogeneity of the deposit was improved, there were still visible differences in particle deposits on different sections of the filters. Large pieces of a brass fitting in the sample line were found on the filter deposit which compromised the mass, copper, and zinc measurements.

The SCAQS sampler also requires the unloading and loading of samples in the field which resulted in a high potential for contamination. Many of the VAQS samples had to be flagged as damaged in the field. Since the SCAQS sampler has no sequential capability, all samples must be changed manually between each sampling interval. This makes the sampler very labor intensive to operate for diurnal sampling.

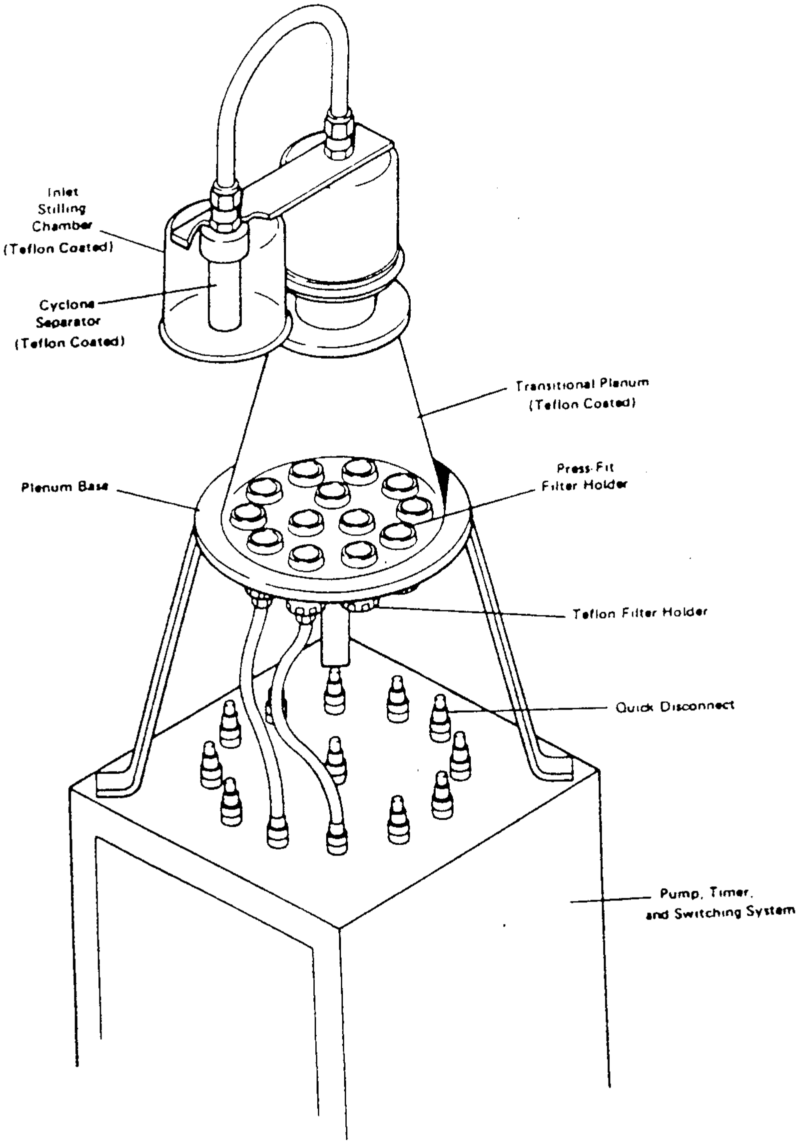


Figure 13. Schematic of a sequential filter sampler.

For all these samplers multiple filter packs are sampled simultaneously. In a typical situation, one filter pack consists of a Teflon-membrane filter followed downstream by a quartz filter. The quartz is separated from the drain disk to minimize transfer from one filter to another. The Teflon filter removes particles for mass and elemental analyses. The quartz backup filter is intended to quantify organic vapor absorption which has been found to interfere with OC measurements. Mueller (1988) recently showed that this system provides a good correction for this artifact. Mueller (1988) confirmed that quartz-fiber denuder systems are unnecessary when this configuration is used.

A second filter pack may consist of a quartz-fiber filter followed downstream by a nylon filter. The deposit on the quartz filter is submitted to ion and carbon analyses. The nylon filter allows the magnitude of volatilized particle nitrate to be determined and added to the nitrate measurement on the front filter to obtain total particulate nitrate. A set of nitric acid denuder tubes may be used to remove gaseous nitric acid from the airstream. Watson et al. (1988b) show that these tubes remove more than 95 percent of gaseous nitric acid from the airstream.

Filter packs are loaded in the laboratory and shipped under refrigeration to and from the field. This not only saves operator time, which will be in great demand at these complex sites, but it minimizes contamination of filters which always occurs when they are loaded and unloaded from filter holders in the field.

Tests conducted at the beginning of the SCENES program showed that at high ambient temperatures (above ~35 °C) the quartz-fiber filters could absorb organic material from the rubber O-rings on Nuclepore filter holders. The rubber O-rings in SCENES were replaced with Viton O-rings, a copolymer of vinylidene fluoride and hexafluoropropylene. Viton O-rings were found to be inert with respect to organic absorption on quartz-fiber filters. The IMPROVE network replaced all Nuclepore rubber O-rings with Viton O-rings for carbon sampling since its commencement in 1987. The Nuclepore filter holders have been modified by drilling the exit hole to 3/8 inch diameter. This is necessary to achieve the desired flow rates. The retainer ring recesses were also machined to accommodate the thickness of multiple filters.

Each SFS is serviced by: 1) verifying and recording post-sampling flow rates; 2) recording elapsed sample time; 3) removing exposed filters; 4) installing unexposed filters; 5) verifying and recording pre-sampling flow rates; and 6) resetting the timer. Inlets are cleaned every 2 weeks and pump exhaust filters are changed every 2 months.

Substrate Sampling for Trace gases and Particles

The choice of filter type results from a compromise among the following filter attributes: 1) mechanical stability; 2) chemical stability; 3) particle or gas sampling efficiency; 4) flow resistance; 5) loading capacity; 6) blank values; 7) artifact formation; 8) compatibility with analysis method; and 9) cost and availability. EPA filter requirements for PM-10 sampling specify 0.3 µm DOP (dioctyl phthalate) sampling efficiency in excess of 99 percent, weight losses or gains due to mechanical or chemical instability of less than a 5 $\mu g/m^3$ equivalent, and alkalinity of less than 25 µequivalents/g to minimize SO_2 and NO_x absorption (Federal Register, 1987a; 1987b; 1987c; 1987d; 1987e; 1987f). Summaries and experimental evaluations of several of these attributes for various filter media have been prepared by Lippman (1983).

The most commonly used filter media for atmospheric particle and gaseous sampling are cellulose-fiber, glass-fiber, Teflon-coated glass-fiber, Teflon-membrane, Nuclepore-membrane, quartz-fiber, and nylon-membrane. None of these materials is perfect for all purposes.

- **Cellulose-fiber** filters meet requirements in most categories with the exception of sampling efficiency and water vapor artifacts. Sampling efficiencies below 50 percent in the submicron region have been observed, but these are highly dependent on the filter weave. Cellulose fiber is hygroscopic and requires precise relative humidity control in the filter processing environment to obtain accurate mass measurements. This substrate has low elemental blanks and is commonly used for chemical speciation of the deposit. The hygroscopic properties of this medium make it especially amenable to filter impregnation for gaseous sampling.

- **Glass-fiber** filters meet requirements in most categories with the exception of artifact formation and blank levels (Witz et al., 1983). Sampling efficiency is very high for all particle sizes. The high alkalinity of these substrates causes SO_2, NO_x, and gaseous nitric acid to be absorbed. Blank levels for most elements of interest are extremely high and variable. Particulate nitrate and ammonium losses have been observed when these samples are stored at room temperature for long periods, but this is probably true of deposits on all types of filter media. Glass-fiber filters may exhibit volatilizable carbon artifacts.

- **Teflon-coated glass-fiber** filters meet requirements in all categories except blank element and carbon levels. Though a small nitric acid artifact has been observed, it is tolerable in most situations. These filters are excellent for ion analyses but not for carbon analyses owing to their Teflon coating.

- **Teflon-membrane** filters meet requirements in all categories except flow resistance and carbon blank levels. Because of their low porosity, it is not usually possible to attain the flow rates needed by the size-selective inlets in high volume sampling, though it is possible to obtain flow rates required for lo-vol and med-vol inlets. These filters cannot be analyzed for carbon because of its presence in the filter material, though they have very low blank levels for ions and elements. Most nondestructive multi-elemental analysis methods use Teflon-membrane filters. The deposit of particles on the filter surface makes these substrates especially amenable to XRF and PIXE analyses.

- **Nuclepore-membrane** filters have low sampling efficiencies, even for small pore sizes (Liu and Lee, 1976; Buzzard and Bell, 1980). They have low elemental blank levels. Nuclepore filters hold an electrostatic charge (Engelbrencht et al., 1980; Chow, 1985) which influences mass measurements unless substantial effort is invested in discharging them. Nuclepore filters are most appropriate for electron microscopic and elemental analyses. Ion analyses are also possible. Carbon analyses are not performed on these substrates because the filter material contains carbon.

- **Quartz-fiber** filters meet requirements in most categories and have artifact properties which are significantly lower than those for glass-fiber filters, though these substrates may absorb organic gases. Trace element blank levels are often too variable for most elemental analyses, though these filters are widely used for carbon analyses. The greatest drawback of quartz-fiber filters is their fragility, which requires extremely careful handling for accurate mass measurements. New formulations have been developed to minimize this drawback (Lundgren and Gunderson, 1975; Witz and Wendt, 1981; West, 1985; McMann, 1986). Various quartz filter formulations have been shown to have low carbon blank levels, though there is evidence that these substrates absorb organic vapors (McDow, 1986). This absorbed OC can equal 10 to 50 percent of the OC measured on these filters (McDow, 1986). Quartz-fiber filters have also been generally found to have low ionic blank levels and minimal absorption of gases such as SO_2, NO_x, and nitric acid (Mesorole et al., 1976, 1979; Pierson et al., 1976, 1983; Coutant, 1977; Appel et al., 1979a, 1984; Spicer and Schumacher, 1979; Watson et al., 1981).

- **Nylon-membrane** filters are used almost exclusively for the collection of nitric acid, although these filters were not originally manufactured for this purpose. However, there is a substantial difference between the properties of filters from different manufacturers. Nylon filters have high flow resistances, which increase rapidly with filter loading. Nylon filters also absorb substantial quantities of SO_2 with variable efficiencies (Japar and Brachaczek, 1984).

Whatman 41 cellulose-fiber filters are commonly impregnated with gas-absorbing solutions to collect gaseous ammonia (NH_3) and SO_2. Several impregnation solutions have been used to absorb gaseous NH_3 and SO_2. These solutions differ with respect to their reactive components and with respect to their formulations. The criteria which must be met by the impregnation solution are: 1) availability of pure reagents; 2) stability of the impregnation solution composition before and after impregnation; 3) low degree of hazard or toxicity; 4) lack of interferences with other pollutants being sampled or with analytical methods; and 5) minimal effects of environmental factors such as temperature and water vapor content.

Sulfuric acid (Okita and Kanamori, 1971; Knapp et al., 1986), oxalic acid (Ohira et al., 1976; Shendrikar and Lodge, 1975), phosphoric acid, and citric acid (Stevens et al., 1985) have been used as the active agent in the sampling of NH_3 on a variety of substrates. Citric acid impregnating solutions best meet the criteria described above.

As part of EPRI/OEN testing, 47 mm diameter Whatman 41 cellulose-fiber filters impregnated with 0.13 µg of citric acid and 0.024 µg of glycerine were shown to absorb more than 4,000 µg of NH_3 with better than 99 percent efficiency. Tests at temperatures ranging from -20 to 25 °C and at high and low relative humidities showed sampling efficiencies for NH_3 in excess of 99 percent. The citric acid/glycerine mixture has been selected for NH_3 impregnation in this project (Fung, 1988).

Potassium carbonate with glycerine has been used in impregnated filters for SO_2 sampling (Forrest and Newman, 1973; Johnson and Atkins, 1975; Daum and Leahy, 1985; Anlauf et al., 1985). Better than 98 percent sampling efficiencies were measured in EPRI/OEN tests of Whatman 41 filters impregnated with a solution containing 10 percent potassium carbonate and 5 percent glycerine with flow rates of 20 L/min, temperatures ranging from -20 to 50°C, and relative humidities greater than 90 percent and as low as 10 percent. Sampling efficiency degraded at relative humidities between 2 percent and

10 percent; some moisture is needed for SO_2 absorption. SO_2 concentrations were 36 and 115 ppb in these tests. The carbonate in the impregnating solution presents interferences to both the ion chromatographic (IC) and automated colorimetric (AC) analyses of extracts from these filters, however. In ion chromatography, the carbonate interferes with the nitrate peak and broadens the sulfate peak. In colorimetric methylthymol-blue analysis, the reaction of the MTB-Ba complex needs to be acidic and the carbonate raises the pH. Steps can be taken to alleviate these in the preparation of the filter extract prior to analysis.

The impregnation solutions consist of:

- 25 percent citric acid and 5 percent glycerol (balance being water) for NH_3 sampling.
- 15 percent K_2CO_3 and 5 percent glycerol solution (balance being water) for SO_2 sampling.

To impregnate filters, 47 mm diameter Whatman 41 cellulose-fiber filter disks are immersed in the impregnating solution for approximately 30 minutes. These disks are then removed and placed in clean Petri slides for drying in a vacuum oven for 5 to 10 minutes. One hundred impregnated filters are immediately sealed in polyethylene bags and placed under refrigeration for later loading into filter holders. One sample from each lot of citric acid filters is submitted to ammonium analysis prior to use. One sample from each lot of potassium carbonate filters is submitted to sulfate analysis before loading to identify SO_2 contamination. The extract is also submitted to soluble potassium analysis to determine that a sufficient quantity of potassium carbonate remains on the filter. Lots are rejected when contamination or insufficient potassium carbonate is discovered.

LABORATORY ANALYSIS OF AEROSOL/GAS SAMPLES

The aerosol chemical properties necessary for input to the receptor models can be measured by a number of different methods. Watson et al. (1988b) provide a justification for the specific measurement methods and these are summarized in this subsection. Particular emphasis is placed on particulate carbon analysis because of their nonroutine nature and importance in elucidating highway particulate impacts.

Gravimetric Analysis

In order to calculate aerosol mass loadings, unexposed and exposed filters are equilibrated at a constant temperature and relative humidity for a minimum of 24 hours prior to weighing. Weighing must be performed on an electromicrobalance with ±0.001 mg sensitivity.

X-Ray Fluorescence Analysis for Elements

X-ray fluorescence (XRF) analysis is used to obtain trace metal signatures on filters for Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Au, Hg, Tl, Pb, and U with an energy dispersive x-ray fluorescence (EDXRF) analyzer.

In XRF, inner shell electrons are removed from the atoms of the aerosol deposit. An x-ray photon with a wavelength characteristic of each element is emitted when an outer shell electron occupies the vacant inner shell. The number of these photons is proportional to the number of atoms present. The characteristic x-ray peaks for each element are defined by 200 eV-wide windows in an energy spectrum ranging from 1 to 50 KeV.

The ability of an x-ray to penetrate matter depends on the energy of the x-ray and the composition and thickness of material. In general, lower energy x-rays, characteristic of light elements, are absorbed in matter to a much greater degree than higher energy x-rays. Larger particles collected during aerosol sampling have sufficient size to cause absorption of x-rays within the particles. Attenuation factors for fine particles (particles with aerodynamic diameter less than 2.5 µm) are generally negligible (Criss, 1976), even for the lightest elements, but these attenuations can be significant for coarse fraction particles (particles with aerodynamic diameter from 2.5 to 10 µm). Correction factors have been derived using the theory of Dzubay and Nelson (1975) and are applied to measurements of PM-10 and samples.

Organic and Elemental Carbon Analysis

There is great controversy regarding the definitions of OC and elemental carbon (EC). All current analytical definitions of OC and EC are operational, that is, they depend on the specified analysis conditions and not on absolute and widely accepted standards. Different researchers have different operational definitions of OC and EC, though the data are usually reported with the same name. This inequivalence of definitions for the same identifier has caused great consternation among the users of these data. When the results labeled OC and EC from different laboratories analyzing the same samples are compared, they often differ by many times their specified precision intervals. The reality is that they should never be expected to be equivalent because they are defined differently.

There are four potential objectives which can be attained by taking organic and elemental carbon measurements in the atmosphere: 1) source apportionment; 2) estimating light extinction and light absorption; 3) comparison with other carbon measurement methods; and 4) evaluation in relation to adverse health effects. The selection of operational definitions for OC and EC depends on which objectives are most important.

The most important objective for this program is the first one, source apportionment. Carbon which evolves at different temperatures and in different atmospheres in source material should have similar properties at the receptor. These properties can be matched during the comparison of modeling results and measurements to provide further validation of the model. This objective requires several operationally defined fractions of the carbonaceous component, which have been or can be measured on particulate source emissions.

Carbon analysis methods with different operational definitions for OC and EC fall into the following categories:

- Solvent extraction of the organics followed by total carbon analysis (Gordon, 1974; Grosjean, 1975, 1985; Appel et al., 1976, 1979b; Daisey et al., 1979; Muhlbaier and Williams, 1982).

- Nitric acid digestion of the organics followed by total carbon analysis (McCarthy and Moore, 1952; Kukreja and Bove, 1976; Pimenta and Wood, 1980).

- Transmission, reflection, or absorption (as estimated by increased temperature) of light to estimate absorption and application of an assumed light absorption efficiency to estimate elemental carbon. Infrared absorbance (Smith et al., 1975), Raman spectroscopy (Rosen et al., 1978), and visible absorbance (Lin et al., 1973; Weiss et al., 1979; Gerber, 1982; Heintzenberg, 1982), and photoacoustic detection (Adams et al., 1989) are variations of this method,

- Thermal combustion including both temperature programming analysis (Muhlbaier and Williams, 1982) and step-wise pyrolysis analysis followed by oxidation using either carbon dioxide (CO_2) or methane (CH_4) detection (Mueller et al., 1971; Patterson, 1973; Merz, 1978; Johnson and Huntzicker, 1979, Johnson et al., 1979; Malissa, 1979; Cadle et al., 1980a, 1980b; Heisler et al., 1980a, 1980b; Mueller et al., 1981; Wolff et al., 1982).

- Combination of thermal combustion and light absorption optical methods (Appel et al., 1976; Dod et al., 1979; Macias et al., 1979; Cadle et al., 1980a, 1980b; Cadle and Groblicki, 198O; Johnson et al., 1981; Novakov, 1982; Rosen et al., 1982; Huntzicker et al., 1982).

Many tests have been undertaken to compare the results of one of these methods with those of other methods. Cadle and Groblicki (1980) applied several methods to quantify the OC in six ambient particulate samples. Solvent extraction yielded 30 to 50 percent of the OC measured by thermal combustion in these tests.

Stevens et al. (1982) compared elemental carbon obtained from thermal combustion and photoacoustic absorption and found the measurements correlated with a 0.8 correlation coefficient. The photoacoustic values were 75 percent of the thermal combustion values for samples taken in Houston, but were 21 percent of the thermal combustion values for samples taken in the Shenandoah Valley. Stevens et al. (1982) also compared the results of the two different thermal combustion methods applied by two separate laboratories to 28 ambient particulate samples. They reported average values of 1.89 and 3.52 $\mu g/m^3$ for OC and 0.85 and 0.37 $\mu g/m^3$ for elemental carbon, respectively, between the two laboratories. These differences were attributed to variations in the thermal combustion analyzers and different analytical procedures.

Johnson (1981) compared elemental carbon derived from Oregon Graduate Center's (OGC) thermal/optical reflectance (TOR) method with light transmission analyses and found the average ratio of thermal combustion to light absorption to be 0.88 ± 0.06 for samples from six sampling sites. Watson (1979) reports results of total carbon measured by the OGC TOR method, by the Oregon Department of Environmental Quality (ODEQ), and by the EPA at Corvallis, Oregon (Watson, 1979; Cooper et al., 1979a, 1979b). The results of this interlaboratory comparison indicated a reasonable equivalence between the EPA and OGC measurements (slope = 1.02, correlation = 0.907) for total carbon.

Comparison with ODEQ yielded a higher correlation of 0.990, but with a linear regression slope of 0.83, with the OGC values as the dependent variable. Johnson (1981) and Shah (1981) report comparisons of OGC, ODEQ, and General Motors Research Laboratory (GMR) thermal combustion analyses of 10 ambient samples. A high correlation was found among total carbon values; however, the slopes of GMR/OGC were 1.46 for OC and 0.74 for EC.

Heisler et al. (1980a; 1980b; 1980c; 1980d) and Mueller et al. (1983) compared carbon as measured by the GMR and Environmental Research and Technology (ERT) thermal combustion methods using 151 filter samples taken in Denver, CO, with an average loading of 208 µg carbon per 47 mm quartz-fiber filter. High correlations (0.96 to 0.99) were found, though GMR's elemental carbon results were systematically higher than ERT's measurements. Cadle and Groblicki (1982) applied many different carbon analysis methods to the same samples. Their agreement was best for total carbon (organic plus elemental), the results ranging from -15 to +26 percent of the mean, and worst for elemental carbon, ranging from -57 to +179 percent of the mean.

The most recent attempt to determine the equivalence of different carbon analysis methods was the CARB Carbonaceous Species Methods Comparison Study (CSMCS) which took place in 1986 (Hering et al., 1989; Lawson and Hering, 1989). A nine-day field study was conducted on the campus of Citrus College, Claremont, CA, and a set of "reference" aerosol samples were distributed to 13 participating laboratories. These samples included total suspended particulate, ambient PM-10 in triplicate, automotive exhaust, woodsmoke, organics (photolysis of trimethylbenzene), and blank quartz filters. Thermal, TOR, thermal/optical transmission, and optical absorption techniques were used. For total carbon, most laboratories were in agreement within ±10 percent for all samples. The coefficient of variation on the triplicate PM-10 analysis ranged from 0.7 to 40 percent from laboratory to laboratory.

This good agreement among different methods for total carbon shows that most laboratories are equivalent for this species. The major differences in the CSMCS were found for the OC and EC fractions, especially for automotive- and woodsmoke-dominated reference samples (Countess, 1988; 1989).

Thermal/Optical Reflectance (TOR) Carbon Analysis

Combustion methods are most commonly used in monitoring networks with many samples. The major differences in the fractions of OC and EC reported by the different combustion methods are functions of several attributes of the analysis: 1) calibration standards; 2) combustion temperatures; 3) temperature ramp rates and duration; 4) pyrolysis suppression or correction; 5) oxidation methods; and 6) combustion atmospheres. Substantial effort has been invested in customizing the thermal/optical reflectance carbon method to meet the needs for light extinction budgets, source apportionment, and interlaboratory comparison.

The principle of the TOR method is the thermal desorption of carbon compounds with a correction for pyrolysis of organic species based on laser reflectance from the deposit surface. The detector is a flame ionization detector with a hydrogen flame. The optical signal is provided by a 10 mW He/Ne laser at a wavelength of 632.8 nm. The evolved carbon is converted to carbon dioxide (CO_2) by a powdered MnO_2 oxidation catalyst heated to 912°C. The CO_2 is converted to methane by a nickel catalyst on firebrick matrix, heated to 650°C.

Combustion for OC takes place in an atmosphere of helium (He), scrubbed of oxygen and hydrocarbon impurities. Combustion for elemental carbon takes place in an atmosphere of oxygen in helium (O_2/He), nominal 2 percent O_2.

Ion Chromatographic Analysis for Chloride, Nitrate, and Sulfate

Anion chromatography for chloride (Cl^-), nitrate (NO_3^-), and sulfate (SO_4^-) is commonly performed with the Dionex (Sunnyvale, CA) ion chromatograph. In IC, an ion-exchange column separates the sample ions in time for individual quantification by a conductivity detector. Prior to detection, the column effluent enters a suppressor column where the chemical composition of one element is altered, resulting in a matrix of low conductivity. The ions are identified by their elution/retention times and are quantified by the conductivity peak area.

Atomic Absorption Spectrophotometric Analysis for Sodium and Potassium

Atomic absorption is generally used to analyze filter extracts for soluble potassium and sodium. A dual hollow cathode lamp emits wavelengths appropriate for sodium and potassium analyses. For potassium, the monochrometer is set at 766.5 nm with a 2.0 nm bandpass. For sodium, the monochrometer is set at 589.0 nm with a 0.7 nm bandpass.

Automated Colorimetric Analysis for Ammonium

An automated colorimetric system is commonly used to measure ammonium concentrations by the indophenol method. Ammonium in the extract is reacted with phenol and alkaline sodium hypochlorite to produce indophenol, a blue dye. The reaction is catalyzed by the addition of sodium nitroprusside. The absorbance of the solution is measured at 630 nm.

CHAPTER 6: ASSESSMENT OF POTENTIAL PUBLIC HEALTH RISKS

INFLUENCE OF MOTOR VEHICLE PARTICLE EMISSIONS ON PUBLIC HEALTH

Particle emissions have been of concern with regard to the presence of inhalable particulate material (PM-10) and lead metal in the environment. EPA (1979) has published criteria for protection of the public health from inhalable particles and for lead. Many emissions control actions, such as phase-out of lead antiknocks in gasoline, diesel passenger car and heavy duty engine particle emissions standards, have been justified mainly on their beneficial effects in achieving safe levels of these pollutants in air. The health effects described in the criteria document for lead neurological disfunction in young children where immature brain membranes allow the passage of lead salts causing possibly irreversible damage. For inhalable particles (e.g. those with diameters < 10 µm), the effects to be expected include general irritation and induction of asthma episodes in particularly susceptible individuals. In both these cases, concern over these particular public health risks has resulted in aggressive regulatory action to limit or eliminate the emissions of these pollutants.

Recent reviews of diesel particle emissions have summarized findings from several U.S. and European studies in which animals were exposed to high levels of particles for extended periods of times. At the very highest levels of exposure, lung tumors were observed in rats. However, it is not clear whether these tumors were caused by exposure to mutating genotoxic agents in the organic materials or by general irritation caused by the massive amounts of soot inhaled by the animals. In either case, the risk analysis previously performed may not be altered importantly by these findings.

Diesel Exhaust Particle Composition

Braddock and Bradow (1975) reported the first studies of diesel exhaust particles in which component material balances were attempted. These authors found the particles emitted from a Nissan passenger car equipped with a 2.1 liter diesel engine to consist mainly of carbon black, accounting for 85-90 percent of the mass. Transmission electron micrographs of the particles showed filament-like aggregates of spherical bodies. These spherical components had characteristic diameters of 150 to about 350 angstrom degrees, approximately the particle size range of the nuclei region particles reported by Kittelson et al. (1988). (NRC, 1982) The particles from this particular car were extremely dry, apparently because of exceptional engine oil control, and only about 10 percent of the mass was extractable in organic solvents. Still, this material was the second most important contributor to particle mass. Sulfur was found as sulfate in the particles, account for 3-5 percent of the mass. Sulfate was found to vary linearly with fuel sulfur; approximately 2 percent of the fuel sulfur was emitted in particles. Lubricant trace elements and wear products account for the final component of particle mass 1-2 percent with these samples.

The first experiments measuring heavy duty diesel engine mass and composition were conducted by collaboration of EPA and Southwest Research Institute in the 1970's (Hare, Springer and Bradow, 1976; Hare and Bradow, 1979). These authors developed methods for measuring and analyzing diesel particles using air dilution techniques (Habibi, 1970) applied to the 13-mode test procedure used for certifying heavy-duty engines at that time. Actually this procedure was quite convenient for developing emissions estimates as a function of engine speed and load. The air dilution procedures involved splitting the raw exhaust, diluting it about 10 fold with filtered ambient air, and filtering the resulting equilibrated particulate material at near room temperatures.

Figure 14 shows the variation in particle mass emission rate per unit time versus engine load for two truck engines used in the project. At peak engine speed, the range of particle emission rate is fairly small, only about a factor of 2.5 from idle to maximum emission. This same trend was also found at lower engine speeds (Hare, Springer and Bradow, 1976). Similar small variability for loads greater than about 50 percent of full rated load were also found for other engines and fuels (Hare and Bradow, 1979).

These authors studied the composition of diesel particles using a variety of fuels and fuel additive compositions as well as engine types. They discovered that particle emissions could be characterized with four major components. For four-stroke cycle engines, including Cummins and Caterpiller types, the major component of diesel particles was black, carbon-black soot accounting for 70 to 85 percent of the material identified.

Organic material could be readily extracted from the particles with low boiling nonpolar solvents such as dichloromethane. This material accounted for approximately 9 to 25 percent of the mass of particulate material. Most of this organic material was hydrocarbon and the molecular weight range of the material incorporated both the higher molecular weight components of the fuel and the engine oil boiling range material.

For the two 2-stroke cycle engines tested, a 6L-71 truck engine and a 6V-71 city bus engine, organic material accounted for more than half the particulate mass recovered. Further, the molecular weight range of this hydrocarbon material was very much greater than that found for the 4-stroke material and corresponded very closely with that of the engine oil. Further, the trace elements found in the particles were consistent with engine oil additives. Lubricant trace elements, calcium, zinc and phosphorous were found in all particle emissions samples and these accounted, collectively for about 2 percent of the particle emission rate. Since the overall particle emission rates were somewhat higher with the 2-stroke cycle engines, the emission rates of the lubricant trace elements were also elevated, proportionately.

The amount of engine oil necessary to account for these observations is 50-60 g/h or about 1 quart every 600 miles. It should be noted that this particle generation characteristic was present in both Detroit engines in these two early studies and in numerous other Detroit Diesel engines in the late 70's and early 80's. By about 1985, this oil control problem seems to have been corrected and very large engine oil contributions to atmospheric PM-10 seem to be relatively uncommon now. However, at very low particle emissions, engine oil can still be a serious concern in certification of heavy duty engines.

Lubricant is a significant contributor to particles emitted from diesel passenger car engines also. Mayer et al. (1980), operated a 5.7 liter GM passenger car diesel engine

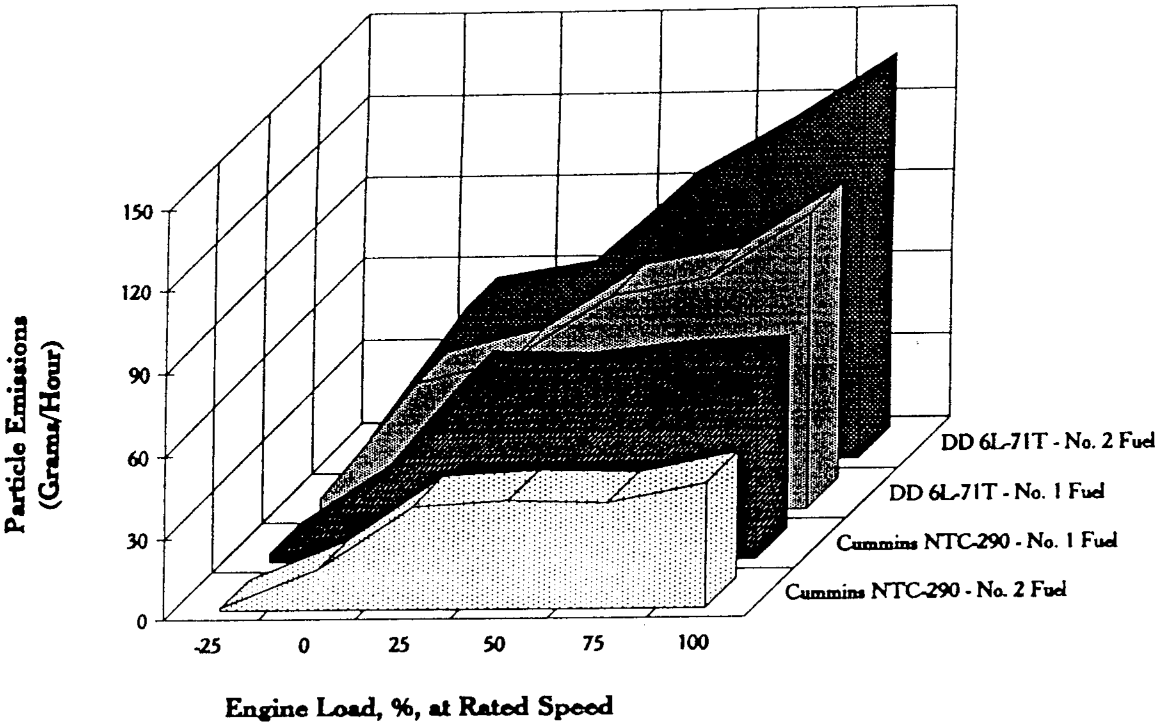


Figure 14. Particle emission rates for two diesel engines and two fuel types used to characterize particle composition.

with a lubricating oil doped with a C28 hydrocarbon radiolabeled with carbon-14. Of the radioactivity found in the particulate material, 100 percent could be recovered by solvent extraction. On the basis of the radiolabel recovery, these authors found up to 60 percent of the organic fraction could be accounted for by the engine oil.

The next most significant component of the particles was sulfate, probably in the form of sulfuric acid (Truex et al., 1986). The two fuel studies (Hare, Springer and Bradow, 1976; Hare and Bradow, 1979) found sulfate to account for about 6 percent of the particulate mass for a typical No. 2 emissions test fuel. Sulfate emission rates varied linearly with fuel sulfur content for all engines tested and accounted for about 1.5 percent of the fuel sulfur charged to the engine.

In addition to these four major components of particulate mass, carbon black, organics (mainly engine oil), lubricant trace elements, and sulfate, small amounts of other components were found which varied in a major way from engine to engine. For example, the Caterpiller engine particles contained small but detectable quantities of lead, manganese, and copper apparently associated with bearing wear. Cummins and Detroit engines also had characteristic patterns of trace metals associated with engine component materials. However, these wear products usually constitute a small portion of the particle mass, usually only 1 to 2 percent. Evidence on the character and emission rates of these four particle components is presented below.

Diesel Soot - Carbon Black Emissions

There is considerable evidence that the combustion formed soot from diesel engines corresponds very well with that from other rich flame sources. For example, Kadota and Henien (1981) reported on a study of diesel soot formation in a diesel spray simulation apparatus. The soot particles collected consisted of agglomerates of 10-40 nm spheres arranged in filaments or loose aggregates of characteristic dimension from 0.2 to 0.4 μm. Corresponding aggregates in rich flat flame burners have been described as "pearl necklaces" (Wagner, 1981). The carbon/hydrogen atom ratio for this soot increases with increasing combustion duration, reaching a maximum of about 10 near the end of combustion. The average particle size, surface area and other physical properties of diesel soot are very similar to those of carbon black from methane flames (Lahaye and Prado, 1981). These observations compare very well with those of Braddock and Bradow (1975) who also found very similar soot particles in aircraft turbine engine exhaust.

Ross et al. (1982b) reported on a physical chemical study of the surface properties of diesel exhaust particles. Samples of diesel particles from engine operated on a highly purified light paraffinic fuel and using a low ash synthetic lubricating oil was compared with that from more typical diesel engine fuel-lubricant combinations and with carbon black. The particulate material from the special fuel-lubricant combination contained very small amounts of organic matter, less than 3 percent. The surface area of this substance, when evacuated at 50°C, was approximately the same as carbon black, about 104 m^2/g, several times greater than that of more conventional diesel soot from an Oldsmobile passenger car. The heat of sorption of benzene on this soot was about 15 kcal/mole or about 6 kcal/mole greater than that for conventional diesel soot. Organic material from the special diesel soot preparation contained mainly pyrene and benzo(ghi)perylene, virtually no other hydrocarbon compounds. The absorption properties of the special preparation diesel soot and of carbon black could be explained by conventional Langmuir adsorption on activated carbon sites (Ross et al., 1982a).

On the other hand, conventional diesel soot from an Oldsmobile passenger car contained a multilayer film of heavy hydrocarbon material. Adsorption of gaseous materials on this material was consistent with solution of the gaseous hydrocarbon in the organic film, not with adsorption on the active carbon sites. Thus, the isosteric heats of adsorption of a number of pure hydrocarbons were identical with the heats of vaporization of the hydrocarbons and the adsorption equilibria were completed predicted by Henry's law absorption in the organic film.

Apparently, this soot-carbon black component of particulate material from uncontrolled diesel engines is very similar to other carbonaceous materials from rich flame combustion. Therefore, measures intended to control diesel particles have concentrated particularly on this particle component. Such measures include trapping the soot in the exhaust pipe and burning off the trapped carbon or modifying the combustion process so as to reduce the formation of this material.

Organic Particulate Material

As shown earlier, diesel particles normally carry an oily film of organic matter; a varying amount of this film might be made up of engine lubricating oil depending on the success of oil control in the engine design or the design of air scavenging measures in 2-stroke engine combustion. Some of the organic material is not lubricant derived, however.

Black and High (1979) presented consistent analysis of gaseous and particle-bound hydrocarbons emitted from a diesel passenger car. These authors showed that hydrocarbons ranging in molecular weight from about C14 to C25 were present in both the gas and particle phases. In fact, there were no clear cut lines of demarcation but, rather, an increasing fraction of the heavier molecular weight hydrocarbons in particles as opposed to the gas phase. However, in the particle phase there was a clear separation between fuel derived and lubricant derived hydrocarbons and evidence for materials from both sources in the emitted particles from an Oldsmobile passenger car where about 2/3 of the organics were lubricant derived. However, from two other passenger cars, a Nissan and a VW Rabbit, there was no clear evidence of lubricant contribution. Hyde et al. (1982), demonstrated similarly high contributions of organic material (likely lubricant) to particulate emissions from eight in-use GM diesel passenger cars and low organic particulate emissions from thirteen Mercedes and VW diesel passenger cars which were retested several times over mileage accumulation from about 60,000 to 100,000 miles.

The earliest studies (Hare, Springer and Bradow, 1976; Hare and Bradow, 1979) had shown that the bulk of the organic material emitted is hydrocarbon. For the 4-stroke cycle particulate material, not dominated by motor oil, solvent separation by compound class had recovered about 10 percent of the organics as moderately to strongly acidic compounds and about 1 percent as organic bases. The neutral compounds were separated by column chromatography to produce paraffinic and hydrocarbon cuts accounting for about 60 percent of the neutral fraction and about equal parts of a transition fraction, later found to contain mainly aromatic ketones, e.g. 9-fluorenone and nitroalkanes, and a like amount of slightly more polar substances, mainly alkoxyaromatics and alkoxyphenols.

The 2-stroke cycle materials contained mainly hydrocarbons with, relatively, smaller quantities of the same substances found in the 4-stroke samples. Rodriguez (1980) later reported GC/MS analysis of these samples demonstrating the prevalence of slightly oxidized hydrocarbon moieties (viz. alkoxides and ketones) as well as nitroaromatic hydrocarbons in diesel particles.

In the IARC and EPA documents, there are long lists of polycyclic aromatic hydrocarbons, their alkyl derivatives, and nitrated and oxygenated compounds which have been found in diesel engine exhausts. Generally, the molecular weight range of polynuclear aromatic hydrocarbons reported in diesel particles ranges from about 152 (acenapthene) to 302 (dibenzopyrene) or in equivalent ring numbers, from two to seven aromatic rings (IARC, 1987). Similarly, numerous nitroarenes have been reported in diesel exhaust particles and in ambient air (Liberti et al. 1984; Paputa-Peck et al., 1983; Areys et al., 1987). However, some of these compounds are also emitted from other sources, gasoline engines, for instance (Bradow et al., 1979), and the relative importance of these substances in defining either particle mass or public risk is not defined by these observations.

Benner, et al. (1989), studied the polynuclear aromatic hydrocarbons present both the gas and the condensed phase in the Baltimore Harbor Tunnel. Factor analysis associated a variety of methyl and dimethyl anthracenes and phenanthrenes, pyrene and benzo(ghi)fluoranthene with diesel emissions, while fluoranthene, benzo(a)pyrene and benzo(e)pyrene were associated primarily with gasoline engine emissions. These results are consistent with earlier studies of gasoline engine PAH emissions.

The Benner, et al. (1989), studies also examined the relative amounts of 3-ring PAH materials present in the two phases. It was found that about 2/3 of the dimethylphenanthrenes and anthracenes were in the gas phase. Pyrene and fluoranthene were about equally divided between the gas and particle phase. This observation was consistent with the observations of Black and High (1979) and also with the quantitative assessments of Yamasaki et al. (1987) who described this partition experimentally along the Langmuir absorption isotherm.

In recent years, this distribution problem has been rediscovered several times. For example, Van Vaeck et al. (1984), have described the volatilization of semivolatile compounds from filters as "blow off" or volatilization artifacts. In fact, this phenomena has previously been described quite adequately by Ross et al. (1982a) in the context of adsorption-desorption measurements. If samples of particles obtained in confined circumstances, e.g. roadway tunnels or emissions experiments, are exposed in ambient air with relatively low concentrations of hydrocarbon material, it is expected that those components with significant vapor pressure will evaporate. This will cause variation some in particle mass estimation; in the case of the observations of Kittelson et al. (1988), this difference was about 15 percent.

For example, McDonald et al. (1982), have shown that diesel emissions samples containing relatively large amounts of fuel derived organics show considerable sensitivity to dilution ratio and sampling temperature. Similarly, Kittelson et al. (1988) have found that about 1/3 of the organic fraction of diesel particles can evaporate in the ambient air over roadways. Black and High (1979), however, have shown that there can be very little sensitivity of the organic fraction to evaporation when lubricating oil is the major constituent of this fraction. All these observations are entirely consistent with one another. Certainly C16 through C20 fuel-derived aromatics can evaporate from filters while C30 and larger lubricating oil components will have insufficient vapor pressure to do so. The constitution of ambient air particles will be defined by the concentrations of semivolatile components in the air, the mass and distribution of particle bound components and the ambient air temperature as predicted by Ross et al. (1982a) and Yamasaki et al. (1984).

Therefore, the organic emissions of diesel vehicles are distributed been the gas and particle phases generally according to fairly well understood physical-chemical principles.

EMISSION OF MUTAGENIC SUBSTANCES IN VEHICLE EXHAUST

Since polynuclear aromatic hydrocarbons have long been recognized to accompany soot from combustion sources, the presence of known carcinogens was investigated in early diesel studies. Hare, Springer and Bradow (1976) and Hare and Bradow (1979) measured small amounts of benzo(a)pyrene (BaP) in diesel exhaust particles, very much smaller amounts than those found in gasoline engine exhaust. The use of alkaline earth smoke suppressants was found to substantially increase BaP emissions principally because the material was blended into the fuel with a heavy aromatic solvent. Attempts to find other carcinogens, such as dimethylnitrosamine in exhaust, either in the gas or particle phases was unsuccessful.

Huisingh et al. (1978), reported on studies of mutating substances found in diesel particle extracts in cellular bioassays with both microbes and with mammalian cell bioassays. For quite a time, the Ames Salmonella reverse mutation bioassay became the major method for detecting and measuring the quantity and activity of these minor trace components in particle emissions, simply because there was no convenient chemical analytical method for this determination. As a result, there are quite a number of papers which have described relative emission rates from vehicle categories in units of Salmonella revertants per mile or per bhp/h.

Bradow et al. (1979), reported relative emission rates of mutagenic materials from older passenger cars (model year 1963 and 1971) as well as emissions controlled passenger cars operated on either leaded or unleaded gasoline in comparison with heavy-duty and passenger car diesel engines. All of these vehicles emitted mutagenic materials. Generally, the diesel vehicles had emission rates about 10 fold greater than SI gasoline vehicles, but the specific emission rates for some times, import noncatalyst vehicles in the 1976-1978 model years and some 3-way catalyst vehicles for instance, had significant mutagenic activity.

The major research activity which searched for particular sources of this activity was lead by Schuetzle and his group at Ford Motor Company, Lewtas (Huisingh) and her associates at EPA in Research Triangle Park, and by Rodriguez at South West Research Institutes. Major work on mutagens in the ambient air was that of Pitts and associates at the Statewide Center for Atmospheric Research in Riverside, California. The results of all these studies have been recently summarized in an excellent chapter of the EPA Diesel Toxicity documents by Barbara Zielinska at Desert Research Institute (EPA, 1991a) and by Dennis Schuetzle in the IARC monograph on carcinogen emissions (IARC, 1987). The major portion of mutagenic materials in diesel extracts is accounted for by nitrated pyrenes and fluoranthenes.

There has been much discussion about the artifactual nature of these emissions. Bradow (1980) and Bradow et al. (1982) have summarized the findings to that time, concluding that some artifactual nitration of exhaust particles can occur on filters; however, the evidence strongly suggested that most of the nitropyrene measured was actually emitted to the air. Pierson et al. (1985), in studies on the Pennsylvania turnpike, agreed with this conclusion.

Herr et al. (1982), conducted an interesting series of experiments aimed at discovering the origins of the mutating materials, whose nature was only poorly understood at the time. These authors operated a diesel engine on a very pure hydrocarbon fuel, a blend of tetradecane and isooctane, using a synthetic engine oil which pyrolyzed readily to form only gaseous products. The carbon obtained from these experiments was very dry, containing no trace elements and only 2 percent extractable organics. The organic substances were mainly pyrene and benzo(ghi)perylene, common components of carbon black (Yergey et al. 1982). Salmonella bioassay of this material revealed the presence of the mutagens sensed in other diesel extracts. The engine was then sealed from air and operated on a synthetic oxidant containing helium, argon, carbon dioxide and oxygen which matched both the heat capacity and thermal conductivity of air. An almost identical soot was obtained which this time contained no mutating agents as measured by a variety of microbial bioassays and in mammalian cell lines as well. When samples of this soot were exposed to NO_2, the mutating materials were generated again. These experiments suggest that the entire source of mutating agent in diesel exhaust is nitration of 4-ring and 5-ring polynuclear aromatic hydrocarbons in the soot (Risby and Lestz, 1983).

Subsequent risk analysis performed either by EPA or by contractors to the U.S. Department of Energy suggested that lifetime risk of cancer from this source at exposure levels of 5-10 $\mu m/m^3$ (Bradow, 1980) is in the order of 10 per million at the extreme. Control measures placed in force in the early 1980s for diesel passenger car particles and projected for heavy duty diesel engines were calculated to drastically reduce exposures. Therefore, EPA concluded that the risk from this material, if any, is minor and controllable (EPA, 1991a).

EMISSION OF ASBESTOS FROM BRAKE AND CLUTCH WEAR

General

Asbestos fibers, used as friction materials in brakes and clutches, are abraded in normal use and emitted into the air. In the mid 1970's, there were major differences of opinion expressed between Dr. Irving Selikoff, on the one hand, and members of the automotive industry, particularly brake suppliers, on the other. There had been significant evidence for a long time that in this wear process, asbestos fibers were metamorphosed into nonserpentine minerals, particularly olivine and montmorillionite. The principal difference of opinion was the degree metamorphosis and subsequent emission rate of asbestos fibers into the air.

Enterline (1973) conducted a risk assessment based on analysis of air samples for asbestos fibers in major U.S. cities. He calculated a lifetime risk of about 100 per million for malignant mesothelioma and 2 per million for other lung cancers associated with nonoccupational asbestos exposures. About half of these would be associated with brake wear. However, German government estimates at the time suggested that no asbestos at all resulted from asbestos brake wear.

Jacko, DuCharme, and Somers (1973) conducted the first credible roadway study of asbestos emissions from brake wear. These authors equipped a vehicle with air tight covers over the disk and drum brakes and operated the vehicle within traffic to sample brake debris. These authors found asbestos in brake debris ranging from a high of 1.65 percent of the mass to a low of 0.03 percent in 90 tests. The mean value was 0.40 percent. Calculating human exposure from the resulting emissions estimates suggested that only an insignificant portion of the urban exposure could have resulted from brake and clutch wear. However, these results were questioned because of the higher-than-normal temperatures experienced in the brake pads.

Later studies by Cha, Carter, and Bradow (1983) and by Williams and Muhlbaier (1982) examined the significance of cooling in greater detail. Cha, Carter, and Bradow (1983) equipped a test car with instrumented brake pads, brake line pressure transducer and speed measuring equipment and operated the vehicle within traffic for several hundred hours. The braking actions experienced in traffic were analyzed for duration, speed range, acceleration rate and brake line pressure, brake pad temperature and cooling rate between successive brake applications. This data was synthesized into a group of representative braking cycles which were then studied on a brake dynamometer. Asbestos from a front wheel disk pad operated over these braking cycles was collected using an air dilution tunnel sampling facility and these samples were analyzed by transmission electron microscopy for asbestos content. Williams and Muhlbaier used arbitrary braking cycles for their studies; however, they carefully designed the sampling and brake cooling system to replicate field operation and then carefully qualified the system for asbestos recovery and analysis.

These two studies provided results which were almost identical in every way and which verified the previous Jacko, DuCharme, and Somers (1973) reports. These two studies find about 0.03 percent of the airborne particulate material to contain asbestos fibers, near the low end of the previous work. Both papers also used exposure models to estimate population exposure for these fibers. Cha, Carter, and Bradow (1983) used results from the RAM model for St. Louis, MO (Bradow, 1980) to estimate exposure while Williams and Muhlbaier (1982) used the Cass lead surrogate approach to estimate exposure in Los Angeles. The two studies each found that asbestos wear could account for less than 1 percent of the fibers found in the air in the respective study area. Therefore, these two independent studies produced completely confirmatory results.

Since these studies were completed, front wheel disk pads for passenger cars have been formulated with metal fiber or carbon fiber materials, rather than with serpentine minerals. Asbestos aftermarket replacement pads are still available and are used for passenger cars. Furthermore, truck and bus brakes still use asbestos formulations almost entirely. Overall, asbestos use is down and exposure must be reduced proportionately; however, asbestos brake wear never seemed to have posed any serious risk to the public health and safety.

Fugitive Emission of Asbestos from Serpentine Rock

Recently, it has been observed that unpaved roadways, particularly in California, have sometimes been covered with crushed serpentine rock containing significant levels of asbestos. For example, some of the most common sorts of rock in the Sierra Nevada and many other regions of California contain chrysotile asbestos in amounts ranging from 7 to 19 weight percent (California ARB, 1990).

The entrainment of asbestos fibers from traffic over these roadways creates quite high levels of asbestos exposure downwind. For example, Stenner et al. (1990) have estimated that the downwind asbestos concentrations can reach several micrograms/cubic meter. By comparison, urban central city concentrations are typically in the range 2 to 20 ng/m^3 (California ARB, 1990; Enterline, 1983). Typical cancer risks from such exposure are calculated to be about 65,000 per million compared to 38 per million for urban exposures (California ARB, 1990). Clearly, this represents a substantial incremental risks for the particular persons so exposed.

CHAPTER 7: CONTROL STRATEGIES FOR HIGHWAY PARTICULATE IMPACTS

CONTROL OF DIESEL PARTICLE EMISSIONS

From the above discussion, it should be clear that spark-ignition engines are no longer considered to be a threat with regard to particle emissions. In recent years, all regulatory action and control system development has been aimed at diesel particle emissions control. The EPA Diesel Toxicity document (EPA, 1991a) and a recent paper by Walsh and Bradow (1991) have summarized the current status of this development.

Manufacturers of heavy duty engines have examined the effectiveness of a variety of diesel soot trap-oxidizers, developing this technology to commercial status. FIAT-IVECO have implemented trap oxidizers in a fleet of buses in northern Italy with some success and a number of other manufacturers have been close to commercialization with similar developments. However, engine controls have been developed even more rapidly.

Currently, all U.S. heavy-duty manufacturers are reporting success in achieving the 0.25 g/bhp/h standard with modifications to combustion chamber shapes, high pressure injection systems and electronic injection timing. These combinations also make possible simultaneous achievement of NO_x emissions standards, previously an important tradeoff issue. The control techniques are only successful, however, with catalytic aftertreatment of exhausts to oxidize heavy organic residues. Even with the best engine oil control designs, some emission of this material is likely (Wachter, 1990). Therefore, most manufacturers are planning only oxidation catalytic converter aftertreatment to reduce organic emissions.

This aftertreatment causes formation of sulfuric acid aerosol, as was reported by Pierson et al. (1974) for early passenger car catalysts. Recently, this problem has been eliminated by successful regulation negotiation between EPA, engine manufacturers and fuel suppliers to produce a fuel sulfur limitation consistent with achieving the emission standard.

An important consideration in this chain of events is a series of observations by Johnson and Bagley (1990). These authors found that trap oxidizer technology successfully eliminated carbon black particles in exhaust, but allowed mutagens to pass through essentially unaltered in the gas phase. It is possible that such a control measure could actually enhance the genotoxicity of these compounds. Inhalation of fine particle-bound organics results in the deposition of only 10 to 15 percent of the material in lung tissue. The balance is exhaled. If the same material is presented in the gas phase, its deposition is 80 to 90 percent of the mass. Thus, implementation of carbon black traps could increase the dose of mutating agents by a factor of 6 to 8. Oxidation catalysts, on the other hand, are thought to be especially selective in eliminating polynuclear aromatic hydrocarbons. It is conceivable that the adoption of this alternate technology will substantially reduce the probable small risk of diesel particle mutagen emissions.

A disturbing feature of the state of knowledge concerning particle emissions is the paucity of recent emissions data, particularly of data on controlled engine types. It is highly desirable that some monitoring studies be conducted to ensure that many of the presently tacit assumptions in these analysis prove actually to be the case in practice.

NEW AND EXISTING MOTOR VEHICLE PM EMISSION STANDARDS - THE EFFECT OF THE CAAA OF 1990

The starting point for a discussion of particulate emissions from motor vehicles must begin with the distinctions between vehicles powered by gasoline and those powered by diesel fuel. Because of the nature and temperature of the combustion processes in the two engine types, the particulate emissions from diesel engines are considerably greater and are more readily apparent. Particulate emissions from gasoline-powered vehicles are sufficiently low that there is little data published concerning these emissions.

Another significant difference between the PM emissions from the two engine types is the situation where control strategies or devices employed to reduce one pollutant result in increased emissions of a second pollutant, commonly called an "emissions trade-off". Where the emissions trade-off exists in gasoline-powered vehicles between NO_x and CO emissions, the emissions trade-off in diesel-powered vehicles is between NO_x and PM emissions. Thus, it is difficult to reduce both pollutants through conventional means. Exhaust gas recirculation (EGR), the primary NO_x emission control technique used at this time, is known to increase diesel particulate emissions. The Federal and California exhaust emission standards acknowledge this emissions trade-off in diesel-powered vehicles by permitting higher NO_x emissions standards for diesels than is permitted for gasoline-powered vehicles in the same class. (This differential is legislatively removed at the Federal level for new vehicles after model year 2003.)

Overall, PM emissions from motor vehicles accounted for 18 percent of the nationwide PM emissions in 1989, with diesel-powered heavy-, medium-, and light-weight trucks accounting for the majority of the emissions. PM emissions from diesel-powered motor vehicles result from soot formed during combustion (40 - 80 percent), heavy hydrocarbons condensed or adsorbed on the soot (10 - 50 percent), and from the formation of particulate sulfate compounds (5 - 10 percent). Due to the increasing stringency of the Federal and California regulations for PM emissions and the results of other non-PM emissions regulations, the PM emissions from individual diesel-powered motor vehicles was estimated to have decreased approximately 35 percent between 1970 and 1990 (Radian, 1988). This compares with decreases of 90 percent, 90 percent, and 67 percent for exhaust hydrocarbons, carbon monoxide, and NO_x, respectively, over the same period of time, with the majority of this improvement realized during the 1970's (MVMA, 1991).

Motor vehicle PM emission standards have been modified, and steadily reduced at the Federal level through provisions of the CAA. The emission standards are divided into two major categories, light-duty vehicles, which includes passenger cars and trucks with a gross vehicle weight rating less than 6,000 pounds; and light-duty trucks, which includes trucks with a gross vehicle rating greater than 6,000 pounds. Each truck subcategory is further divided by the loaded vehicle weight or the test weight of the vehicle. The units for the standards are g/m. The standards must be maintained over the useful life period of the standard, which includes a period of performance and a mileage. For example, a 5 year/50,000 mile standard must be maintained for either 5 years or 50,000 miles, whichever first occurs.

The CAAA of 1990 established PM emission standards for motor vehicles in two sections within Title II, Sections 203, Emission Standards for Conventional Motor Vehicles, and 229, Clean Fuel Vehicles. The CAAA of 1990 also established PM emission standards for buses in Title II: Section 207, Emission Control Diagnostics and Buses, and Section 227, Urban Buses. These sections amend Section 202 of the 1977 CAA.

Prior to passage of the CAAA of 1990, the PM emission standard for passenger cars, model years 1991 through 1993, was .20 g/m with a reduction to .08 g/m (for a period of 5 years or 50,000 miles) and .10 g/m (for a period of 10 years or 100,000 miles) for model year 1994. For model years 1991 through 1995, the PM emission standard for all light-duty trucks is .13 g/m.

The CAAA of 1990 maintained the PM emission standard for passenger cars at the .08 g/m/.10 g/m levels and extended these standards to model years 1994 through 2006, with a gradual increase in the percentage of the fleet that would have to achieve the standards. The CAAA of 1990 also extended these PM emission standards to light-duty trucks under 5,750 pounds (test weight), also with a phase-in period. The PM emission standard for light-duty trucks over 5,750 pounds (test weight) was established as .12 g/m, also with a phase-on period. The light-duty truck standards for trucks with a gross vehicle weight rating less than 6,000 pounds apply to model years 1995 and after; the light-duty truck standards for trucks with a gross vehicle weight rating in excess of 6,000 pounds apply to model years 1996 and after.

The provisions of Section 229, the Clean Fuel Vehicle program - Fleet Vehicle provisions, generally extends the .08/.10/.12 g/m PM emission standards to the vehicles within this program for slightly longer useful life periods. There is also a phase-in period for these standards which coincides with other provisions for the program.

The PM emission standard for buses was established as .25 g/bhp/h for model years 1991 and 1992 and .10 g/bhp/h for model years 1993 and after in Section 207. Section 227 stipulates that the PM emission standard for model years 1994 and after must be no less than 50 percent of the PM emission standard for model year 1990 buses.

Particulate Traps to Reduce Motor Vehicle Emissions

One method under investigation to meet the increasingly more stringent PM emission standards for diesel-powered motor vehicles is the particulate trap, either alone or coupled with a catalyst system. Although there are development projects underway, none have been commercialized at this time for either on-road or off-road applications. Particulate traps are placed in-line on the exhaust system of a vehicle. The trap consists of a fabric or ceramic element housed within a metal canister, occasionally fitted with a screen filter on the inlet end. After a period of use the particulate trap is removed and cleaned of the collected material with the trap re-installed on the vehicle. Studies have been conducted using various fuel additives and catalyst systems to increase the efficiency of the particulate trap in use. These fuel additives have included iron, copper, or manganese. (Although the 1977 CAA Amendments permitted the maximum manganese concentration in gasoline to be .0625 grams per gallon (Section 211), the CAAA of 1990 repealed its possible use, unless a waiver was granted by the EPA Administrator (Section 219).) There have also been studies conducted to determine the possible health effects from the

PM emissions from engines utilizing these fuel additives, due to concerns about the hazardous air pollutants emitted which contain the fuel additive by-products (HEI, 1988).

Increased PM Emissions Due to Tampering

There are a number of modifications to motor vehicles which lead to increased PM emissions. When intentionally performed, either by physically altering the vehicle or misfueling, these modifications are referred to as tampering. Tampering can be grouped into two principal categories: 1) modifications to emission control devices and 2) modifications related to combustion processes. The former category includes the removal, disabling, or bypassing of control devices such as catalytic converters, electronic controls, traps, and the EGR system. The disabling of these control devices can also occur through misfueling of a vehicle. The latter category includes physically modifying some aspect of the combustion system, such as modifying the fuel/air ratio, advancing or retarding the timing, or incorrectly repair or rebuilding an engine component.

The results of tampering on the emissions from a vehicle can be substantial; the removal of a trap-oxidizer or the modification of the injection timing, for example, will result in increased emissions in excess of 100 percent for some individual vehicles (Radian, 1988). Overall, the tampering of vehicles was estimated to increase the California statewide emissions of PM emissions from diesel-powered vehicles by 70 percent in 1990 and 100 percent in 2000 (Radian, 1988).

Tire Wear and PM Emissions from Motor Vehicles

It was thought that the PM emissions associated with tire wear represented a significant source of these emissions. This possible link lead to a closer examination and preliminary regulation development by the air pollution control agencies in areas where ambient PM concentrations were approaching or exceeding the NAAQS. One air pollution control district, the South Coast Air Quality Management District (SCAQMD [Los Angeles]), proposed the use of radial tires in lieu of bias-ply tires as an air quality management plan control strategy to reduce PM emissions from this source. (Because most new cars are equipped with radial tires, this control measure was directed at the tire replacement market, and sought to replace the use of bias-ply tires with radials on older model cars.) Results of studies have shown that while tire life has increased as much as 200 percent by the use of radial tires and the resulting PM emissions are reduced, the net ambient air quality improvement is negligible for several reasons: 1) PM emissions are, initially, very small from bias ply tires, 2) the size distribution of the PM emissions lead to the particles being rapidly deposited along the roadway margins, within 10 to 20 meters, with little or no material transported beyond these margin areas, and 3) the size distribution of the emitted PM allows rapid deterioration of the individual particles through oxidation with ambient oxygen or ozone. (Personal communication with Randy Clark, Michelin Tire Services.)

CONTROL STRATEGIES FOR SOURCES IN NONATTAINMENT AREAS

Review of State Implementation Plans

Areas with air quality monitoring data in exceedance of the applicable NAAQS for PM-10 are designated as nonattainment areas. In order to be re-designated as attainment, the State in which the nonattainment area is located must submit a State Implementation Plan (SIP) that includes both an emission inventory for all sources thought to be contributing to the pollutant concentrations and a control strategy plan designed to decrease the levels of the pollutant below the applicable NAAQS. Frequently, an air quality modeling demonstration will also be included that takes into account the decrease in emissions resulting from the implementation of the control strategies and presents information for selected receptor locations indicating that the levels are below the NAAQS.

The CAAA of 1990 provided for a number of areas that were in violation of the PM-10 NAAQS to submit SIPs within one year of the date of enactment of the legislation. As a consequence, many of the PM-10 nonattainment area SIPs were due on November 15, 1991. However, few of these areas had actually submitted SIPs by the deadline. As a consequence, Pechan was only able to review four SIP emission inventory submittals. The four areas reviewed included Silver Valley, Idaho; Medford-Ashland, Oregon; Klamath Falls, Oregon; and Presque Isle, Maine. The information received for Silver Valley, Idaho included only the emission inventory submittal and did not contain any control strategy information. However, by examining the emission inventory, the major source contributors can be determined and a highway source assessment can be made.

Silver Valley, Idaho

Total point source emissions of PM-10 within the nonattainment area were calculated to be 62.94 tons/year. By contrast, emissions from area sources (all values in tons/year) were 181 for fugitive road dust (134 from paved road resuspension, including road sanding and salting and 34 from unpaved road emissions), 85.2 for woodstoves/fireplaces, 38.5 from building construction, 9.8 from tailpipe emissions, 2 from aircraft, 1.3 from open burning, 0.9 from coal combustion, 1.5 from tire and brake wear, 1.4 from railroads and locomotives, 0.4 from road construction, 0.4 from heating oil combustion, and 0.7 from natural gas combustion. Thus, based on the emission levels calculated for the SIP inventory, approximately 27 percent is derived from paved road resuspension, 7 percent from unpaved roads, 2 percent from tailpipe emissions, 0.3 percent from tire and brake wear and 0.08 percent from road construction activities. Clearly, any control strategy implemented in this nonattainment area for highway sources would be most effective if its focus was on control of emissions from resuspension of paved roadway material and unpaved roads.

Presque Isle, Maine

Total point source emissions of PM-10 in the Presque Isle nonattainment area totaled 171 tons/year. Emissions from highway source included 2.9 tons/year from tailpipe emissions for gasoline vehicles, and 4.4 for diesel vehicles. Road dust (paved) contributed 1095 tons/year from truck traffic and 1912 from automobiles. This road dust contribution was attributed primarily to road sanding and salting during winter months. Area source combustion processes contributed a total of 251 tons/year. Thus, 88 percent of all emissions in this nonattainment area were attributable to paved road resuspension. 0.21 percent were estimated to be from direct tailpipe emissions. Road construction and tire and brake wear emissions were not considered in this inventory.

Control strategies suggested to bring about attainment of the PM-10 NAAQS in Presque Isle included more careful control of the silt content of anti-skid materials, better quality assurance of the hardness of anti-skid materials, removal of anti-skid materials from the road surface so that the loading on the road does not exceed 10 g/m^2, utilize a special clean up effort in the curb lane to remove material build-up, more frequent water flushings to remove silt material and speed control by utilizing a daytime traffic control sequence at night where blinking yellow lights were utilized in the past.

Medford-Ashland, Oregon

PM-10 emissions in the Medford-Ashland nonattainment area can be broken down into the following source categories (all values in tons per year): wood products industry sources (1275), residential wood heating (1777), fugitive dust (1008), backyard and agricultural burning (83), transportation sources (290) and other sources (241). The two major fugitive dust sources are paved (430) and unpaved (489) road dust emissions. Transportation emissions include tailpipe and tire wear emissions (225), off-highway vehicles (58) and railroad and aircraft (7). Thus highway particulate sources account for 24 percent of all PM-10 emissions. Again, paved and unpaved road dust are the largest highway contributors with 9 percent and 10 percent, respectively.

Control strategies include continuing to pave unpaved roads, curb and gutter shoulders on paved roads, and control of mud and dirt trackout from industrial, construction and agricultural operations.

Klamath Falls, Oregon

Sources of PM-10 emissions in the Klamath Falls nonattainment area included industrial sources (189 ton/year), residential woodheating (1202), backyard and agricultural burning (173), fugitive dust (230), and transportation sources (230). Paved road resuspension is attributed with 112 tons/year, unpaved roads with 53, tailpipes and tire wear with 97 and off-highway vehicles with 12 ton/year. Thus, highway sources contribute 13 percent to the total PM-10 emissions. As with the other areas reviewed, the main contributors are paved road resuspension and unpaved roads.

Control strategies in the Klamath Falls nonattainment area for highway sources included a 60 percent reduction in winter road sanding emissions via use of liquid road deicing techniques in lieu of rock aggregate, application of less road sanding material and rapid cleanup of used road sanding aggregate and mandatory cleanup of trackout from unpaved areas onto highway right-of-ways.

Clearly the examination of the SIP inventories and control strategies for the nonattainment areas examined indicate that the major source of highway particulate emissions that the States must deal with in order to maintain the NAAQS are paved road resuspension emissions and emissions from unpaved roads.

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