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Delucchi, Mark A. McCubbin, Donald R.

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THE CONTRIBUTION OF MOTOR VEHICLES AND OTHER SOURCES TO AMBIENT AIR POLLUTION

Report #16 in the series: The Annualized Social Cost of Motor-Vehicle Use in the United States, based on 1990-1991 Data

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Mark A. Delucchi¹

Donald R. McCubbin²

¹Institute of Transportation Studies University of California Davis, California 95616

(916) 967-1915 phone and fax madelucchi@ucdavis.edu

²Department of Economics University of California, San Diego La Jolla, California 92093

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TABLE OF CONTENTS

ACKNOWLEDGMENTS	i
REPORTS IN THE SERIES	ii
LIST OF ACRONYMS	iii
TABLE OF CONTENTS	v
16. THE CONTRIBUTION OF MOTOR VEHICLES AND OTHER	
SOURCES TO AMBIENT AIR POLLUTION	1
16.1 MODELING AIR POLLUTION AND THE CONTRIBUTION OF MOTOR	
VEHICLES	
16.1.1 Background	1
16.1.2 Modeling pollution formation and estimating the	
contribution of motor vehicle emissions to ambient	
pollution	2
16.2 ESTIMATES OF EMISSIONS: THE EPA'S OFFICIAL EMISSIONS	
INVENTORY (OEIP',I,C), AND OUR CORRECTIONS TO THE EPA	
ESTIMATES (ECP',I)	
16.2.1 Background	11
16.2.2 Estimates of VOCs, NOx, and CO emissions from	
mobile sources (MOBILE5a model)	12
16.2.3 Estimates of PM and SOx exhaust emissions from	4.77
mobile sources (PART5 model)	17
16.2.4 Estimates of PM dust from paved roads (AP-42	20
Volume 1, and PART5 model)	23
16.2.5 Estimates of PM dust from unpaved roads (AP-42,	2.4
Volume 1)	34
16.2.6 Estimates of PM emissions from construction,	2.4
including road construction (AP-42, Volume 1)	
16.2.7 Summary of correction factors	33
QUALITY MONITOR	26
16.3.1 Conceptual approach to air-quality modeling	
16.3.2 The Gaussian model	
16.3.3 The results of the model	
16.3.4 Comparison with other estimates	
16.3.5 Long-range transport	
16.4 Atmospheric chemistry: the contribution Of Motor	02
VEHICLES TO OZONE	62
16.4.1 Background	
16.4.2 Alternative simple methods for estimating the	02
contribution of precursors to ozone formation	63

16.5 ATMOSPHERIC CHEMISTRY: THE FORMATION OF SECONDARY	
SULFATE AND NITRATE PARTICULATES FROM EMISSIONS OF NOX,	
SO2, AND NH3	70
16.5.1 Background	70
16.5.2 Formation of ammonium sulfate from SO2 and NH3	
emissions	71
16.5.3 Formation of ammonium nitrate from NOx and NH3	
emissions	76
16.5.4 Other contributors to secondary particulate formation	79
16.5.5 Secondary organic aerosols (SOA)	
16.5.6 Size distribution of ammonium sulfate, ammonium	
nitrate, and organic aerosols	81
16.5.7 Formal model of ambient particulate levels after a	
change in emissions	82
16.6 COMPARISON OF OUR MODELING RESULTS WITH THE SOURCE-	
APPORTIONMENTS FROM CHEMICAL MASS-BALANCE STUDIES	85
16.7 References	
ABBREVIATIONS USED IN THE TABLES IN THIS REPORT	98
TABLES	
TABLE 16-1. CORRECTIONS TO THE EMISSIONS INVENTORY: THE RATIO OF	
OUR ESTIMATE OF EMISSIONS TO THE EPA'S (1995D) OFFICIAL	
ESTIMATES	99
TABLE 16-2. PM AND OTHER EXHAUST EMISSIONS FROM HIGH-MILEAGE, IN-	
USE LIGHT-DUTY GASOLINE VEHICLES COMPARED TO PART5	
MODEL EMISSIONS	100
TABLE 16-3. PM EXHAUST EMISSIONS FROM IN-USE HEAVY-DUTY VEHICLES	
TESTED OVER A TRANSIENT CYCLE ON A CHASSIS DYNAMOMETER	101
TABLE 16-4. COMPARISON OF MOTOR VEHICLE PM EXHAUST EMISSIONS	
BACK-CALCULATED FROM FIELD STUDIES AND EMISSIONS	
CALCULATED BY THE PART5 MODEL (GRAMS/MILE)	102
TABLE 16-5. CALCULATION OF TRAVEL FRACTIONS AND AVERAGE VEHICLE	102
WEIGHTS, FOR USE IN THE PART5 MODEL APPLIED IN TABLE 16-4	
AND TABLE 16-6	106
TABLE 16-6. CALCULATION OF TOTAL PM EMISSIONS FROM TRAFFIC, USING	100
PART5/AP-42	100
TABLE 16-7. COMPARISON OF EMFAC7F AND MOBILE5A ESTIMATES OF	100
PM EMISSIONS	100
	109
TABLE 16-8. MOTOR-VEHICLE AND FUGITIVE-DUST EMISSIONS OF PM IN	
URBAN AREAS OF THE U.S. IN 1990, ACCORDING TO THE OFFICIAL	110
EPA EMISSION INVENTORY (MILLION TONS)	110

TABLE 16-9. SOURCE CONTRIBUTIONS TO AMBIENT PM10, AS ESTIMATED BY	
CHEMICAL MASS-BALANCE STUDIES	111
Table 16-9 (continued).	112
TABLE 16-9 (CONTINUED).	113
TABLE 16-10. SOURCE CONTRIBUTIONS TO AMBIENT PM2.5, AS ESTIMATED BY	
CHEMICAL MASS-BALANCE STUDIES	116
TABLE 16-11. THE RATIO OF ROAD-DUST PM TO MOTOR-VEHICLE EXHAUST	
PM: CMB SOURCE APPORTIONING VERSUS THE EMISSIONS	
INVENTORY	118
TABLE 16-12. ATMOSPHERIC RESIDENCE TIME AS A FUNCTION OF PARTICLE	
SIZE	119
TABLE 16-13. COMPARISON OF MOTOR VEHICLE PM EMISSIONS BACK-	
CALCULATED FROM FIELD STUDIES AND EMISSIONS CALCULATED	
BY PART5/AP-42 STUDIES OUTSIDE OF THE U.S. MIDWEST	
(GRAMS/MILE)	121
TABLE 16-14. SIZE DISTRIBUTION OF PARTICLES FROM VARIOUS FUGITIVE	
DUST SOURCES (PERCENT OF PARTICLES BELOW INDICATED SIZE)	123
TABLE 16-15. ESTIMATES OF CONTRIBUTION TO AIR QUALITY, RELATIVE TO	
CONTRIBUTION OF LDVS, PER KG OF EMISSIONS, BASED ON	
SIMPLE DISPERSION MODELING: ASSUMED VALUES OF INPUT	
PARAMETERS	124
Table 16-15 (continued)	127
Table 16-15 (continued)	
TABLE 16-16: STATISTICS REGARDING AQCRS AND COUNTIES WITHIN	
AQCRs	131
TABLE 16-17. STATISTICS FOR MAJOR POINT SOURCES	132
TABLE 16-18. OUR ASSUMPTIONS AND CALCULATIONS REGARDING SETTLING	
AND DEPOSITION VELOCITY AND REACTION RATES OF PARTICLES	
AND GASES	134
TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR DNP',I,C, AND	
DNP',I,OC, THE CONTRIBUTION TO AMBIENT POLLUTION PER U	
NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE	
CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY	
MOTOR-VEHICLES	135
A. Urban monitors, emission sources within the county, low-	
COST CASE	135
B. Urban monitors, emission sources within the county,	
HIGH-COST CASE	136
C. Urban monitors, emissions outside the county, small	
AQCRS, LOW-COST CASE	137
D. Urban monitors, emissions outside the county, small	
AQCRS, HIGH-COST CASE	138

E. URBAN MONITORS, EMISSIONS OUTSIDE THE COUNTY, LARGE	
AQCRS, LOW-COST CASE	139
F. URBAN MONITORS, EMISSIONS OUTSIDE THE COUNTY, LARGE	
AQCRS, HIGH-COST CASE	140
G. AGRICULTURAL MONITORS, EMISSION SOURCES WITHIN THE	
COUNTY, LOW-COST CASE	141
H. AGRICULTURAL MONITORS, EMISSION SOURCES WITHIN THE	
COUNTY, HIGH-COST CASE	142
I. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY,	
SMALL AQCRS, LOW-COST CASE	143
J. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY,	
SMALL AQCRS, HIGH-COST CASE	144
K. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY,	
LARGE AQCRS, LOW-COST CASE	145
L. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY,	
LARGE AQCRS, HIGH-COST CASE	146
TABLE 16-20. EPA-ESTIMATED EXPOSURE FACTORS FOR DIFFERENT PM	
EMISSION SOURCES (EPA, 1994B)	147
TABLE 16-21. DIESEL ENGINES IN THE SOUTH COAST AIR BASIN, 1982: FUEL	
USE, EMISSIONS, AND CONTRIBUTION TO TOTAL PARTICULATE	
POLLUTION	149
TABLE 16-22. OZONE SENSITIVITY TO VOC AND NOX EMISSIONS	
TABLE 16-23. EMISSIONS, POCP-WEIGHTED EMISSIONS, AND POCP-	
ADJUSTMENT FACTORS FOR VARIOUS VOC-EMISSION SOURCES	153
TABLE 16-24. ADJUSTED SALES OF DISTILLATE FUEL OIL IN ARIZONA,	
CALIFORNIA, AND NEVADA IN 1993, BY TYPE OF END USE (103	
GALLONS)	154
TABLE 16-25. SOURCE-SPECIFIC FACS BY LAND COVER TYPE	
TABLE 16-26. COMPARISON OF SOURCE-APPORTIONMENTS FROM CHEMICAL	
MASS-BALANCE STUDIES (CMB) WITH MODELING RESULTS	
PERCENTAGES OF PM10 ATTRIBUTABLE TO FOUR SOURCES	156
FIGURES	
FIGURE 16-1. MOTOR-VEHICLE EMISSION SOURCES, OTHER EMISSION	
SOURCES, AND RECEPTOR SITES IN COUNTIES IN AN AIR-QUALITY	
CONTROL REGION	158
FIGURE 16-2. MODELED REPRESENTATION OF MOTOR-VEHICLE EMISSION	
SOURCES, OTHER EMISSION SOURCES, AND RECEPTOR SITES IN	
COUNTIES IN AN AIR-QUALITY CONTROL REGION	159
FIGURE 16-3. DISPERSION OF POLLUTION FROM A POINT SOURCE	

16. THE CONTRIBUTION OF MOTOR VEHICLES AND OTHER SOURCES TO AMBIENT AIR POLLUTION

16.1 MODELING AIR POLLUTION AND THE CONTRIBUTION OF MOTOR VEHICLES

16.1.1 Background

In this Report, we explain how we model the contribution of motor-vehicles and other emissions sources to ambient air pollution.

In Reports 11, 12, and 13 of this social-cost series (see the list at the beginning of this report), we develop dose-response functions that estimate changes in human health, crop production, and visibility as a function of changes in ambient air pollution:

$$\Delta E = f(\Delta P, O) = f(PI, PP, O)$$
 [0]

where:

 ΔE = the change in the effect of interest (human health, crop production, or visibility)

 ΔP = the change in ambient air pollution

O = other variables (such as population or incidence rate)

PI = the initial pollution level

PP = the pollution level after the change in pollution -- in this social-cost analysis, the level after removing all anthropogenic emissions, or 10% or 100% of motor-vehicle related emissions

The initial pollution level, PI, is the actual ambient air quality in each county in the U. S. These data, and the data for any of the other variables O, such as population, are discussed in Reports 11, 12, and 13. *In this report, we discuss how we estimate PP, the pollution level after removing anthropogenic emissions, or 10% or 100% of motor-vehicle related emissions.*

Note that, when we estimate the pollution level after removing motor-vehicle related emissions, we estimate the effects of a specific, "marginal" change in pollution: the difference between actual pollution (PI) and, what pollution would have been had motor-vehicle-related emissions been reduced by 10% or 100% (PP). We did consider as an alternative estimating the effect of all anthropogenic air pollution and then assigning a fraction of this total effect to motor vehicles, but for two reasons rejected this alternative. First, some of our dose-response functions (in Reports 11, 12, and 13) are nonlinear, which means that the change in effects (the responses) depends not only on the difference between PI and PP (the "doses"), but on the absolute magnitudes of PI and PP as well. A decrease in pollution from 15 units to 10 units does not necessarily

result in the same change in effects as does a decrease from 10 units to 5 units or from 5 units to zero units. If all of the dose-response functions were linear, then effects would be a function only of the difference between PI and PP, and one would have to specify only this difference, and not the absolute values of PI and PP. But as this is not the case, we must specify the absolute magnitudes of PP and PI.

Second, because ozone formation is a nonlinear function of two precursor pollutants, NO_X and VOCs, the only way to model the real nonlinear effect on ozone of motor-vehicle ozone-precursor emissions is to model actual ozone levels with and without motor vehicle precursor emissions. It simply is not meaningful to model the elimination of all anthropogenic pollution and then use some ad-hoc rules or "apportioning" factors assign a fraction of this eliminated pollution to motor vehicles.

In short, we perform a "with/without" analysis: we estimate the health, agriculture, or visibility effects of the difference between total air pollution (with motor-vehicle-related emissions) and air pollution with 10% or 100% of motor-vehicle-related emissions eliminated. To estimate the difference in pollution due to motor-vehicle emissions, we use data on ambient air quality, a detailed emissions inventory, emissions correction factors, and a simple air-quality dispersion model.

16.1.2 Modeling pollution formation and estimating the contribution of motor vehicle emissions to ambient pollution

Recall that our task in this report is to estimate PP, the pollution level without motor-vehicle related emissions (equation 0). In each county, we estimate PP on the assumption that the ratio of PP to PI (initial pollution in each county) is equal to the ratio of the *modeled* PP to *modeled* PI:

Assume :
$$\frac{PP}{PI} = \frac{PP*}{PI*}$$
 (1a, 1b)
 $PP = PI \cdot \frac{PP*}{PI*}$

where:

PP = the estimated actual pollution level after the change in pollution (eliminate all anthropogenic emissions, or eliminate 10% or 100% of motor-vehicle-related emissions)

PI = the actual total ambient pollution level (data from air-quality monitors [EPA, 1993]; discussed in Reports 11, 12, and 13)

PP* = the modeled level of pollution after the change in pollution

PI* = the modeled level of total ambient pollution.

Thus, in order to estimate PP, we must develop a model of ambient pollution, and estimate the ratio of PP* to PI* in each county.

In general, ambient air pollution at particular time and place is a function of the amount of pollutants emitted per unit time, the physical dispersion of the emissions from the emissions source to the site where the ambient pollution is being measured, and chemical transformations of pollutants. Dispersion and chemical transformations are a function of topography, meteorology, the mix of pollutants, and other factors. Formally:

$$PI_{P}^{*} = f(E_{P',i}; D_{P',i}(d,h,m,t...); C_{P'\to P}(s,m,t...))$$

$$PP_{P}^{*} = f(E_{P'\land,i}; D_{P',i}(d,h,m,t...); C_{P'\to P}(s,m,t...))$$
(2a, 2b)

where:

 PI_p^* = the modeled initial level of ambient pollution P, at a particular time and place

 PP_p^* = the modeled level of pollution P at a particular time and place, after the change in emissions

P = the ambient pollutant, measured at the ambient air-quality monitors and included in health, crop, or visibility damage functions: carbon monoxide (CO), ozone (O₃), nitrogen oxides (NO_X), total suspended particulate matter (TSP), particulate matter less than 10 microns in aerodynamic diameter (PM₁₀), and particulate matter less than 2.5 microns (PM_{2.5})¹

 $E_{p',i}$ = emissions of P' from source i, over some time period

p' = the emitted pollutant: CO' (--> CO), PM_{2.5-10}' (also called "coarse" PM₁₀) (--> PM₁₀), PM_{2.5}' (--> PM_{2.5}, PM₁₀), NO_x' (--> NO₂, O₃, PM₁₀, PM_{2.5}); volatile organic compounds (VOCs'; --> O₃, PM_{2.5}), SO₂' (--> PM₁₀, PM_{2.5}), ammonia (NH₃' --> PM₁₀, PM_{2.5})

 $D_{p',i}(d,h,m,t...)$ = the dispersion of emissions P' from source i, as a function of distance (d), height (h), meteorology (m; e.g., wind, temperature), topography (t), and other factors

 $C_{p'-->p}(s,m,t...)$ = the chemical transformation of emissions of P' to ambient pollutant P, as a function of the mix of pollution (s), meteorology (m), topography (t), and other factors

 $^{^{1}}$ We do not include sulfur dioxide (SO₂) as an ambient pollutant because we do not attribute any health, visibility, or agricultural effects to SO₂ per se. However, we do account for the contribution of SO_X emissions to ambient particulate levels.

In Report #11, we also estimate the health effects of toxic air pollutants, but the method of estimating the motor-vehicle contribution to toxic air pollution is different from the method, outlined in this report, of estimating the motor-vehicle contribution to other ambient pollution. The analysis of the damage cost of motor-vehicle toxics is presented in Report #11.

 $E_{p'}$, i = emissions of P' from source i over some time period, minus the emissions that are presumed to be eliminated; in other words, the emissions of P' from source i that remain after the hypothetical change in emissions has occurred

Note that we distinguish between ambient air pollutants (P), measured at airquality monitors, and emitted pollutants (P'), which disperse, and in some cases participate in chemical reactions, to become ambient, measured pollutants. Emitted pollutants can be the same chemical compounds as ambient pollutants (e.g., carbon monoxide [CO] is emitted, and also is an ambient pollutant), or can be involved in chemical reactions that produce ambient pollutants (e.g., volatile organic compounds [VOCs] are emitted, and are involved in the atmospheric formation of ozone).

To model the link between emissions and ambient air pollution we make several simplifications:

- **I).** We assume that in each county c, the ambient pollution measured at the airquality monitors is a function of:
 - i) emissions in county c, and
 - ii) emissions from other counties in the same Air Quality Control Region² (AQCR) as county c.

In essence, we model emissions from two source areas, or bands: the county of the monitor, and the band of counties around the county of the monitor. As explained next, we do this as a compromise between the impossible task of modeling emissions from every individual source and the oversimplification of having only one set of emission sources per air basin.

Recall that we estimate ambient air quality, as measured at EPA-ambient airquality monitors, in each county. Ideally, we would model air quality in each county as a function of emissions from every source that contributes in any way to air quality in the county. This would require that we formally locate and characterize every individual emissions source, define air basins and pollution transport regions, and model air quality as a function of all effective emissions sources. Unfortunately, we do not have the data or resources to be able to do such detailed modeling for every county and air basin the U. S.

Rather than model the effect on air quality of every individual emissions source, one can define bands or regions of emissions, each with an effective "center" of emissions, and model the effect on air quality of emissions from these bands. The greater the number of bands or regions (as aggregations of emissions sources), the greater the precision, but the greater the data and analytical requirements. Our balance is to choose two emissions "bands," or areas: the county of the air-quality monitor in question, and the counties outside of this county but within the same AQCR. Within the county, we will estimate the actual effective location of different source categories

²Air quality control regions are defined in the Code of Federal Regulations (Section 40: Part 81).

(highway vehicles, power plants, off-road vehicles, construction, and so on). In the outside counties, we will assume a single effective location for all emission sources. We discuss this in more detail in Section 16.3.

- **II).** We ignore the transport of pollution from one AQCR to another, and assume that pollution within an AQCR is a function only of emissions within the AQCR. This assumption obviates the difficulties of analyzing long-range pollutant transport, and hence greatly simplifies our analysis. Of course, as discussed a bit further in Section 16.3, on dispersion modeling, we recognize that in some areas, such as the Northeastern U.S., long-range transport is important, and ideally should not be ignored.
- **III).** We assume that emissions of precursor pollutants P' disperse as P' from the source to the receptor (the ambient air-quality monitor), and then at the receptor undergo any chemical transformations to produce ambient pollutant P. For example, we assume that VOC and NO_X emissions disperse as such from anywhere in the AQCR to the receptor in the county of interest, and at the receptor then are converted into ozone (O3). We make this assumption because we cannot easily model chemical transformations as a function of the distance from the source.
- **IV).** In equation 1, we estimate the *ratio* PP*/PI*; we do not estimate PI* and PP* individually in units of concentration ($\mu g/m^3$). We do this because there is less uncertainty in modeling dispersion from one source *relative* to another than in modeling dispersion in absolute terms. Our model estimates the dispersion of emissions from non-motor-vehicle sources relative to dispersion of emissions from light-duty motor vehicles. With this relative model of dispersion, we can estimate the ratio PP*/PI*, but not PP* and PI* individually. We discuss this more below and in section 16.3.
- **V).** In the cases where we model the chemical transformation of precursor emissions to ambient pollutants (VOCs, NO_X --> O_3 ; NO_X , SO_X , NH_3 , VOCs --> PM_{10} , $PM_{2.5}$), we ignore meteorology and topography and assume that the ambient pollution is a function only of the amount precursor emissions at the site of the monitor.

A simple model of pollutant formation

With these assumptions, we consider a simple model of pollutant formation:

$$\left(\sum_{i} \left(E_{P1',i,c} \cdot D_{P1',i,c} + E_{P1',i,oc} \cdot D_{P1',i,oc} \right) \right)$$

$$| PI_{P,c} *= C_{P' \to P} \left| \sum_{i} \left(E_{P2',i,c} \cdot D_{P2',i,c} + E_{P2',i,oc} \cdot D_{P2',i,oc} \right) \right|$$

$$| ...$$

(3a, 3b)

where:

$$E_{P',i,oc} = \sum_{o \in R_c} E_{P',i,o}$$

PI_{p,C}* = the modeled level of total ambient pollution P "received" or formed at air-quality monitors in county C, in a year, given the baseline emissions PP_{p,C}* = the modeled level of total ambient pollution P "received" or formed at air-quality monitors in county C, in a year, after the change in emissions subscript P = the ambient pollutant, measured at ambient air-quality monitors subscript C = the county of interest (i.e., the county for which air quality and the cost of air pollution are estimated)

subscript P' = the emitted pollutants

subscript R_C = the AQCR that contains county C

subscript OC = all counties other than county C in AQCR R_C

subscript O = a county other than C in AQCR R_C (all O together make OC)

 $C_{p'-p}$ = the chemical transformation of emissions of precursor pollutants P' (P1', P2',...) to ambient pollutant P (discussed below; this transformation function is assumed to be the same in every county, and to be independent of the source of the emissions)

 $E_{p1',i,c}$, $E_{p2',i,c}$... = yearly baseline emissions of precursor pollutants P1', P2'... from emissions source i in county C

 $E_{p1',i,oc}$, $E_{p2',i,oc}$... = yearly baseline emissions of precursor pollutants P1', P2'... from emissions source i in all counties except C in AQCR R

- $D_{p1',i,c}$, $D_{p2',i,c}$ = the fraction of emissions of precursor pollutants P1', P2'... from source i in county C that reaches the ambient air-quality monitor in county C
- $D_{p1',i,oc}$, $D_{p2',i,oc}$ = the fraction of emissions of precursor pollutants P1', P2'..., from source i in all counties except C in AQCR R, that reaches the ambient air-quality monitor in county C
- $E_{p1'}$, $E_{p2'}$,
- Ep1'^,i,oc, Ep2'^,i,oc ... = yearly emissions of precursor pollutants P1', P2'... from source i in all counties except C in AQCR R, after the change in emissions
- $E_{p',i,o}$ = emissions of pollutant P' from source i in county O in AQCR R_{c} (for simplicity, we leave the notation for P' general, and do not write out separate equations for P1', P2', P1'^, and P2'^)

Now, recall that we will model pollution with 100% of anthropogenic emissions eliminated, and with 10% and 100% of emissions related to motor-vehicle use eliminated. Emissions "related" to motor-vehicle use comprise direct emissions, such as evaporative, tailpipe and road dust emissions, and "indirect" emissions from sources such as the production of motor fuel at refineries, the assembly of motor vehicles, the servicing of motor vehicles, the manufacture of materials used in motor vehicles, road construction, and so on. Because so many sources are related to motor-vehicle use in one way or another, we incorporate formally into our model a motor-vehicle share factor, which is the share of emissions, from each source in the emissions inventory, that is related to motor-vehicle use. From some of the sources in the inventory (such as highway construction, and of course motor-vehicles themselves), all of the emissions are attributable to motor-vehicle use; from other sources (such as agricultural operations), none of the emissions are attributable to motor vehicle use; and from still other sources (such as petroleum refineries), some portion of the emissions are attributable to motorvehicle use. Thus, for the cases in which we eliminate 10% or 100% of motor-vehiclerelated emissions:

$$E_{P^{\wedge},i} = E_{P',i} - E_{P',i} \cdot k \cdot MS_{P',i} = E_{P',i} \cdot (1 - k \cdot MS_{P',i})$$

and

$$\left(\sum_{i} \left(E_{P1',i,c} \cdot D_{P1',i,c} + E_{P1',i,oc} \cdot D_{P1',i,oc}\right) \cdot \left(1 - k \cdot MS_{P1',i}\right)\right) \\
| PP_{P,c} * = C_{P' \to P} \left|\sum_{i} \left(E_{P2',i,c} \cdot D_{P2',i,c} + E_{P2',i,oc} \cdot D_{P2',i,oc}\right) \cdot \left(1 - k \cdot MS_{P2',i}\right)\right| \\
| ...$$
4a, 4b

where:

MS_{p,i} = the motor-vehicle-related fraction of emissions of precursor pollutant P' (P1', P2'...) from emissions source i; that is, of the emissions of pollutant P', from source i, MS_{p,i} is the fraction that is related to motor-vehicle use (e.g., all tailpipe emissions from motor-vehicles are related to motor-vehicle use; some fraction of refinery emissions is related to motor-vehicle use, and no fraction of emissions from agricultural tillage is related to motor-vehicle use) (estimated in Report #10 of this social-cost series)

k = 1.0 in the case in which 100% of motor-vehicle-related emissions are removed, and 0.10 in the case in which 10% of motor-vehicle-related emissions are removed

i = sources of emissions of P' (includes all sources in the emissions inventory: motor vehicles, power plants, industries, businesses, farms, and so on).

In the case in which we eliminate 100% of anthropogenic emissions, $E_{p^{,i}}$ is equal to emissions from natural sources.

Now, with two more adjustments, our model of pollutant formation will be complete. First, note that in equations 2, 3, and 4, we have a term for annual county-level emissions of pollutant P' from source i: $E_{p',C,i}$ (for the county C with the airquality monitor of interest) or $E_{p',OC,i}$ (for all counties except C in AQCR R_C). Now, the emissions data that we have are the EPA's (1995d, 1995e) official inventory of emissions in every county of the U. S., in 1990. (We discuss these estimates below.) Let us designate the official EPA county-inventory estimate of emissions of pollutant P' from source i as: $OEI_{p',C,i}$, or $OEI_{p',OC,i}$. It appears that most of these official inventory estimates — the OEI — are reasonably accurate. However, we do know that the official inventory (OEI) over— or under-estimates emissions of some pollutants from some sources. Therefore, in general, we will assume that the true county-level emissions of pollutant P' from source i ($E_{p',C,i}$, $E_{p',OC,i}$) are equal to the official estimate of emissions multiplied by a correction factor:

$$E_{P',i,c} = EC_{P',i} \cdot OEI_{P',i,c}$$

$$E_{P',i,oc} = EC_{P',i} \cdot \sum_{o \in R_c} OEI_{P',i,o}$$
(5)

where:

 $OEI_{p',i,C}$ = the EPA's official emission-inventory estimates of emissions of pollutant P' from source i in county C (data from EPA, discussed below)

 $OEI_{p',i,o}$ = the EPA's official emission-inventory estimates of emissions of pollutant P' from source i in county O (any county other than C in AQCR R_c) (data from EPA, discussed below)

 $EC_{p',i}$ = our emissions-inventory correction factor, equal to the ratio of our estimate of true emissions of pollutants P' from source i to the EPA's official estimate (discussed below; this factor is 1.0 for most sources i, and is assumed to be the same in every county).

Second, we will normalize the dispersion terms in equation 4, $D_{p',i,C}$ and $D_{p',i,OC}$, to the dispersion of direct emissions of fine PM from light-duty motor-vehicles in county C. We define a normalized dispersion, DN:

$$DN_{P',i,c} = \frac{D_{P',i,c}}{D_{fPM',LDV,c}}$$

$$DN_{P',i,oc} = \frac{D_{P',i,oc}}{D_{fPM',LDV,c}}$$

where:

 ${\rm DN_{p',i,C}}$ = the fraction of emissions of precursor pollutants P' from source i in county C that reach the ambient air-quality monitor in County C, relative to the fraction of direct emissions of fine PM from light-duty motor-vehicles in county C that reach the ambient air quality monitor in county C

 $DN_{p',i,OC}$ = the fraction of emissions of precursor pollutants P' from source i in all outside counties OC (all counties except C in AQCR R_C) that reach the ambient air-quality monitor in county C, relative to the fraction of direct emissions of fine PM from light-duty motor-vehicles in county C that reach the ambient air quality monitor in county C

Dfpm',LDV,c = the fraction of direct emissions of fine PM from light-duty motor-vehicles in county C that reach the ambient air quality monitor in county C

Note that the dispersion term always is normalized with respect to LDV emissions of fine PM in County C. That is, even the dispersion of emissions in all outside counties, OC, is normalized to the dispersion of LDV fine PM emissions in County C. Because every DN term -- for every pollutant, from every source and location -- is normalized with respect to the *same* Dfpm',LDV,C, we properly may add together *any* product of emissions (E) normalized dispersion (DN). Thus, the pollution contribution of emissions outside county C is additive with the contribution of emissions in County C, because both contributions are estimated with respect to the same baseline (Dfpm',LDV,C). Similarly, with all DN estimated relative to Dfpm',LDV,C, we may add up the contributions of fine PM, coarse PM, sulfate PM, and nitrate PM, where each contribution is estimated as the product of normalized dispersion and emissions, in order to determine the total contribution of different sources to total ambient PM10 (which consists of directly emitted fine PM, directly emitted coarse PM, and nitrates and sulfates).

We now have our final general model of ambient pollution, shown here for the case in which we eliminate 10% or 100% of motor-vehicle-related emissions:

$$PI_{P,c} *= D_{fPM',LDV,c} \cdot C_{P' \to P} \left(\sum_{i} PT \, 1'_{i}, \sum_{i} PT \, 2'_{i}, \dots \right)$$

$$PP_{P,c} *= D_{fPM',LDV,c} \cdot C_{P' \to P} \left(\sum_{i} PT \, 1'_{i} \cdot \left(1 - k \cdot MS_{P1',i} \right) \sum_{i} PT \, 2'_{i} \cdot \left(1 - k \cdot MS_{P2',i} \right) \dots \right)$$

$$\frac{PP_{P,c} *}{PI_{P,c} *} = \frac{C_{P' \to P} \left(\sum_{i} PT \, 1'_{i} \cdot \left(1 - k \cdot MS_{P1',i} \right) \sum_{i} PT \, 2'_{i} \cdot \left(1 - k \cdot MS_{P2',i} \right) \dots \right)}{C_{P' \to P} \left(\sum_{i} PT \, 1'_{i}, \sum_{i} PT \, 2'_{i}, \dots \right)}$$

$$PT \, 1' = EC_{P1',i} \cdot \left(DN_{P1',i,c} \cdot OEI_{P1',i,c} + DN_{P1',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P1',i,o} \right)$$

$$PT \, 2' = EC_{P2',i} \cdot \left(DN_{P2',i,c} \cdot OEI_{P2',i,c} + DN_{P2',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P2',i,o} \right)$$

where all terms are as defined above.

In the case in which we eliminate 100% of anthropogenic pollution, there are two changes to the numerator of the $PP_{p,C}^*/PI_{p,C}^*$ ratio: the (1-k·MS_{p1',i}) term is dropped, and the $OEI_{p1',i,C}$ become emissions of pollutant i from natural sources in county C.

Notice that the $D_{fpm',LDV,C}$ terms will cancel out when we take the ratio of PP to PI, in equation 1. Thus, we do not have to estimate any "absolute" dispersion factors; rather, we need estimate only dispersion factors relative to light-duty motor-vehicle dispersion factors (the DN terms). This is important because there is less uncertainty in estimating pollution dispersion from one source relative to another than in estimating dispersion per se.

In this most general form, the model applies to ambient pollutants, such as ozone (O_3) and secondary particulates $(PM_{2.5} \text{ and } PM_{10})$, that form via chemical reactions that involve emissions of precursor pollutants P'. However, in the case of ambient pollutants CO, NO₂, and "direct" PM_{10} and $PM_{2.5}$, we ignore atmospheric chemistry. In these cases, the ambient pollutants are the same as the emitted pollutants, and the model simplifies to:

$$PI_{P,c} *= D_{fPM',LDV,c} \cdot \sum_{i} \left(EC_{P',i} \cdot \left(DN_{P',i,c} \cdot OEI_{P',i,c} + DN_{P',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P',i,o} \right) \right)$$

$$PP_{P,c} *= D_{fPM',LDV,c} \cdot \sum_{i} \left(EC_{P',i} \cdot \left(1 - k \cdot MS_{P',i} \right) \cdot \left(DN_{P',i,c} \cdot OEI_{P',i,c} + DN_{P',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P',i,o} \right) \right)$$

$$\frac{PP_{P,c} *}{PI_{P,c} *} = \frac{\sum_{i} \left(EC_{P',i} \cdot \left(1 - k \cdot MS_{P',i} \right) \cdot \left(DN_{P',i,c} \cdot OEI_{P',i,c} + DN_{P',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P',i,o} \right) \right)}{\sum_{i} \left(EC_{P',i} \cdot \left(DN_{P',i,c} \cdot OEI_{P',i,c} + DN_{P',i,oc} \cdot \sum_{o \in R_{c}} OEI_{P',i,o} \right) \right)}$$

There are sophisticated models of emissions, dispersion, and atmospheric chemistry. However, it is time consuming and expensive to run all of the best models for every region in the U.S. To keep our task manageable, we will:

- use the results from the best available emissions models;
- treat dispersion very crudely;
- use an extremely simple nonlinear model of tropospheric ozone chemistry;
- greatly simplify tropospheric aerosol chemistry.

As we stated in the beginning of this report, we will use our air-quality model to estimate the change in air quality for our dose-response functions for human health (Report #11), crop damages (Report #12), and visibility (Report #13). The application of the model is virtually identical in all three cases (human health, crops, and visibility). In the case of human health and visibility, we model pollution at urban air-quality monitors, because health and visibility costs are greatest in urban areas (broadly defined, to include suburban areas). In the case of crop damage, we model pollution at agricultural monitors. As we shall see in section 16.3, this dichotomy (urban or agricultural) affects but one parameter in the entire model -- the distance from the emissions source to the receptor (the air-quality monitor).

In the remainder of this report, we present our analysis of emissions, emission-correction factors, dispersion, and atmospheric chemistry. As a check, we will compare our estimates of the motor-vehicle contribution to ambient pollution with analyses of the chemical composition of pollution captured at ambient air-quality monitors.

16.2 ESTIMATES OF EMISSIONS: THE EPA'S OFFICIAL EMISSIONS INVENTORY (OEIP',I,C), AND OUR CORRECTIONS TO THE EPA ESTIMATES (ECP',I)

16.2.1 Background

The EPA (1995d, 1995e) has produced a detailed, county-by-county emission inventory, which provides estimates of emissions of all criteria pollutants, from a wide variety of biogenic and anthropogenic sources, for every county in the U.S. (The 1995d report has the inventory for PM, VOCs, NO_X , and SO_X [biogenic emissions excluded], and the 1995e report has the inventory for biogenic emissions of VOC and NO_X .) We use these estimates as our starting point in estimating the motor-vehicle contribution to ambient air pollution. However, even though these official estimates are the best that have been published, many of them are very uncertain, and a few are thought to be seriously in error.

Consequently, we examined the uncertainty of some of the emissions estimates in the EPA inventory. If an official estimate of emissions of some pollutant, P', from source i seemed accurate, or if we did not have any reason to question it, we used it as is in equations 6 or 7 above -- that is, we implicitly assigned a value of 1.0 to the correction factor, $EC_{p',i}$, for that pollutant from that emissions source. Otherwise, we estimated a correction factor (other than 1.0) to apply to the official estimate to make it, in our view, more accurate.

In the official inventory, emissions calculated as the product of an emission factor, which is given in grams of emission per unit of activity (e.g., grams per mile of travel by light-duty cars), and total activity (e.g., miles by light-duty cars):

*Emissions = emission factor (grams emitted/unit activity) * units of activity.*

Uncertainty in emissions estimates, then, is related to uncertainty either in the emission factors or in the activity levels.

It appears that most total activity levels are known reasonably well. For example, estimates of total vehicle miles of travel (VMT) -- the activity which is multiplied by gram/mile emissions (from a computer model called MOBILE5A) to produce total grams of emission -- probably are accurate to within better than 10%, although the uncertainty in the estimates of VMT by heavy-duty trucks might be greater than this (Guensler et al., 1991).

The emission factors, however, can be very uncertain. Emission factors for stationary sources (such as petroleum refineries) and area sources (such as road construction activities) are documented in the EPA's voluminous emission-factor handbook, known as AP-42 Volume 1 (EPA, 1995a). Emission factors for VOCs, CO, and NOx for the various classes of motor vehicles are estimated in grams/mile by an EPA computer model, called MOBILE5A. (California has its own version, called EMFAC7F.) Emission factors of PM and SO_X are estimated by a separate EPA computer model, similar to the MOBILE model, called PART5.

Our investigation of the uncertainty of emission factors used to estimate $OEI_{p',i,c}$ led us to the following conclusions.

- \bullet First, it is likely that the MOBILE5A model underestimates real-world gram/mile emissions of VOCs, CO, and NO_X from light-duty gasoline-powered motor vehicles.
- Second, it is possible that the PART5 model underestimates real-world PM emissions from heavy-duty diesel vehicles, although there is little evidence one way or the other.
- Third, it is very likely that AP-42 overestimates emissions of PM₁₀ road dust and substantially overestimates emissions of PM_{2.5} road dust.
- Finally, it is likely that AP-42 overestimates emissions of PM₁₀ and PM_{2.5} from road construction.

In the following sections we detail these conclusions, and develop the correction factors that we apply to the official emissions estimates (EPA, 1995d, 1995e) to produce what we believe are more accurate estimates.

16.2.2 Estimates of VOCs, NOx, and CO emissions from mobile sources (MOBILE5A model)

Background

The MOBILE5A computer model estimates gram/mile emissions of VOCs, CO, and NOx from several classes of gasoline and diesel-fuel vehicles. The model calculates emissions for a particular year, as a function of the mix of vehicles in the fleet, VMT by vehicle class, vehicle speed, ambient temperature, fuel characteristics, characteristics of inspection and maintenance programs, and other factors. The model is built on the basis of emissions tests of vehicles in use, which are tested mainly but not exclusively over a standardized drive cycle known as the Federal Test Procedure (FTP). The most recent

version of the model, MOBILE5A -- the version used to produce the county-by-county emissions estimates in the official inventory EPA, 1995d, 1995e) -- was released in 1993.

Shortcomings of the MOBILE model

By the late 1980s, evidence had accumulated that the then-current version of the EPA's emission-factor model, MOBILE3, greatly under-predicted emissions of VOCs and CO from light-duty gasoline vehicles. In 1991, a seminal report by the National Research Council (1991) concluded that "measurements from roadside tests, tunnel studies, and remote-sensing of in-use vehicles provide consistent and compelling evidence that vehicles on the road have substantially higher CO and VOC emissions than current emissions models predict" (p. 288). Analyses of the relative abundance of VOCs, CO, and NO_X in the atmosphere, and of the composition of ambient VOCs, also indicated that emissions of VOCs and CO from mobile sources were underestimated. The models appeared to underestimate VOC and CO emissions by a factor of 2 or 3.

The MOBILE3 performed poorly for several reasons (NRC, 1991; EPA, 1995b):

- 1). It underrepresented the proportion of vehicles with extremely high emissions (called "super emitters").
 - 2). It did not include running-loss and resting-loss evaporative emissions of VOCs.
- 3). It underestimated the rate at which emissions increase as a vehicle accumulates mileage.
- 4). It did not account for or properly represent so-called" off-cycle" emissions, which are produced at very high speeds and during hard accelerations and transient fuel enrichment.
- 5) It probably underestimated the total number of starts that occurred with a cool or cold catalyst.
- 6) It did not represent well the effect of air conditioning on emissions (the use of air conditioning greatly increases NO_X emissions).

In the late 1980s and early 1990s, the EPA conducted extensive testing of in-use vehicles, and revised subsequent versions of the model. Compared with MOBILE3, the most recent version of the model, MOBILE5A, has a more accurate representation of super-emitters, includes running and resting-loss emissions (MOBILE3 did not), and assumes that emissions increase much more rapidly with mileage (EPA, 1995b). As a result, the current version of the EPA's emission-factor model, MOBILE5A, predicts much higher emissions than did the previous versions, and appears to predict real-world emissions much more closely (EPA, 1995b; Auto/Oil Air Quality Improvement Program, 1995).

However, MOBILE5A still suffers from shortcoming 4) to 6) in the list above: it does not properly represent "off-cycle" emissions, it probably underestimates the total number of cold starts, and it does not represent well the effects of air conditioning

(EPA, 1995b; German, 1995)³. As a result, MOBILE5A still apparently underestimates CO, VOC, and perhaps NO_X emissions from light-duty gasoline vehicle, although not by nearly as much as did MOBILE3.

<u>Off-cycle emissions.</u> "Off-cycle" emissions, which result from speeds and transient conditions that are more extreme than those in any part of the standard FTP emissions test, are thought to be an especially large source of excess or under-predicted emissions. Recent studies of real driving patterns have shown that speeds and accelerations are higher than in the FTP (Ross et al., 1995; German, 1995).

During a hard acceleration, emissions per second increase by as much as a 1000-fold. In an illustrative calculation, Ross (1994) estimates that a hard acceleration occurs about once every 3 miles, and produces 28 grams of CO, and 0.7 grams of HC -- or 9 g/mi CO, and 0.23 g/mi HC. Similarly, Ross et al. (1995) estimate that "command enrichment," which is an increase in the fuel/air ratio in response to demands for high power, causes excess emissions of 7.3 g/mi CO and 0.12 grams/mile HC, under average driving. These excess emissions are large compared to the current emission standards of 3.4 g/mi CO, and 0.4 g/mi HC, and imply that MOBILE5A might be significantly underestimating real emissions (especially CO emissions). German (1995) suggests that the MOBILE5A also does not properly model off-cycle NO_X emissions.

<u>Number of starts with cooled down catalyst.</u> A cold catalytic converter does not catalyze reactions well, and hence does a poor job of reducing engine-out emissions. As a result, the tailpipe emissions from a cold vehicle are quite high, but drop fairly rapidly as the engine warms the catalytic converter to its effective operating temperature.

When an engine is turned off, the catalytic converter, which is heated by exhaust gases, begins to cool immediately, and is cold within 45 to 60 minutes (German, 1995). This behavior, combined with the poor performance of the catalyst when cold, means that five 1-mile trips one hour apart will produce much more pollution than does one 5-mile trip. In other words, gram/mile emissions -- which is what MOBILE5A estimates -- are a function of the total number of times a vehicle is started with a cold or cool catalyst.

It appears that in reality there are many more cold or cool starts than is assumed in MOBILE5A. The emission factors in MOBILE5A are based on the FTP, which is 7.5 miles long and assumes that 43% of all vehicle starts are "cold" starts. Recent limited data on trip patterns indicate that the *fraction* of trips that are begun with a cool or cold catalyst might be accurate, but that the average trip length is much less than 7.5 miles (EPA, 1995b; German, 1995). Assuming that total VMT is correct, this means that there are many more starts, and hence many more starts with cold or cool catalysts, than is assumed in MOBILE5A. This, in turn, means that the average emissions per mile are higher than estimated by MOBILE5A, because as mentioned above during cold start

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³Ross et al. (1995) provide an excellent discussion of real-world emissions from passenger cars, although they do not estimate the extent to which the MOBILE5A model mis-estimates emissions.

and cold-transient driving the catalytic converter is cold and relatively ineffective at reducing engine-out emissions.

<u>Air conditioning.</u> In most FTP tests, the vehicle's air conditioning is not on, and consequently the MOBILE5A emissions model, which is based largely on FTP emissions data, does not account for the effect of air conditioning on emissions. In recent tests reported by EPA (1995b), the use of air conditioning increased VOC emissions by 25%, CO emissions by 51%, and NO_X emissions by 92%, over the full FTP, albeit under extreme conditions of high temperature and high humidity. The large increase in NO_X emissions has come as something of a surprise, and by itself suggests that MOBILE5A might significantly underestimate drive-cycle, year-round average emissions.

How much is MOBILE5A off?

The EPA is gathering data on these problems to be able to improve the next version of the model, MOBILE6 (EPA, 1995b). In the meantime, though, we face the question of the extent to which the current mobile-source emission inventory, developed with MOBILE5A, still underestimates emissions of VOCs, CO, and perhaps NO_X from light-duty gasoline vehicles. Unfortunately, there are few quantitative estimates of the extent of the underestimation. The discussion above suggests that emissions of CO are substantially underestimated, and that emissions of VOCs are underestimated less than are emissions of CO. There is some evidence that under some conditions NO_X actually is overestimated (EPA, 1995b; Auto/Oil Air Quality Improvement Program, 1995), but when all of the factors discussed above (off-cycle emissions, cold starts, and especially air conditioning use) are considered, it is more likely that NO_X emissions will be found to be underestimated than overestimated.

The following four studies help us quantify the extent to which the MOBILE5A model might be in error:

1). A comparison of ambient ratios of $CO:NO_X$ and $VOCs:NO_X$ with emission ratios of $CO:NO_X$ and $VOCs:NO_X$ indicates that the 1991 version of California's emission model, EMFAC7E, underestimates mobile-source CO emissions by a factor of 1.5 and mobile-source VOC emissions by a factor of about 2.2 (Fujita et al., 1992). The subsequent version of EMFAC7F, similar to EPA's MOBILE5A, estimates higher VOC emissions than does EMFAC7E, but according to a recent study (Fujita et al., 1995) still underestimates mobile-source emission factors for VOCs. Fujita et al. (1995) used VOC profiles of motor-vehicle VOC exhaust and other VOC emissions sources to estimate the motor-vehicle contribution to measured ambient VOC concentrations in seven urban areas in the San Francisco Bay Area and San Joaquin Valley. They compared this estimated ambient contribution with the ratio of motor-vehicle VOC emissions (estimated using EMFAC7F) to total estimated VOC emissions in each area. Exhaust and evaporative emissions contributed 70 to 74% of the measured ambient VOCs in the seven urban areas (excluding biogenic VOCs and acetone), but only 43% of the estimated primary anthropogenic VOC emissions. (See also Magliano et al., 1993.)

There are three reasons why the ambient chemical-mass-balance source apportionment to motor vehicles might exceed the emissions-inventory apportionment

to motor vehicles: 1) in the source apportionment of ambient concentration, the portion attributed to motor-vehicles actually might include some non-vehicular sources that have a VOC profile similar to the motor-vehicle profile; 2) the ambient monitors used in the source apportioning might capture a greater percentage of motor-vehicle emissions than of other emissions, most likely because the monitors are closer to motor vehicles; 3) the motor-vehicle VOC emission factors might be underestimated. However, if underestimation of VOC emissions accounts for all of the discrepancy estimated by Fujita et al. (1995), then EMFAC7F underestimated VOC emissions by a factor of 3.4 (!), because motor-vehicle emissions would have to have been 3.4 times higher in order for their share of total emissions to have been 72% (assuming that all other sources in the inventory were correctly estimated). We believe, however, that part of the discrepancy between the 72% ambient share and 43% estimated emission-inventory share was due to the second possibility, that the monitors generally captured a larger fraction of motor-vehicle emissions than of other emissions. Thus, this study suggests that EMFAC7F underestimates VOC emissions by less than a factor of 3.4

- 2). Recently, German (1995) of EPA has made preliminary estimates of the extent to which in-use emissions from a low-emitting vehicle in the year 2020 will exceed the levels predicted by the current model, MOBILE5A. He estimates that VOC emission will be 1.15 times higher than predicted by MOBILE5A, that CO emissions will be 1.47 times higher, and that NO_X emissions will be 1.35 times higher.
- 3). The estimates, cited above, of Ross et al. (1995) -- that command enrichment causes excess emissions of 7.3 g/mi CO and 0.12 grams/mile HC, under average driving -- suggest that, on account of command enrichment alone, CO emissions might be underestimated by as much as a factor of 2.0, and HC emissions by a factor of at least 1.1.
- 4). Finally, a comparison of California's forthcoming updated emission-factor model, EMFAC7G, with the current version, EMFAC7F, gives some indication of the extent to which MOBILE5A underestimates real-world emissions. EMFAC7F, the current California model, is similar to EPA's MOBILE5A. The forthcoming update, EMFAC7G, accounts better for high-emitting vehicles, real-world driving patterns, inspection and maintenance programs, and the distribution of starts than does EMFAC7F. In other words, EMFAC7G accounts for many of the factors that cause MOBILE5A to underestimate real-world emissions. The ratios of EMFAC7G to EMFAC7F estimates of emissions from all vehicles in the South Coast Air Basin in summer 1990 are: VOCs 1.29, CO 1.81, and NO_X 1.41 (California Air Resources Board, 1995).

German's (1995) preliminary estimates pertain to a low-emitting vehicle in the year 2020. Because we are working with the 1990 emission inventory, we are interested in the extent to which MOBILE5A under-predicted emissions from a "fleet average" vehicle in 1990. We expect that generally, MOBILE5A under-predicts emissions from a fleet average vehicle in 1990 by *at least* as much as it under-predicts emissions from a low-emitting vehicle in the year 2020, because the fleet average vehicle in 1990 will be have higher baseline emissions, and greater variation in emissions as a function of the

drivecycle and the number of cold starts. In support of this, we note that the difference between EMFAC7G and EMFAC7F decreases from the year 1990 to the year 2000.

With these considerations, we assume, in our low-cost case, that actual emissions of CO from light-duty gasoline cars and trucks are 1.5 times higher than estimated in the official MOBILE5A-based inventory, that actual emissions of VOCs are 1.1 times higher, and that NOx emissions are 1.2 times higher. In our high–cost case, we assume that actual emissions of CO from light-duty gasoline cars and trucks are 1.8 times higher than estimated in the official MOBILE5A-based inventory, that actual emissions of VOCs are 1.3 times higher, and that actual emissions of NO $_{\rm X}$ are 1.4 times higher. These adjustments are summarized in Table 16-1 below.

<u>Corrections to VOCs, NO_X, and CO emissions from diesel vehicles or heavy-duty</u> <u>gasoline vehicles?</u> For several reasons, we believe that the MOBILE5A estimates of VOC, NO_X, and CO emissions from diesel vehicles and HDGVs are not seriously in error, and consequently do not make any corrections to the official inventory estimates of these emissions.

First, the MOBILE5A model underestimates emissions from LDGVs mainly because the emission control system of LDGVs is not very effective under certain conditions that are not well represented in the database underlying the MOBILE5A model. However, because diesel vehicles do not have catalytic converters, computer-controlled air/fuel ratios or evaporative control systems (because diesel fuel is not volatile), one would expect that emissions from conditions not represented in the MOBILE5A model would not be as radically different from emissions under modeled conditions as is the case with LDGVs when the emission control system essentially stops working.

Second, the available data show that diesel vehicles do not produce significant emissions of CO or VOCs anyway.

Third, recent tunnel studies indicate that MOBILE5A predicts NO_X emissions from diesel vehicles reasonably well (Auto/Oil Air Quality Improvement Program, 1995).

Therefore, we assume that the MOBILE5A model accurately predicts VOC, CO, and NO_X emissions from diesel vehicles, and make no correction to the diesel-vehicle emissions inventory of these pollutants. We assume also that MOBILE5A model accurately predicts emissions of these pollutants from heavy-duty gasoline vehicles, and so make no correction to that inventory either.

16.2.3 Estimates of PM and SO_X exhaust emissions from mobile sources (PART5 model)

The EPA's PART5 model, similar in structure to the MOBILE5A model, calculates gram per mile exhaust emissions of PM and SO_X from the same classes of vehicles included in the MOBILE5A model discussed above. In addition to exhaust PM and SO_X emissions, PART5 estimates road dust and tirewear emissions, although it does not estimate brakewear emissions. We discuss corrections to the road dust

emissions inventory in Section 16.2.4. Tirewear and brakewear are relatively insignificant and we do not attempt to make any corrections to the inventory. We focus on exhaust emissions in this section. SO_X exhaust emissions are calculated on the basis of the sulfur content of the fuel, which is known reasonably accurately, and with the reasonable assumption that most sulfur is burned to SO_2 and a small fraction to sulfate (EPA, 1995c). Hence, the estimates of SO_X exhaust emissions are fairly accurate.

However, the estimates of PM exhaust emissions are suspect. We are concerned most about the accuracy of the estimates of PM exhaust emissions from heavy-duty diesel vehicles (HDDVs), because HDDVs emit many times more PM per mile than do light-duty gasoline vehicles. It appears that PM exhaust emissions from diesel vehicles are calculated on the assumption that the vehicles meet the pertinent model-year PM emission standards, with little if any deterioration over time (Walsh, 1995; this can be confirmed by comparing the output of PART5 with the relevant emissions standards). Guensler et al. (1991) and Ostria (1996) describe the history of the testing.

In 1983 and 1984, the EPA tested 30 in-use heavy-duty diesel engines. The engines were removed from their chassis, and tested "as is" (i.e., without being tuned up) over the heavy-duty transient cycle for new engines, on an engine dynamometer. The results for eight of the engines were problematic, and discarded. The test results for the remaining 22 engines (9 medium-heavy and 13 heavy-heavy engines) were used to establish the EPA's official PM emission factors (Guensler et al., 1991; Ostria, 1996).

Each engine was tested one to four times. For the heavy-heavy engines, average PM emissions ranged from 0.58 to 2.14 g/bhp-hr (Guensler et al. 1991). (It is not clear if this is TSP or PM10.) Assuming a conversion factor of 2.3 (1982 model year) to 3.0 (1978 model year) bhp-hr/mi (EPA, 1985), we calculate PM emissions of 1.33 to 6.42 g/mi.

After these initial tests of the 22 engines "as received", the EPA tuned up and retested 7 of the medium-heavy and 6 of the heavy-heavy engines. After this tune up, the engines emitted more NO_X but less HCs (Guensler et al., 1991). Because PM emissions generally change in the same direction as do HCs, and the opposite direction from NO_X , we can presume that the PM emissions also decreased after tune-up.

It is not clear which set of test results -- before tune up, or after tune up -- the EPA used to establish its baseline emission factors. Guensler et al. (1991) speculate that the official emission factors are based on the results of the tests conducted *after* the engines were tuned up. In support of this, we note that PART5 estimates that in 1983, HDDVs emitted 2.1 g/mi PM, which is at the low end of the range of emissions from the 13 heavy-heavy engines tested "as is" in 1983/1984 (1.33 to 6.42 g/mi, above). If PART5 is based on the results for tuned-up engines, then obviously it understates real-word PM emissions.

Perhaps more seriously, it appears to us that none of the engines tested were super-emitters, because even the highest level measured in the tests (2.14 g/bhp-hr) is less than one would expect from a badly smoking engine. Given that the small amount of super-emitters that one typically observes in a fleet can significantly raise fleet-average emissions, the omission of super-emitting engines from the emissions tests would have resulted in a significant underestimate of real-world emissions.

The 22 engines tested in 1983 and 1984 had accumulated from 29,000 to 410,000 miles at the time of testing (Guensler et al., 1991). It is not clear, however, if the mileage distribution was representative of the fleet average at the time, or if the EPA accounted for the effect of mileage in establishing its baseline emission factors (Guensler et al., 1991). In fact, in general, it is not clear if the vehicles selected were broadly representative of the in-use fleet.

Guensler et al. (1991) also note that the trucks in the real world may idle more than is assumed in the HDTC, and that the emissions inventory apparently does not account for emissions from truck engines being run to provide auxiliary power for refrigeration and other purposes.

It appears, then, the PART5 PM emission factors are based on emissions from a small number of tuned-up engines driven over a somewhat idealized test cycle. Now, to some extent it is reasonable to assume that emissions from HDDVs don't get significantly worse as the vehicles age, because HDDVs do not have the elaborate emission-control hardware that LDGVs have. However, HDDVs that are out of tune, run poorly, or driven in ways not properly represented in the emissions tests, probably will have significantly higher exhaust emissions than are measured during the engine certification tests.

The PART5 model has input variables for the drive cycle (cruising or transient), vehicle speed, altitude (high or low), and inspection & maintenance (I&M) (in force or not) (EPA, 1995c). This implies that the PART5 model estimates PM emissions for different drive cycles, vehicle speeds, and so on. However, it turns out that this is not the case. Virtually all of the PM emissions in PART5 are a function only of the model year and type of vehicle.

In PART5, exhaust PM emissions from HDDVs -- which contribute the bulk of PM emissions from the highway transportation sector -- depend only on the model year of the vehicle. They do not depend on drive cycle, speed, altitude or inspection & maintenance (I & M). The model produces a single emission factor for every model year, everywhere in the country, on every road. It appears that the emission factors are based on the results of vehicles tested over the official heavy-duty-vehicle test cycle, the heavy-duty transient test cycle (HDTC).

The calculation of exhaust emissions from LDVs is more complicated. LDV PM emissions are equal to lead + sulfate + carbon PM emissions. Carbon emissions, which usually are the largest fraction of the total, depend only on model year and vehicle type. Lead emissions depend on the model year, speed, and drive cycle, and other factors. However, in all our scenarios, lead emissions are zero or trivial, because there is so little unleaded gasoline. Direct sulfate emissions depend on speed (but not drive cycle), type of gasoline, and other factors. However, the sulfate emissions don't vary that much by speed. Thus, virtually all PM emissions from LDVs are a function only of model year and vehicle type. Most likely, the emission factors for LDVs are based on the results of vehicles tested over official light-duty-vehicle test cycle, the Federal Test Procedure (FTP).

Thus, the PART5 model has, in effect, "fixed" PM emission exhaust factors for different vehicle classes and model years-- factors that are independent of the speed, drive cycle, and altitude.

Now, given this, how might the Emission Inventory (EI), which is calculated with PART5, be wrong? There are two ways in which the EI might be wrong. First, the emission factors themselves, for whatever drivecycle was used, might be inaccurate, most likely because the tested vehicles were not representative. For example, perhaps no super emitting vehicles were included in the data base from which the emission factors for PART5 were taken. Second, the drive cycle might not be representative of the "average" driving conditions in the country, which presumably it is supposed to be. For example, there might be a lot more idling and hard accelerating in the real world than is present in the HDTC.

We suspect that both problems are present: that there are more high-emitting vehicles in the real world than are tested in the FTP and HDTC, and that there is more high-emitting driving in the real world than in the drive tests. Our impression is that the first problem, super-emitters, is more serious.

Ideally, to check PART5, one would compare its predictions with exhaust emissions data taken from in-use "as-is" vehicles, tested over a variety of realistic driving conditions. To our knowledge, few if any such data exist, mainly because they are expensive to collect. (And if there were such data, PART5 probably would have incorporated them.) As we discuss next, the available emissions data suggest, but certainly do not prove, that PART5 is underestimating exhaust emissions of PM.

Evaluating the accuracy of PART5 exhaust emission estimates

Chassis dynamometer tests of in-use vehicles. Recently, the Desert Research Institute (Sagabiel, 1995) measured exhaust emissions from 23 high-mileage, in-use light-duty gasoline vehicles, over the IM240 emissions test, and found that PM exhaust emissions: A) varied by over two orders of magnitude⁴, and B) generally were much higher than predicted by PART5 (Table 16-2). These results are important because they pertain to high-mileage in-use vehicles, pulled off of the road and tested without modification. Six of the vehicles smoked visibly, and emitted about ten times more PM than did vehicles that didn't smoke. Even the non-smoking vehicles, however, emitted more PM than predicted by PART5 (0.05 g/mi in the tests versus 0.02 g/mi predicted by PART5). These results suggest that PART5 models relatively new, properly functioning light-duty gasoline vehicles, not a real "in-use" fleet with some old or malfunctioning vehicles. We expect that this conclusion applies to heavy-duty diesel vehicles as well.

21

65% of the emissions.

 $^{^4}$ At least one earlier study corroborates this. Hanson and Rosen (1990) measured aerosol black carbon in the exhaust of gasoline vehicles driving up a hill in Berkeley in 1985, and found that emissions varied by more than two orders of magnitude, and that 20% of the vehicles -- the "high emitters" -- accounted for

If we assume that some of the fleet are old or malfunctioning vehicles ("superemitters"), then the total levels of emissions are much higher than those predicted by PART5. About 10% of the fleet are super-emitters (the results from Sagabiel suggest that the fraction of super-emitters could be higher)⁵, and super-emitters emit roughly five to ten times more than normal vehicles. Now, if we start with the assumption that the normal vehicles emit 0.02 g/mi, as assumed by PART5, we end up with LDGV fleet emissions being 1.4 to 1.9 times higher than predicted by PART5. Alternatively, if we assume that the normal vehicles emit 0.05 g/mi, we end up with LDGV fleet emissions 3.5 to 4.75 times higher than predicted by PART5.

The EPA has measured PM exhaust emissions from in-use heavy-duty diesel vehicles (HDDVs) and heavy-duty gasoline vehicles (HDGVs), driven over the transient test cycle on a chassis dynamometer (Black et al., 1984; Dietzmann et al., 1980). The test results, and the corresponding predictions from PART5, are shown in Table 16-3. PM emissions from the in-use HDDVs vehicles increase with increasing mileage, and at 60,000 miles -- well below the midpoint of the life of an HDDV -- already are at or above the level predicted by PART5. This suggests to us that a fleet of HDDVs, with more than 100,000 miles of travel on average, emits more exhaust PM than is predicted by PART5. Of the five HDGVs tested, four emitted close to the amount predicted by PART5, but three of these had new or nearly new engines. The fifth HDGV emitted several times more PM than predicted by PART5. Thus, we expect, again, that a real in-use HDGV fleet, with some high-mileage vehicles, and a few high-emitting vehicles, will emit considerably more PM than is predicted by PART5.

In sum, tests of in-use vehicles indicate there is very wide range of PM exhaust emissions, and that on average the whole fleet in-use emits much more than do the new, properly functioning vehicles which seem to be the basis of the PART5 model.

<u>Measurements of on-road emissions</u>. We have found three studies of on-road emissions from HDDVs. In 1983, Pierson and Brachaczek measured the ambient airborne PM at the exit of the Allegheny and Tuscarora Mountain Tunnels on the Pennsylvania Turnpike, and with these and other data, back-calculated the HDDV emission rate⁶. More recently, Whittorf et al. (1994) and Gertler et al. (1995) reported the results of a similar experiment at the Fort McHenry Tunnel in Baltimore, Maryland. Finally, Balogh et al. (1993) measured the PM concentration along a university road that had heavy bus traffic, and back-calculated the bus emission rate.

In Table 16-4, we compare the results of these studies with the estimates of the PART5 model specified for the same conditions. In all three cases, PART5 estimates of

⁵ Regarding CO emissions, Ross et al. (1995) classify vehicles in two groups: 90% of the vehicles emit CO at about the normal FTP-measured rate, and 10% emit at a much higher rate.

⁶Pierson and Brachaczek (1983) summarize the method: "Known traffic and air fluxes are combined with net (tunnel minus intake) tunnel-air pollutant concentrations to derive mg/km emission rates of the various species observed. Correlation against the changing traffic composition gives emission-rate estimates resolved as to vehicle type" (p. 2).

PM exhaust emissions are comparable to those in the original studies, after we corrected the original studies so that they estimate exhaust emissions in the transient drive cycle. (Details of the corrections are given in the Table 16-4.) These results are consistent with the fact that PART5 appears to take little account of super-emitters, because the majority of emissions from super-emitters occur during transient driving. Since the field studies examined vehicles that are cruising, super-emitters should not create a large discrepancy between the PART5 and field-study estimates.

The ratio of exhaust PM to road-dust PM in the emissions inventory versus the same ratio measured at ambient air-quality monitors. As discussed below, the ratio of emissions of road dust to exhaust emissions from highway vehicles, in the EPA's (1995d) emissions inventory, is many times higher than the ratio of dust to motor-vehicle exhaust at ambient air-quality monitors. If the ambient ratios are accurate, and if the differences between the ambient ratios and the emissions ratios cannot be explained by differences in emissions dispersion, then the AP-42-based estimates of road-dust emissions are too high, or the PART5-based estimates of highway-vehicle PM emissions are too low, or, most likely, both occur.

<u>PART5 versus EMFAC7F.</u> One basis, albeit still a weak one, for quantifying the degree to which PART5 underestimates exhaust emissions from HDDVs is a comparison of the PM emission factors from PART5 with the PM emission factors from California's emission-factor model, EMFAC 7F. We ran PART5 and EMFAC7F for the year 1990, and got the results shown in Table 16-7. The EMFAC7F estimates of exhaust PM from HDDVs are about 1.8 times as high as the PART5 estimates. Although, the EMFAC7F tirewear estimates are at least an order of magnitude higher than the PART5 estimates, this does not qualitatively affect the results since tirewear is a small fraction of emissions.⁷

Why are CARB's EMFAC7F estimates higher than the EPA's PART5 estimates? According to Guensler et al. (1991), CARB had used the EPA's estimates until 1988, when CARB modified the EPA emissions factors to reflect inspection and maintenance practices in California. CARB developed its new estimates for EMFAC7F on the basis of a report by Radian Corporation, which reviewed the original data used to establish the

⁷However, we suspect that neither PART5 nor EMFAC7F is correct about tirewear: PART5 assumes that tirewear emissions are proportional simply to the number of wheels, so that a bus is predicted to have the same emissions as does a car, and only twice the emissions of a motorcycle. It is inconceivable that a bus emits only twice as much tirewear PM as does a motorcycle. EMFAC7F is more realistic in this respect, in that it estimates the same tirewear emissions for buses as for HDDVs. However, for two reasons, the EMFAC estimates appear to us to be too high all the way around.

First, back-of-the envelope calculations of the total amount of tire material worn away from tires suggests that the wear rate per mile is much less than is estimated by EMFAC7F. Second, in a study of ambient particulate matter associated with motor-vehicles in an expressway tunnel, Pierson and Brachaczek (1983) estimated that tires contributed only 1% of the total motor-vehicle PM emission rate of about 0.30 g/mi. The PART5 model estimates 0.008 g/mi PM from LDGV tires, and 0.036 g/mi PM from HDDV tires; for the 80% LDGV/20% HDDV traffic mix in the Pierson and Brachaczek (1983) study, PART5 estimates about 0.014 g/mi -- several times higher than the estimate of 0.003 g/mi from Pierson and Brachaczek (1983). The EMFAC7F estimates, of course, are even further off.

EPA (PART5) factors, plus additional information. The Radian report apparently estimated a factor to adjust the EPA's estimates upwards to account for high emissions from poorly maintained vehicles (Guensler et al, 1991). This adjustment factor might partially explain why the EMFAC7F estimates are so much higher than the PART5 estimates.

Our conclusion

The data reviewed above suggest that PART5 underestimates emissions from real on-road vehicles, primarily because PART5 takes little, if any, account of superemitters. In our low-cost case, we assume that the PART5 model underestimates PM emissions by a factor of only 1.5. In our high-cost case, we assume that PART5 underestimates emissions by a factor of 2.0.

16.2.4 Estimates of PM dust from paved roads (AP-42 Volume 1, and PART5 model)

Motor vehicle traffic kicks up the dust on the road. Some of this "emitted" road dust is small enough to be suspended in the air as particulate matter. Surprisingly, such "re-entrained road dust," as it is called, is by far the largest source of particulate matter in the official emissions inventory for the United States (EPA, 1995d): in 1990, road dust from paved and unpaved roads accounted for nearly half of all PM10 emissions in the U.S. emissions inventory.

Because road dust apparently is such a large source of emissions, it is important to determine if the emission factors used to calculate road-dust emissions are accurate. In this section, we present evidence that the current EPA (1995a) AP-42 emission factors, used in the PART5 model, substantially overestimate emissions of PM₁₀ and especially PM_{2.5} from paved roads. (We briefly discuss emissions from unpaved roads in the following section)

The paved road-dust equations

In the official emissions inventory, emissions of road dust from paved roads (RDP) are calculated with the following formulas:

$$TP = RDP + Ta + Ti + B =$$
 (D1)

$$k (sL/2)^{0.65} (W/3)^{1.5}$$
 (D2)

$$RDP = TP - Ta - Ti - B$$
 (D3)

where:

TP = total PM emissions due to motor vehicles on paved roads: tailpipe PM + road-dust PM + tire-wear PM.

RDP = emissions of road-dust particulate matter from paved roads (g/mi)

Ta = tailpipe emissions of PM (grams/mile; calculated from the PART5 model, discussed above)

- Ti = tire-wear emissions of PM (grams/mile; given in grams/mile for various vehicle classes, in the PART5 model)
- B = brake-wear emissions of PM (grams/mile; assumed to be zero in the application of equations D1 and D2
- k = multiplier to obtain different PM size classes (EPA, 1995a; e.g., to get emissions of TSP, k = 38; to get emissions of PM₁₀, k = 7.3; to get emissions of PM_{2.5}, k = 3.3)
- sL = the silt loading on the surface of the road (grams/meter²) (values from EPA, 1995a)
- W = the average weight of vehicles on the roadway (tons).

Equation D2 is presented in AP-42 (EPA, 1995a), and equation D3 (without the brakewear term B) is given in the PART5 model. It is important to note that dust emissions from paved roads are calculated by subtracting tailpipe (Ta), tirewear (Ti), and brakewear (B) emissions from empirically estimated *total* emissions from motor-vehicle traffic on paved roads (TP). Contrary to the implication in the emission-factor handbook, AP-42 (EPA, 1995a), equation D2 does not predict road-dust emissions per se; rather, it predicts total motor-vehicle-related emissions. (AP-42 is misleading because it presents equation D2 but not equation D3, and states that equation D2 predicts "dust emissions from vehicle traffic on a paved road" (p. 13.2.1-1). However, the equations are applied correctly in the EPA's PART5 model.) Thus, in order to assess the accuracy of emission inventory for paved road dust, we must evaluate the components of equation D3: total emissions, tailpipe, tirewear, and brakewear.

Possible sources of error in the paved-road-dust emission-factor equations

<u>Total PM emissions due to motor vehicles on paved roads (TP).</u> Again, the AP-42 equation for dust emissions from paved roads, equation D2 above, does not estimate road-dust emissions per se, but rather all PM in a plume attributable to motor-vehicle traffic. The equation is derived from a regression analysis of many calculated emission rates. The emission rates are calculated on the basis of PM concentrations measured in the plume of total PM emitted by traffic on paved roads. To characterize the PM plume, the investigators place PM monitors at various positions downwind and upwind of traffic, and measure the concentration gradient. The difference between downwind and upwind concentrations is input into a dispersion equation to back-calculate the PM emission rate (Midwest Research Institute, 1993). Because the investigators are measuring ambient PM in a plume emanating from the road, they are capturing tire wear and exhaust PM and even brakewear as well as road dust.

As just mentioned, the most recent AP-42 emission factor (equation D2) was estimated from a regression analysis of emission rates calculated from plume measurements of PM. In 1993, Midwest Research Institute (MRI, 1993) compiled all of the available PM emission tests, including relatively old tests used to estimate earlier versions of the emission-factor equation. (Most of the tests were conducted in the west and midwest of the U.S.) After screening out the unreliable tests (for example, those that

did not have enough PM samplers to reliably characterize the plume), MRI had 65 emission tests for PM₁₀ (fewer for PM_{2.5}). For each test they had the total PM emission factor as the dependent variable, and silt loading, mean vehicle weight, mean vehicle speed, and mean number of wheels as possible explanatory variables. Using stepwise multiple linear regression, they estimated various emission models, and found that the best model was equation D2 above: emissions as a nonlinear function of silt loading and vehicle weight.

Given this, we wish to know how this emission-factor equation might err, particularly, how it might over-predict real emissions, as seems to be the case. In the following, we discuss four possible sources of error in the estimation of the emission-factor equation, plus an additional source of error in the estimation of the emission inventory.

1). In the original field studies, the ambient PM concentration, used to characterize the PM plume, might have been measured incorrectly by each sampler. Logically, the "first" source of potential error is analytical or instrumental: the actual PM measurements taken by each sampler (in the studies used to develop the emission-factor equation) might have been wrong. If the PM measurements were wrong, then of course the calculated emissions were wrong.

We are unable to evaluate this possibility ourselves. However, Cowherd (1995a) of MRI, which performed many of the original field tests, believes that generally the measurements were reasonably accurate. According to Cowherd (1995a), PM was measured by a high-volume cascade sampler, with 3 substrates. A residual-allocation procedure was used to account for overloaded filters. Still, Cowherd (1995a) points out that the PM concentrations along the roads were relatively high -- higher than normal ambient concentrations measured away from roads -- and speculates that the samplers might have measured the relatively high concentrations less accurately. In any case, he believes that the samplers were more likely to have given incorrect results for PM2.5 than for PM10.

2). The PM plume was not correctly characterized, because too few measurements were taken. As mentioned above, the emission rates are derived from the observed plume. If the plume is not accurately characterized, then the back-calculated emissions are wrong.

This is a potentially serious source of error. Ideally, one should characterize a plume by taking simultaneous measurements in several different locations at several heights. However, some studies have sampled PM at only one or two points along the road. For example, Balogh et al (1993), who argued that the then-current AP-42 emission factor equation overestimated emissions by at least an order of magnitude, used only one downwind monitor to characterize the PM2.5 plume from the roadway. They acknowledged that this was the "critical limitation" in their study, and could have introduced as much as a two- or three-fold error (p. 30).

However, this probably was not so great a source of error in the estimation of the AP-42 emission-factor equation, because the data base used in the regression that

produced the equation included only those field tests in which at least four downwind monitors had been used (MRI, 1993).

3). The value of atmospheric parameters in the dispersion equation used to calculate the emission rate in the original studies might be incorrect. The back-calculated emission rates depend heavily on how one specifies the dispersion equation that relates emissions to measured concentrations.

In their review of the original emission studies, MRI (1993) notes that in two studies the specification of the dispersion equation resulted in "conservatively" high estimates of emission rates. However, MRI (1993) excluded those studies from its regression analysis, and we have no basis for believing that the studies that were included in the MRI database systematically mis-specified the dispersion models used to calculate emission rates.

4). The roads in the emission-test database might not be representative of most roads in the U.S. As mentioned above, most of the emissions tests were conducted many years ago, on paved roads in the west and midwest. It seems likely that roads in the west and midwest have more dust on the surface than do roads in the eastern U.S.. Also, according to Cowherd (1995b), MRI, which performed many of the original emissions tests, tended to look at relatively dirty roads. Moreover, Cowherd (1995b) of MRI believes that roads today generally are cleaner than were roads 15 or 20 years ago. Together, these considerations suggest that the emission-test database used to develop the AP-42 emission-factor (equation D2 above) might comprise roads that generally had much more silt than does the average road today. If so, then an equation estimated on a more representative database might have had a functional form different from that of equation D2.

These four sources of error apply to the emission-factor equation itself. However, it is possible that the equation is correct, but that the emissions inventory nevertheless is not; that is, that there is an additional potential source of error in the application equation of D2 to estimate the emissions inventory:

5). In the estimation of the emissions inventory, the EPA might assume incorrect values for sL, the silt loading, in equation D2. Equation D2 is used to estimate motor-vehicle PM emissions in every county of the United States. To do this estimation, the EPA must assume the mean vehicle weight (W) and the mean silt loading (sL) on every class of road in every county. The mean vehicle weight can be estimated reasonably accurately from complete, detailed national data on vehicle travel by vehicle class and type of road. However, there is no national inventory of silt loading. Rather, the EPA (1995a) has a very limited database for sL, which must be extended to types and locations of roads not covered in the data base.

The EPA (1995a) emission-factor handbook presents the results of measurements of sL, on a variety of roads, and suggests generic values to be used to calculate emissions in the absence of site-specific data. However, most of the measurements were of roads in Montana, and nearly all of the remaining measurements were in Colorado, Utah, Nevada, and Arizona. Moreover, there are very few measurements of the silt

loading on freeways. Thus, there are no data on the silt loading of roads in either the East Coast or the West Coast where most people live.

It is likely that the silt loading on roads in the major urban centers of the East Coast or West Coast is less than the loading on roads in Montana. If it is, then the emission inventory, which presumably is calculated by assuming generic values for sL derived from the available data in AP-42, overestimates PM emissions. We believe that this is the major source of error in the inventory of emissions of dust from paved roads.

<u>Tailpipe, tirewear, and brakewear emissions of PM.</u> Equation D3 shows that road-dust emissions of PM are equal to total motor-vehicle emissions minus tailpipe, tirewear, and brakewear emissions of PM. Thus, if any of these last three are misestimated, then paved-road-dust emissions are mis-estimated.

Above, we suggest that the PART5 model might underestimate tailpipe emissions of PM from heavy-duty diesel vehicles. If this is true, then by equation D3, emissions of road dust are overestimated.

The PART5 model estimates considerably lower emissions of tirewear than does California's EMFAC7F (Table 16-7). If EMAC7F is correct, then road-dust emissions again are overestimated. However, as noted above, we believe that the EMFAC7F estimates are too high. The PART5 estimate is incorrect in calculating tire wear as a function of the number of wheels, and there is some evidence that even the PART5 estimates are too high (Pierson and Brachaczek, 1983). In any case, tirewear emissions are a relatively small fraction of total emissions.

The EPA apparently assumes that in equation D3, brakewear emissions are zero, probably on the grounds that in the emissions tests used to estimate the parameter TP in equation D2, few vehicles were braking. We do not know whether brakewear emissions really should be zero in equation D3, but we do believe that it does not matter, because brakewear emissions are much smaller than tailpipe or road-dust emissions (Cha et al., 1983; Pierson and Brachaczek, 1983; EPA, 1985; Watson et al., 1994b; PART5 model).

Overall, we believe that tailpipe emissions of PM from HDDVs might be significantly underestimated. If so, then by virtue of equation D3, calculated PM dust emissions from paved roads are overestimated.

<u>Summary of possible sources of error:</u> The foregoing indicates: i) that it is possible that PM2.5 was inaccurately or insufficiently measured in the original emissions tests; ii) that the emission-factor equation might apply only to paved roads with a relatively high silt loading; iii) that most paved roads actually have a lower silt loading than is assumed in the calculation of the emission inventory; and vi) that emissions from HDDVs might be a larger part of total road-dust + vehicle emissions than currently is estimated.

In the next section, we attempt to estimate the degree to which emissions of PM dust from paved roads are overestimated.

Analysis of evidence that PM dust emissions from paved roads are overestimated

There is compelling evidence that the paved-road-dust emission factors in AP-42 (EPA, 1995a), and the current emissions inventories based on those factors, overestimate emissions of PM₁₀ and PM_{2.5} from paved-road dust. First, the ratio of road-dust PM actually measured in ambient air to motor-vehicle-exhaust PM actually measured in ambient air is a lot less than the ratio of estimated paved-road-dust emissions to estimated motor-vehicle exhaust emissions. Second, it appears that the AP-42 equation substantially over-predicts PM emissions from roads in the Eastern and Western U. S. Third, relatively recent measurements of the size distribution of paved-road-dust particulate matter indicate that the PM_{2.5}/PM₁₀ ratio assumed in the emissions inventory is too high. We discuss each of these next.

I. <u>Chemical-Mass-Balance (CMB) source apportionment studies versus the emissions inventories</u>. In the emissions inventory, emissions of PM₁₀ from paved roads in urban areas are almost 20 times higher than exhaust emissions of PM₁₀ from highway vehicles, and emissions of PM_{2.5} from paved roads are about 10 times higher than emissions of PM_{2.5} from highway vehicles (Table 16-8). This is because within the range of vehicle weights and silt loadings typically specified, the PART5 emission model estimates that gram per mile emissions of paved-road dust are an order of magnitude higher than gram per mile exhaust emissions. However, chemical analyses of the sources of PM in ambient air (called "chemical-mass-balance source-apportionment" studies) indicate that the ambient concentration of PM₁₀ road dust (actually, geological material) is less than twice the ambient concentration of PM₁₀ from motor-vehicles, and that the ambient concentration of PM_{2.5} road dust (geologic material) is much less than the ambient concentration of PM_{2.5} from motor vehicles (Table 16-11).

Table 16-9 shows the contribution of geologic sources, motor-vehicle exhaust, secondary particulates, and miscellaneous emissions sources to measured ambient PM₁₀ levels, as determined by chemical mass-balance (CMB) models, in many cities of the U.S. Table 16-10 shows the same for PM_{2.5}. In Table 16-9, geologic PM₁₀, which includes dust from wind erosion, unpaved roads, and agriculture, as well as dust from paved roads, accounts on average for 32-35% of all PM₁₀ (depending on whether one estimates the share with respect to explained or measured PM₁₀). Exhaust emissions of PM₁₀ from motor vehicles account on average for about 20% of all PM₁₀8. Thus, the ratio of all ambient geologic PM₁₀ to all ambient motor-vehicle-exhaust PM₁₀ is less

29

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⁸Analyses of specific "marker" pollutants give similar percentage shares to motor vehicles. Rogge et al. (1995) used hopanes and steranes as markers for vehicular emissions in Los Angeles, and found that 11% to 26% of atmospheric fine particle mass could be attributed to vehicular exhaust.

than 2:1 -- quite a bit lower than the ratio of emitted paved-road dust to emitted highway-vehicle exhaust, in the official PM₁₀ emissions inventory (OEI) (Table 16-8)⁹.

In theory, there are four reasons that the CMB source-apportionment ratio might be lower than the emissions-inventory ratio. First, the "primary geologic" category in the CMB studies probably includes more than just paved-road dust. However, in many CMB analyses, the chemical composition of "primary geologic" material is taken to be the composition of road dust, so that what is being identified as "geologic" most likely is mainly road dust. Moreover, the "motor-vehicle exhaust" category in the CMB studies undoubtedly includes some PM from off-highway mobile sources that produce PM species very similar to those produced by mobile sources. (In the emissions inventory, off-highway transportation sources emit as much PM as do highway sources [EPA, 1995d].) It thus is likely that the true ratio of ambient road-dust PM to ambient motor-vehicle-exhaust PM is close to the ratio of geologic PM to motor-vehicle PM reported in the CMB studies¹⁰.

Second, PM from motor-vehicle exhaust generally is smaller than PM from roads, and hence disperses further and stays in the atmosphere longer than does PM from roads. This will result in a greater fraction of motor-vehicle PM₁₀ or PM_{2.5} than road-dust PM₁₀ or PM_{2.5} being captured at the ambient monitors and hence included in the CMB studies, which in turn will tend to make the ratio of CMB road-dust share to CMB motor-vehicle share lower than the ratio of OEI road-dust emissions to OEI motor-vehicle emissions¹¹.

Work by Wiman et al. (1990), on particulate residence time, allows us to estimate the relative length of time that road dust and motor vehicle exhaust PM stays in the atmosphere. Wiman et al. (1990) present an equation that gives the estimated residence time for particulates of varying sizes in different sections of the atmosphere (Table 16-13). To use this equation, we first estimate the average diameter of road dust and exhaust PM. Several pieces of evidence help us to estimate the average diameter:

⁹Whittorf et al. (1994), estimated that, at a bust stop in Manhattan in 1993, road dust accounted for about 10% of total PM from all sources, and 15% of motor-vehicle-related PM. Thus, in this study, the ratio of road-dust to motor-vehicle PM was less than 1.0

¹⁰Watson et al. (1994b) define motor-vehicle exhaust as "those particles emitted directly from the tailpipe," (p. 32), and surmise that "most of the geologic material originated from paved roads" (p. 32). This implies that the primary-geologic: primary-motor-vehicle ratio from the CMB studies refers to substantially the same thing as the paved-roads: highway-vehicles ratio from the emissions inventory.

 $^{^{11}}$ Table 16-11 shows that the OEI ratio substantially exceeds the CMB ratio for any particle size class. Although the ratio of OEI road-dust to OEI motor-vehicle emissions of coarse PM₁₀ is closer to the ratio of CMB-estimated ambient geologic coarse PM₁₀ to ambient motor-vehicle coarse PM₁₀ than are the OEI ratios for PM₁₀ or PM_{2.5}, it still is not equal to the CMB ratio. Note, though, that in the CMB studies, very little coarse PM₁₀ was attributed to motor-vehicles anyway. Hence the ratio of geologic coarse PM₁₀ to motor-vehicle coarse PM₁₀ is very sensitive to small absolute changes in the amount of coarse motor-vehicle PM₁₀ predicted, and consequently might not be particularly meaningful.

- i.) First, there are estimates from three studies that use a large set of data collected in California in 1986.
 - Chow et al. (1994a) indicate that most PM2.5 road dust is between 0.5 and 2.5 μ m, with a mean of around 1.5 μ m, and that most PM10 road dust is around 6.0 μ m (see Table 16-14).
 - Walsh (1995) reports that combustion particles can range from 0.01 to 1.0 μ m, and that dusts generally are above 2.0 μ m. For diesel vehicles, 96.2% is less than 10 μ m, 92.3% is less than 2.5 μ m, and 91.8% is less than 1.0 μ m. For dust, the results are 52.3% (10 μ m), 10.7% (2.5 μ m), and 4.5% (1.0 μ m). The data are from emissions characterizations conducted in California in 1986.
 - Kao and Friedlander (1995) estimate that 11-30% of PM₁₀ dust is PM_{2.2}, using the same set of data that are used in Table 16-14 (Chow et al.).
- ii) Flagan (1993) has a graph showing that combustion fumes range from 0.01 to 1.0 μ m, and that mechanically generated particles range from 1.0 to 100 μ m.
- iii) Cahill and Wakabayashi (1993) state that "accumulation" mode particles, which are mainly combustion particles, range from 0.2 to $0.8~\mu m$.
- iv) In the Allegheny Mountain Tunnel in the Pierson and Brachaczek (1983) study, the mean mass of all vehicle aerosol was 0.15 μ m. This, though, includes the relatively large road-dust aerosol too.

Based on this evidence we assume the following average particulate sizes:

PM_{2.5} and PM₁₀ exhaust = $0.1 \mu m$ PM_{2.5} dust paved roads = $1.5 \mu m$ PM₁₀ dust paved roads = $6.0 \mu m$.

Table 16-12 presents the residence time for particulate sizes in two regions of the atmosphere, and it presents the ratio of residence time for a given PM size and residence time for $0.1~\mu m$ PM. Assuming emissions stay below 1.5 kilometers and allowing for uncertainty in mean size of the two classes (particularly road-dust), it appears that PM2.5 from exhaust stays in air roughly 5% to 20% longer than does PM2.5 from paved roads, and that PM10 from exhaust stays in the air 2 to 4 times longer than does PM10 from paved roads. This suggests that, in our comparison of the OEI dust:MV ratio with the CMB dust:MV ratio for PM2.5, we should assume that a factor of 1.05 to 1.2 of the ratio of the ratios is due to the different residence times. In the case of PM10, we assume that a factor of 2.0 to 4.0 of the ratio of the ratios is due to different residence times.

Third, as discussed above, it is likely that motor-vehicle exhaust emissions are underestimated in the emissions inventory. However, they probably are not underestimated by more than a factor of 1.5 to 2.0, which still leaves unaccounted for the bulk of the discrepancy between the CMB road-dust:motor-vehicle ratio and the OEI ratio.

Finally, emissions of dust from paved roads probably are overestimated in the emissions inventory. In the case of PM₁₀, differential dispersion of motor-vehicle versus road-dust PM, and the underestimation of motor-vehicle emissions, together can account for no more than a factor of about 3-8 out of the 10-fold difference between the OEI and CMB PM₁₀ road-dust/motor-vehicle ratios (Table 16-11). Regarding PM_{2.5}, differential dispersion and an underestimation of emissions can account for no more than a factor of 1.6 to 2.4 of the 30-fold difference between the PM_{2.5} ratios. The remaining differences between the PM₁₀ and PM_{2.5} ratios indicates that paved-road-dust emissions of PM₁₀ are overestimated by a factor of 1.25 to 3.33, and that PM_{2.5} emissions are overestimated by a factor of 12.4 to 18.9¹².

We determine the factor $(\alpha_{pm_{10}})$ by which PM₁₀ road-dust is overestimated with the following equation:

$$\frac{\left(OEI\ road-dust\ PM_{10}\ share\right)\times\alpha_{pm_{10}}}{\left(OEI\ motor\ vehicle\ exhaust\ PM_{10}\ share\right)\times\beta_{pm_{10}}\times\chi_{pm_{10}}}{CMB\ road-dust\ PM_{10}\ share} = \frac{17.1}{1.7} = 9.9,$$

when:
$$\alpha_{pm_{10}} = \beta_{pm_{10}} = \chi_{pm_{10}} = 1$$

where:

 α_{pm10} = road-dust PM10 emission-correction factor

 β_{pm10} = exhaust PM10 emission-correction factor

 $\chi_{pm10}\text{=}$ differential dispersion factor between exhaust and road-dust PM_{10}

We expect this ratio to be equal to one, not 9.9. To make the equation equal to one, we make corrections for exhaust PM₁₀ and for the differential dispersion of exhaust versus paved-road dust, and then solve for the paved-road-dust correction factor so that the overall ratio is one. We do analogous calculations for PM_{2.5}, although the ratio of ratios now equals 29.8 (not 9.9 as for PM₁₀) (see Table 16-11):

32

¹²This refers to the unrevised PM_{2.5} OEI that we use. The revised emissions inventory (Table 16-8) apparently is overestimated still by a factor of roughly 7.4 to 11.3.

$$\begin{split} &\alpha_{pm_{10}}=1.24,\ when\ \beta_{pm_{10}}=2,\ \chi_{pm_{10}}=4\\ &\alpha_{pm_{10}}=3.33,\ when\ \beta_{pm_{10}}=1.5,\ \chi_{pm_{10}}=2\\ &\alpha_{pm_{2.5}}=12.42,\ when\ \beta_{pm_{2.5}}=2,\ \chi_{pm_{2.5}}=1.2\\ &\alpha_{pm_{2.5}}=18.92,\ when\ \beta_{pm_{2.5}}=1.5,\ \chi_{pm_{2.5}}=1.05. \end{split}$$

II. <u>Measurements of total roadway emissions in the east and west of the United States.</u> As discussed above, the AP-42 emission factor equation is derived from measurements of total roadway emissions (exhaust + tirewear + brakewear + road dust) in the Midwest, and the silt-loading values that are input to the equation to generate the emissions inventory are based on measurements done mainly in the Midwest, many years ago. We believe that silt loadings, and hence roadway emissions, are much lower in the major urban areas of the U.S. today than they were in the Midwest many years ago. As indirect evidence of this, we have compiled the results of several studies of total roadway emissions done outside of the areas where the original AP-42 field studies were done (Table 16-13). In every case, AP-42 substantially over-predicts total roadway emissions.

We hasten to point out this evidence is only indirect. The tests in Table 16-13 measure, and the AP-42 equation predicts, all emissions across a roadway: exhaust emissions, tirewear emissions, brakewear emissions, and road-dust emissions. If silt loadings on the roads tested in Table 16-13 are much lower than the values assumed in the application of AP-42, then AP-42 will over predict roadway emissions. But this is not the only reason that AP-42 might over predict total roadway emissions: it is possible that the fleet-average exhaust emission rate implicit¹³ in the AP-42 equation is much higher than the actual exhaust emissions in the studies of Table 16-13. However, there is no evidence that this is the case. We believe that it is most likely that actual silt loadings along the roadways tested in Table 16-13 were much lower than the values assumed in the application of AP-42.

Why might the silt loadings on the roads of Table 16-13 be so much lower than the levels assumed in AP-42? There are several reasons:

i). *Tunnel studies:* Pierson and Brachaczek (1983) emphasize that one would expect to find very little soil at the *exit* of a tunnel, because, as they put it, "the

¹³Note that it is the PART5 model, not AP-42, that probably underestimates exhaust PM emissions in general. The PART5 model explicitly predicts exhaust PM emissions for individual vehicle classes, on the basis mainly of dynamometer tests. By contrast, the AP-42 roadway-emissions equation is a statistical representation of all emissions (exhaust + road dust +...) from the entire in-use fleet, on the road. Recall that road-dust emissions are calculated by subtracting PART5-estimated exhaust and tire-wear emissions from AP-42 estimated total roadway emissions.

entrainment process...had been turned off some 1623 meters up the road" (p. 17). In other words, the nearest source of soil was a mile away. They argue that the road-dust percentage should be much higher "in the open."

- ii). Seattle, Washington study: Seattle has a relatively high number of days of precipitation, although the annual precipitation is roughly average. A lot of days of light rain might tend to suppress emissions of road dust: Nicholson et al. (1989) found that "the presence of a small amount of moisture seemed to greatly inhibit resuspension" (p. 1425). (The EPA recently has revised its calculation of emissions of dust from paved roads to account for the effects of rain.)
 - iii). California freeway studies: According to Cahill et al. (1994):

California freeways generally are not sanded or salted..The high freeway speeds and presence of trucks sweep freeways with hurricane-velocity winds several times each minute. The roadway itself is slightly greasy to the touch, with very low surface silt loading. Thus, the only re-suspended soils can come from the roadways' margins, which usually are paved well away from traffic flow. The major sources are located well away from the traffic lanes, allowing them to stabilize, often with vegetation...Other factors my be operating as well, e.g., less rapid deterioration of the exhaust system..(p. 24).

Summary. The results summarized in Table 16-13 suggest that AP-42 substantially overestimates emissions of PM₁₀ and PM_{2.5}, and is thus consistent with the inferences drawn from comparing CMB source apportioning with emission-inventory ratios. However, the results in Table 16-13 are not particularly helpful in quantifying the size of the overestimate since they consider total emissions rather than just paved-road-dust emissions.

III. In the equation for dust emissions from paved roads (equation (D2) and (D3) above), the size-class multiplier "K" is 4.6 for PM₁₀, and 2.1 for PM_{2.5}, for paved roads (EPA, 1995a), which means that PM_{2.5} is assumed to be 45% of PM₁₀. However, in the actual emissions inventory that we use, PM_{2.5} is 42% of PM₁₀ from road dust. We do not know why the percentage in the actual inventory differs slightly the percentage in the emission-factor equation. The evidence, summarized below, indicates that this percentage is too high.

- i). Cowherd, (1995b), who is re-evaluating the road-dust emissions inventory for EPA, suggests that PM_{2.5} from road dust is about 10% of PM₁₀ from road dust. Wilson (1995) of EPA suggests an even lower percentage.
- ii). The EPA contractor who prepares the emission inventory recently has revised the PM2.5/PM10 ratio for paved-road dust to 25% (Barnard, 1996).
- iii). The data of Table 16-10 indicate that PM_{2.5} from geologic material is on the order of 10% of PM₁₀ from geologic material. Source apportionment studies in Reno, Sparks, and Verdi, Nevada (Watson et al., 1988a) indicate a somewhat higher PM_{2.5}/PM₁₀ ratio of about 20% to 40%.

Note that this is the $PM_{2.5}/PM_{10}$ ratio for geologic PM in general. However, the $PM_{2.5}/PM_{10}$ ratio for road-dust specifically probably is similar, because most of the

other components of the general "geologic" category -- dust from wind erosion, unpaved roads, and agriculture -- apparently have PM2.5/PM10 ratios similar to that for paved roads (Table 16-14), and because paved-road dust probably is a substantial fraction of general geological PM, especially in cities.

- iii). Chow et al. (1994a) measured the size distribution of several sources of fugitive dust particles, and found that in Fresno, PM_{2.5} from paved road is 19% of PM₁₀ from paved roads (Table 16-14).
- iv). Kao and Friedlander (1995) cite an estimate, in a 1989 report (*Determination of Particle Size Distribution and Chemical Composition of Particulate Matter from Selected Sources in California*) for the California Air Resources Board, that 11% to 30% of geologic PM10 is in the PM2.2 size class. 14 v). There are data that indicate that road-dust PM2.5 is 35% or more of road-dust PM10, but they are weak or incomplete. Partial data from a study by Pierson and Brachaczek (1983; summarized in Table 16-2 here) suggest that road-dust PM2.5 is about 35% of road-dust PM10. Pierson and Brachaczek (1983) identified the source of all motor-vehicle related PM emissions generated at the exit of a tunnel on an expressway. According to the authors, all of the elements Ca, Sc, Ti, V, Si, and K, and about half of the elements Li, Mg, Al, Cr, Fe, and Zr in the airborne particles were from local soils. The authors give the median size of Ca, Fe, Mg, Ti, and Cr particles. Of these, Ca has the largest median size (4 µm). A graph in their paper shows that 95% of Ca was PM10, and 35% was PM2.5. In other words, PM2.5 was a little over 35% of the PM10 of one of the largest soil particles.

Also, the EPA's *Air Emissions Species Manual* (1990) reports that PM_{2.5} from paved roads is 57% of PM₁₀ from paved roads. That estimate, however, is merely an "extrapolation" of data from a 1984 Canadian study.

The evidence of Tables 16-10, and 16-14, and the opinion of the experts cited above, suggests that PM_{2.5} from paved roads 10% to 30% of PM₁₀ from paved roads. As we noted above, the actual inventory shows that PM_{2.5} is 42% of PM₁₀. Since we are adjusting the actual emissions inventory, we apply our adjustment to the 42%, not the 45%, baseline. If this is so, then road-dust PM_{2.5} in the emission inventory that we use is overestimated by about 1.4X to 4.2X, where X is the factor by which PM₁₀ is overestimated.

Our conclusions

CMB source-apportionment studies of ambient PM₁₀, measurements of PM emissions from road traffic, and analyses of the size distribution of dust particles from paved roads, indicate that:

35

 $^{^{14}}$ Kao and Friedlander (1995) used the term "crustal sources" which we take to mean geologic sources.

- 1) AP-42 and the emissions inventory overestimate emissions of PM₁₀ from paved roads, perhaps by several-fold (probably because on average silt loadings are several times lower than is assumed in the application of equation D2); and
- 2) the published AP-42 coefficient (not the recently revised coefficient; see Table 16-8) overestimates the ratio of $PM_{2.5}$ to PM_{10} from paved roads, by a factor of 1.4 to 4.2.

Given all of this, we assume, first, that PM₁₀ road dust is overestimated by a factor of 1.25 to 3.33. Then, we assume that the ratio of PM_{2.5} to PM₁₀ is overestimated by a factor of 1.4 to 4.2, so that PM_{2.5} emissions are overestimated by (1.25 x 1.4 =) 1.75 to (3.33 x 4.2 =) 14.0¹⁵. The adjustment factors we give in Table 16-1 are the inverse of these factors.

16.2.5 Estimates of PM dust from unpaved roads (AP-42, Volume 1)

PM10. We have not come across any evidence that the EPA's (1995d) estimate of PM10 dust emissions from unpaved roads is seriously in error, and as a result, we do not make any corrections to the PM10 emissions inventory for unpaved roads.

PM2.5. In AP-42, and in the actual emissions inventory that we use (EPA, 1995d), emissions of PM2.5 from unpaved roads are assumed to be 0.264 of emissions of PM10 from unpaved roads. As discussed above, the PM2.5/PM10 ratio for paved roads appears to be 0.10 to 0.30. Now, the data of 16-14 indicate that the PM2.5/PM10 ratio should be slightly lower for unpaved than for paved roads: 0.16 for unpaved roads, versus 0.19 for paved roads. This seems reasonable. Considering all this, we assume that the PM2.5/PM10 ratio for unpaved roads should be 0.08 to 0.25. Thus, to correct the EPA's assumed ratio of 0.264 to our range of 0.08 to 0.25, we must multiply by 0.30 to 0.95. These are the correction factors of Table 16-1.

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 $^{^{15}}$ This range for PM_{2.5} is slightly less than the range of 2.4 to 18.9 suggested by the comparison of the OEI with the CMB studies. However, we believe that the 1.75 to 14.0 range is more accurate, because the evidence regarding the error in the PM_{2.5}/PM₁₀ ratio is solid.

16.2.6 Estimates of PM emissions from construction, including road construction (AP-42, Volume 1)

In our official emissions inventory, fugitive dust from construction is one of the largest sources of anthropogenic PM₁₀ (Barnard, 1996) (Table 16-8). However, as we explain here, we suspect that construction emissions are overestimated. It is important to estimate construction emissions accurately not only because road construction emissions (which is attributed to motor-vehicle use) are nontrivial in a few places (such as Los Angeles) (EPA, 1995d), but because construction emissions appear in equations 6 and 7.

According to the EPA (1995a), the quantity of dust emissions from construction operations is proportional to the area of land being worked and to the level of construction activity. Thus, in the EPA's recent *Compilation of Air Pollutant Emission Factors* (1995a), emissions from road construction are calculated with the following generic emission-factor equation:

Total Suspended Particulate (TSP) emissions = 1.2 tons/acre/month of activity.

This obviously is a very generic, highly aggregated emission factor, useful, as the EPA (1995a) properly cautions, only for developing estimates of overall emissions from construction scattered throughout a geographical area¹⁶.

But even applied broadly, this emission factor has several weaknesses. First, it is based on only one set of field studies, published in 1974, of concentrations of total suspended particulate (TSP) surrounding apartment and shopping center construction projects (EPA, 1995a). We speculate that these old studies were of especially dusty construction sites, and hence overestimate dust emissions from most construction sites. We emphasize, though, that this is speculation

Second, in the original field studies there were no measurements of PM₁₀ or smaller, and consequently the published emission factor is for TSP only. However, the EPA's (1995d) official emission inventory assumes that PM₁₀ from construction is 22% of TSP from construction (Solomon, 1995), which seems reasonable to us.

Third, the emission factor assumes that construction activity occurs 30 days per month (EPA, 1995a), and the EPA's (1995d) official PM emissions inventory apparently accepts this assumption (Solomon, 1995). We believe that 21 days per month is more reasonable.

Finally, we note that the chemical-mass balance analyses of the chemical composition of ambient particulates typically find very little PM enhanced in lime or calcium (Table 16-9).

With these considerations, we believe that true PM₁₀ emissions from construction are 10% to 50% of the official emission-inventory estimates.

37

¹⁶The EPA (1995a) recommends that emissions from a particular construction site be analyzed with respect to the specific activities at the site: drilling, blasting, loading, bulldozing, etc.

In the official PM emission inventory that we use, PM2.5 from construction is estimated to be only 2% of PM10 from construction. However, this 2% factor apparently is off by an order of magnitude, because recently the EPA contractor who does the official emissions inventory revised the PM2.5 fraction to 20% (Barnard, 1996). Thus, to correct PM2.5 emissions from construction, we first multiply by 0.1 to 0.5 (the correction factors for PM10, above), then multiply by 10 (to correct 2% to 20%), giving an overall correction factor of 1.0 to 5.0.

16.2.7 Summary of correction factors

Our investigation of the uncertainty of some of the emission factors related to motor-vehicle use indicated that MOBILE5A underestimates emissions of VOCs, CO, and perhaps NO_X from light-duty gasoline vehicles; that PART5 underestimates PM emissions from heavy-duty diesel vehicles; and that AP-42 overestimates emissions of road dust and dust from road construction (but underestimates PM_{2.5} from construction). Accordingly, we have estimated adjustment factors, to correct for the likely extent of over- or under-estimation. These adjustment factors -- $EC_{p',i}$ in equations 6 and 7 -- are summarized in Table 16-1.

16.3 THE DISPERSION OF EMISSIONS FROM SOURCE TO AMBIENT AIR-QUALITY MONITOR

16.3.1 Conceptual approach to air-quality modeling

Recall that in order to estimate the motor-vehicle contribution to ambient air pollution, we must estimate a normalized "dispersion" term ($DN_{p',i,c}$ and $DN_{p',i,o}$ in equations 6 and 7), which is the fraction of emissions from source i that is captured by the ambient-air quality monitors, normalized to the same fraction for light-duty-vehicle tailpipe emissions of fine PM. In this section, we develop a simple Gaussian dispersion model and use it to estimate the normalized dispersion terms. We will estimate one set of DN terms for urban air pollution (for the analysis of health effects [Report #11], and the analysis of visibility [Report #13]), and another for air pollution in agricultural areas (for the analysis of crop damages [Report #12]).

As discussed above, we assume that in each county, the pollution measured at the receptor R (the air-quality monitor) is determined by emissions within the county, and emissions from all other counties in the same AQCR. Figure 16-1 depicts emissions sources and receptors in counties in an AQCR. In this figure, there is one receptor (R), or air-quality monitor, near the center of each county. Motor-vehicle (MV) and other (O) emission sources are scattered around the monitor, as indicated. The indicated motor-vehicle sources are the effective emissions center (or band) of the aggregation of actual motor-vehicle emissions throughout a portion of the county. The other emission sources are large point sources, such as power plants or industries, or the center or band of area sources such as farms.

Within-county emissions. We will model emissions from 13 categories of emissions sources (MV + 12 "O" ["other"] categories) within every county. Specifically, we will make assumptions about the location and other characteristics of emissions sources, and then use a Gaussian plume model to calculate the contribution to concentration. As indicated in Figure 16-1, we assume that, on average, motor-vehicle sources are closer to the monitors (receptors) than are other sources. This model is developed later in this section.

Out-of-county emissions. Out-of-county emissions are trickier. If the prevailing wind blows diagonally across the AQCR as shown in Figure 16-1, from County 1 to County 8, then emissions from County 1 blow first into County 2, then into County 3, then into Counties 4 and 6, then into Counties 5 and 7, then into County 8, and finally out of the AQCR and into the next AQCR. Emissions from County 2 blow first into County 3, then into Counties 4 and 6, and so on. Generally, emissions from each upwind county blow into the downwind counties. The emission plume spreads out and becomes ever more dilute as it moves further from the source through the more remote counties.

In the scenario just described, the receptor R in County 8 receives emissions from the upwind sources within the county plus emissions from the 7 upwind counties; the receptors in Counties 5 and 7 receive emissions from the upwind sources within the counties plus emissions from the 5 upwind sources; and so on, up to the receptor in County 1, which receives emissions from the upwind sources within the county plus any emissions that blow in from the adjacent AQCR region. Therefore, in general, the contribution of out-of-county emissions is determined by the emissions from the upwind counties.

Ideally, we would model the specific upwind emission sources that affect air quality in each and every county in the U.S. This, however, is beyond our scope. Instead, to model the effect of out-of-county emissions, we make three simplifying assumptions. First, we group counties into AQCRs, and assume that the AQCRs are isolated pollution-mixing basins. With this assumption, we can close the bounds of our analysis around the AQCR, and, in terms of Figure 16-1, ignore pollution that blows out of County 8 into the AQCR downwind, or into County 1 from the AQCR upwind.

Second, we assume that the average contribution of out-of-county emissions to receptors throughout the AQCR can be represented by the contribution to a receptor at the center of the AQCR. In terms of Figure 16-1, we assume that contribution of upstream emissions to the receptor in County 3 is close to the average of the contributions to the receptors in Counties 1 and 8, or the average of the contributions to the receptors in Counties 2 and 4, 5, 6, and 7. With this assumption, we can model air quality in each county as if the county were at the center of the AQCR.

Third, we assume that the all of the out-of-county sources are located at single distance directly upwind of the central receptor in the county of interest. If in Figure 16-1 we are interested in air quality at the receptor R in County 3, then to estimate the out-of-county contribution, we collapse all of the sources in the upwind Counties 1 and 2 into a single effective source, at some distance outside of County 3. (If within in AQCR,

some sources tend to be located further from the center than others, then our assumption will be incorrect.)

With these assumptions, we model every county as a circle at the center of a circular AQCR, as shown in Figure 16-2. At the very center is the air-quality monitor, R. Upwind of R, within the circle of the county, are the motor-vehicle (MV) and other (O) emission sources, located at different distances from R according. All of the outside emission sources are located at a radial distance r_0 . We discuss our assumptions for r and r_0 later.

16.3.2 The Gaussian model

Ours is a Gaussian model of a pollutant plume from an emissions source to a receptor. It relates the pollutant concentration at the receptor to the strength of the emissions source, the distance from the source to the receptor, the wind speed and angle, the stability of the atmosphere, and other factors. Gaussian dispersion models are widely used in air quality modeling, especially for regulatory purposes, because they produce results that agree reasonably well with experimental data, are easy to manipulate mathematically, and are well grounded theoretically (Hanna et al., 1982). The EPA's "Industrial Source complex Dispersion Models," used for regulatory purposes to model emissions from elevated point sources, is built upon the Gaussian plume equation (EPA, 1995f).

The Gaussian plume dispersion equation. With a variety of restrictive assumptions, one can derive the following Gaussian plume dispersion model, in which pollutant concentration at the point (0, 0, z) in the Cartesian coordinate system is a function of: the rate of emissions from a source located at (x, y, h_s) ; the velocity of the wind, which by convention is assumed to be oriented parallel to the x axis; the magnitude of x, y, and h (height); the height of the atmospheric region in which pollutants can mix (called the "mixing layer); the deposition or settling of pollutants; turbulent diffusion, and other factors (EPA, 1995f; Horowitz, 1982; Hanna et al., 1982; Ermak, 1977):

¹⁷The stability of the atmosphere, as pertains to Gaussian dispersion modeling, is described by the behavior of a parcel of air displaced vertically and adiabatically from its original position. In a stable atmosphere, the parcel will return to its original point. In an unstable atmosphere, the parcel will accelerate away from its original point. In a neutral atmosphere, the parcel will not be accelerated. The atmosphere is unstable close to the surface on a sunny day, neutral on a windy and cloudy day or night, and stable near the surface at night or at any time in an elevated inversion layer (Hanna et al., 1982).

Pasquill (1974) defines six categories of stability: A - extremely unstable, B-moderately unstable, C-slightly unstable, D- neutral, E-slightly stable, and F-moderately stable. Values for some of the dispersion parameters, such as σ_y and σ_z , have been developed for the different Pasquill stability classes, and as a result the Pasquill stability classes are widely used. Of course, there are other measures of stability. For example, the Richardson number and the Monin-Obukhov length are direct measures of stability which account for the effects of both mechanical mixing and buoyancy forces (Hanna et al., 1982). Hanna et al (1982) show Golder's analysis of the relationship between Pasquill's stability categories and the Monin-Obukhov length.

if $h < z_i$ and $\sigma_z < a \cdot z_i$, then we have the following Gaussian dispersion

$$C(x,y,z,t) = \frac{E}{2\pi \cdot w \cdot \sigma_{y} \cdot \sigma_{z}} \cdot e^{\left(-0.5\left(\frac{y}{\sigma_{y}}\right)^{2}\right)} \cdot D1 \cdot \left(e^{\left(-0.5\left(\frac{z_{r}-h}{\sigma_{z}}\right)^{2}\right)} + e^{\left(-0.5\left(\frac{z_{r}+h}{\sigma_{z}}\right)^{2}\right)} + S1 + D2\right) \cdot D3$$
(8a)

if $h < z_i$ and $\sigma_z > a \cdot z_i$, then we assume the concentration is homogeneous vertically

$$C(x, y, z, t) = \frac{E}{2\pi \cdot w \cdot \sigma_{y} \cdot \sigma_{z}} \cdot e^{\left(-0.5\left(\frac{y}{\sigma_{y}}\right)^{2}\right)} \cdot D1 \cdot \left(\frac{\sigma_{z} \cdot \sqrt{2\pi}}{z_{i}} + D2\right) \cdot D3$$
 (8b)

if $h > z_i$ then the pollution stays above the mixed layer , and at the surface we have :

$$C(x,y,z,t) = 0 (8c)$$

where:

h = the "effective" height of the emissions source above the ground (meters)

 z_i = the mixing height of the atmosphere (meters)

a = constant expressing the maximum σ_{z_i} as a fraction of mixing height z_i

C(x,y,z,t) = the concentration of pollution, due to emissions from source i, at time t and coordinates x, y, and z (g/m³)

E = the mass of pollutant emitted per unit time from source i (continuously) (g/sec)

x = the distance from the emissions source i to the receptor r, along the direction of the wind (meters)

y = the distance from the source to the receptor, perpendicular to the direction of the wind (meters)

 z_r = the height of the receptor above the ground (meters) (the receptor is taken to be located at the point (0, 0, z) in the Cartesian coordinate system

w = the mean wind velocity at height h, taken to be along the \mathbf{x} axis $(m/s)^{18}$

 $^{^{18}}$ The EPA (1995f) uses the wind speed at the stack height h_s , rather than at the effective height h_s . However, Hanna et al. (1982) state that in the Gaussian equation, the wind speed is the average throughout the plume depth, but that in practice the wind speed at h_s (not h_s) is used.

- σ_y = the horizontal diffusion parameter: the standard deviation of the distribution of the concentration C in the direction perpendicular to the wind; this is a function of time, where time is given by \mathbf{x}/w_X -- the time it takes a particle to be transported by the wind from the source to the \mathbf{x} -axis coordinate of the receptor (meters)
- σ_Z = the vertical diffusion parameter: the standard deviation of the distribution of C in the vertical direction; this also is a function of time, where time is given by \mathbf{x}/\mathbf{w}_X (meters)
- S1 = term to account for multiple reflections of the plume off of the ground and the inversion layer (unitless)
- D1 = term to account for settling of particles (unitless)
- D2 = term to account for deposition of particles and reactive gases (unitless)
- D3 = term to account for removal of pollutant by chemical reaction (unitless)

This model assumes that the earth's surface is a plane, the atmosphere is homogenous, vertical wind speed is zero, particles are perfectly reflected from the surface and the underside of the inversion layer, emission sources are constant over time, and the effects of turbulent diffusion in the direction of the wind are negligible in comparison to the effects of transport by the wind (Horowitz, 1982). To the extent that these assumptions are incorrect, the model will mis-estimate pollutant concentrations. Gaussian dispersion models can mis-estimate absolute concentrations by an order of magnitude or more.

However, we will use the model not to estimate absolute pollutant concentration, but rather to compare the dispersion of motor vehicle exhaust emissions relative to the dispersion from other sources. Our purpose is to estimate the *fraction* of total pollution that is due motor vehicles, not the absolute amount of total pollution. We expect that some of the errors involved in estimating absolute concentrations in effect cancel out when one is estimates the contribution of one source *relative* to the contribution of another. It is with this expectation that we proceed to use the Gaussian model to estimate DNp', i, c and DNp', i, o in equations 6 and 7 and above.

To get a rough approximation of the normalized dispersion term $(DN_{p',i,C})$ and $DN_{p',i,O}$ in equations 6 and 7), we can divide C(x,y,z,t) for emissions source i by C(x,y,z,t) for motor-vehicle exhaust emissions (actually, light-duty motor-vehicle exhaust emissions):

$$DN_{P',i} = \frac{c(x,y,z,t)_{P',i}}{c(x,y,z,t)_{P',m}} = \frac{e^{\left(-0.5\left(\frac{yi}{\sigma_{yi}}\right)^{2}\right)} \cdot D1_{i} \cdot \left[e^{\left(-0.5\left(\frac{z_{r}-h_{i}}{\sigma_{zi}}\right)^{2}\right)} + e^{\left(-0.5\left(\frac{z_{r}+h_{i}}{\sigma_{zi}}\right)^{2}\right)} + S1_{i} + D2_{i}\right] \cdot D3_{i}}{e^{\left(-0.5\left(\frac{y_{m}}{\sigma_{ym}}\right)^{2}\right)} \cdot D1_{m}\left[e^{\left(-0.5\left(\frac{z_{r}-h_{m}}{\sigma_{zm}}\right)^{2}\right)} + e^{\left(-0.5\left(\frac{z_{r}+h_{m}}{\sigma_{zm}}\right)^{2}\right)} + S1_{m} + D2_{m}\right] \cdot D3_{m}}$$

$$(9)$$

where the subscript "m" refers to light-duty motor-vehicle exhaust pipe emissions, and "i" refers to any other source (including road dust). (The $\mathrm{DNp'}$, i for equation 8b is calculated analogously.) We assume that the emission rate E is the same for all sources, and hence cancel it out in the equation, because we are interested in the relative contribution to concentration *per unit of emissions*.

We now turn to the problem of estimating the parameters in equations 8a and 8b. Because most of the parameter values (e.g., effective height of emissions, h) vary from emissions source to emissions source, we will have to estimate different values for different emission sources. (That is, in general, $DN_{p'}$ for, say, trains, is different than $DN_{p'}$ for say power plants.) To keep our task manageable, we will estimate different sets of parameter values, and hence different $DN_{p',i}$, for 13 different categories of emissions, shown in Table 16-15. We chose these 13 categories as a compromise between the demand for accuracy, which calls for more categories, and the demand for simplicity, which calls for fewer categories.

Where there is considerable uncertainty or site variability for a parameter, we assume low and high values, where "low" results in a low dollar cost attributable to motor-vehicle air pollution, and high to a high dollar cost. Note that a lower dollar cost results from a higher value for $\mathrm{DN}_{p',i}$, for non-motor-vehicle sources, and vice-versa because a higher value for $\mathrm{DN}_{p',i}$ means that the non-motor-vehicle sources are responsible for a larger share of the ambient air pollution. Hence, our low case, which is a low-cost case, corresponds to the high for $\mathrm{DN}_{p',i}$ (for non-motor-vehicle sources), and vice versa. We experiment with different combinations of parameter values to determine which parameter bound gives the low-cost result, and which gives the high-cost result.

The non-motor vehicle sources, for which the high $DN_{p',i}$ result in low costs for motor vehicles, are all sources except LDVs, HDVs, paved roads, and unpaved roads. For the motor-vehicle related sources (LDVs, HDVs, paved roads, and unpaved roads), the relationship between the low and high DNi values and the low and high cost results is not uniform, mainly because the parameter values for some of these must be estimated relative to the parameter values for LDVs.

Distance from source to receptor. In the formulation above, the vector from the source to the receptor is decomposed into a vector along the wind direction (x), and a vector perpendicular to the wind direction (y). Rather than estimate the length of these decomposed vectors (x and y) directly, it is more intuitive to estimate directly the distance from the source to the receptor (r), and the angle θ between the wind vector and the source-receptor vector, and to calculate x and y from these directly estimated quantities:

$$x = r \cdot \cos(\theta)$$

$$y = r \cdot \sin(\theta)$$

where:

x, y are as defined above

r = distance from the source to the receptor (meters) (discussed below)

 θ = the angle between the wind vector and the source-receptor vector (degrees) (discussed below)

The distance from the source to the monitor (r). We estimate two sets of source-receptor distances: one to agricultural monitors (for the analysis of crop damages, in Report #12), and another to urban monitors (for the analysis of the human health and visibility costs of air pollution). In the case of human health and

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¹⁹There actually is a complication here: in nearly all of the 13 emission-source categories, at least some emissions are related to motor-vehicle use. For example, some of the VOC emissions in the "solvent" category arise from painting cars. Ideally, we would separate all of the motor-vehicle related emissions sources, and treat the low and the high for them differently than from the low and the high for non-motor-vehicle-related sources. (High costs for motor vehicles result from high DNi on all sources related to motor-vehicle use, but low DNi on all sources not related to motor-vehicle use.) However, to keep our analysis manageable, we have not done this. For the purpose of estimating low and high motor-vehicle costs, we treat all emission sources other than motor vehicles, paved roads, and unpaved roads as non-motor-vehicle sources.

visibility we refer to urban monitors because the bulk of health and visibility damages due to air pollution occur in urban and suburban areas. (However, see the qualifications in Reports 11 [health effects] and 13 [visibility].)

As mentioned in sections 16.1.2 and 16.3.1, we assume that in each County C, air quality measured at a monitor is a function of: 1) emissions from within County C; and 2) emissions outside of County C but in the same AQCR as County C. Let us consider each of these in turn.

- 1). Distance to emission sources within the county. As explained in section 16.3.1 and illustrated in Figure 16-2, we estimate distances from the monitor to 13 emission source categories *within* the county in question. Our assumptions regarding these 13 distances are shown in Table 16-15, and explained in the notes thereto. Generally, we assume that motor vehicles are relatively close to urban monitors, but relatively far from agricultural monitors.
- 2). Distance to emission sources outside of the county, but in the same AQCR. As explained in section 16.3.1 and illustrated in Figure 16-2, we assume that all of the out-of-county sources are located at single distance directly upwind of the central receptor in the county of interest. In Figure 16-2, this is the distance r_0 . It is reasonable to expect that this distance is a function of the size of the AQCR, and the average size of counties within an AQCR: the bigger r_r (the radius of the AQCR, if it the AQCR were a circle) and r_c (the radius of the average-size county within the AQCR, if the county were a circle), the bigger r_0 , the parameter of interest. Thus, to estimate r_0 , we first should calculate r_r and r_c .

In Table 16-16, we present statistics on r_r and r_c for all 241 AQCRs in the U.S. Because there is a considerable range in the size of AQCRs and the counties within them, we divided the universe of AQCRs into small (less than 11,000 mi²; 154 AQCRs) and large (greater than or equal to 11,000 mi²; 87 AQCRs). We thus estimate two r_0 values: one for small AQCRs, and another for large AQCRs. This is slightly more precise than estimating a single r_0 for all AQCRs²⁰.

Given the radius of the average county (the average county size being equal to the area of the AQCR divided by the number of counties in the AQCR), and the radius of the AQCR, we can calculate r_0 , the radius to the outside-of-county sources, as:

$$r_o = r_c + a \cdot (r_r - r_c)^b$$

²⁰⁻

²⁰It would be most accurate to calculate a separate r_O for each AQCR, and then calculate a separate set of normalized dispersion factors (DN_i) for *each* AQCR, rather than one set for small AQCRs and another for large AQCRs. However, to do this, we would have to add the entire air-quality model, which we developed in spreadsheet program, to the separate SAS program that manipulates the huge air-quality data base and applies the dose-response functions to calculated air quality in each county. Rather than move the entire air quality model from the spreadsheet to the SAS program, we calculate the DN_i in the spreadsheet program, and then transfer the results to the SAS air-quality program. To keep the amount transferred manageable, we calculate the DN_i for only two AQCR size classes, small and large.

What should be the value of the coefficients a and b? If concentration were a linear function of distance, and sources were distributed uniformly throughout the AQCR, it would be reasonable to assume that in effect all outside-of-county sources were located halfway between the edge of the county and the edge of the AQCR (a = 0.5, b = 1.0). However, concentration in fact is a nonlinear function of distance, such that closer sources contribute disproportionately more to concentration than do further sources. This suggests that effective distance should be closer to the county edge than the AQCR edge. We assume that a = 1.0, and b = 0.93, so that we have:

$$r_o = r_c + (r_r - r_c)^{0.92}$$

As shown in Table 16-16, we estimate r_0 = 120 km for large AQCRs, and 56 km for small AQCRs.

Note that the distance from source to receptor is the only parameter that has different values in the crop-damage analysis than in the health and visibility analyses.

The angle between the wind vector and the source-receptor vector (θ). We assume that all sources, including motor vehicles, are located randomly -- i.e., with no discernible pattern -- with respect to one another, the wind vector and the receptor. Put another way, we assume either a homogeneous density of sources throughout an area, or more generally any density distribution that does not make θ differ from one source to another. With this assumption, on average the angle between the wind vector (which by convention is parallel with the X axis) and the source-receptor vector will be the same for all source classes. For analytical convenience, we assume a value of 0° for all sources; that is, we analyze the case in which all emission are directly upwind of the receptor.

It turns out that the angle between the wind vector (which runs along the x-axis) and the vector from the monitor/receptor (at the origin) to the source is a more powerful determinant of DN_i then is the absolute distance r from the source to the monitor. In general, as this angle increases for any source i, the contribution of source i to the concentration at the monitor (receptor) decreases, because the emissions from i must move an increasing distance sideways in a decreasing amount of time in order to reach the monitor before being blown downwind of it. In the extreme, if this angle is 90° or greater, source i will be downwind of the monitor, and will contribute nothing to the concentration measured at the monitor.

Thus, if the wind/source-receptor angle (θ) for any source i is greater than the angle for motor vehicles, then source i is located on the **x**-axis (windward) side of the line from the monitor to the motor-vehicle source, and is more nearly parallel with the wind. This will result in a relatively high DN_i, even when source i is much further from the monitor that is the motor-vehicle source. Conversely, if the wind/source-receptor angle (θ) for any source i is less than the angle for motor vehicles, DN_i will be relatively

small. We have assumed that on average, source i and the motor-vehicle source make the same angle with the wind vector.

Note too that the direction of the wind turns with height, according to the "Ekman spiral" (Hanna et al., 1982). However, under the assumption that the source-receptor vector is random with respect to the wind vector, this altitude effect does not affect our assumption that θ is equal to 0° .

Effective source height (h). The Gaussian equation represents the diffusion of a pollutant plume that is released with no initial net vertical velocity. That is, in the Gaussian equation itself, there is no term that represents an initial vertical velocity of the pollutant plume. Thus, strictly speaking, the equation should applied at the point of zero net vertical velocity. In general, this point will *not* be the "mouth" of the exhaust stack at the physical height of the stack h_S, because at h_S, at the mouth of the stack, the exhaust gases are rising, perhaps rapidly, on account of their high temperature and initial exit momentum from the stack.

The relevant height in the Gaussian model, then, is some height above stack height h_S -- the "effective" source height h, at which the pollutant plume has stopped or nearly stopped rising on account of its initial buoyancy and momentum. This "effective" source height h therefore is given by:

$$h = h_S + \Delta h$$

where:

h = the effective source height in the Gaussian equation (meters)

h_S = the height of the actual stack or exhaust at the actual point of release (meters)

 Δh = the rise in the plume due to the pollutants being emitted from the stack at high velocity or high temperature (meters)

Physical stack height (h_S). Our assumptions are shown in Table 16-15, and explained in the notes thereto. Note that we have, and use, data on the actual stack heights of major point sources in five categories: i) fuel combustion, electric utilities; ii) fuel combustion, industrial; iii) fuel combustion, other (mainly residential wood combustion); iv) chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes; and v) solvent utilization, storage and transport, and waste disposal and recycling (Table 16-17). The calculated results for DN_i are somewhat sensitive to plausible variations in the assumed value of h_S.

Plume rise (Δh). The change in the plume height, Δh , is a function of the stackgas exit velocity, the diameter of the stack, the temperature of the gas at the stack, the ambient temperature, wind speed, atmospheric stability, and other factors (Hanna et al., 1982; EPA, 1995f). (Hanna et al., 1982). For motor vehicles and some building vents, Δh is close to zero (Hanna et al., 1982). However, for elevated sources, Δh can be 2 to 10 times h_S (Hanna et al., 1982).

Various sources give equations for Δh depending on the stability of the atmosphere, and whether the plume rises mainly on account of the heat (buoyancy) or the velocity (momentum) of the emitted gases. In our calculations below, we assume a neutral atmosphere. Regarding buoyancy versus momentum, Hanna et al. (1982) state that typically after only 50 meters, buoyancy dominates momentum. Hence, we model buoyancy-dominated plume rise.

Formally, we modify slightly the EPA's (1995f) formula for Δh for buoyancy-dominated rise in near-neutral atmospheric conditions:

$$\Delta h = \frac{\beta \cdot Fb^a \cdot x^b}{\sqrt{w^2 + \left(\frac{v_g}{4}\right)^2}}$$

$$Fb = \frac{g \cdot v_s \cdot d_s^2 \cdot (T_s - T_a)}{4T_s}$$

where:

Δh and x are as defined above

w = the mean wind velocity at height h, taken to be along the \mathbf{x} axis (m/s; see below)²¹

 v_g = the ground speed of the exhaust source (m/sec; assumptions by source category are shown in Table 16-15)

Fb = the buoyancy flux parameter (m^4/sec^3)

g = the gravitational constant (9.8 m/sec²)

 $v_{\rm S}$ = the velocity of the stack gases (m/sec; assumptions by source category are given in Table 16-15)

 d_S = the inside diameter of the end of the exhaust (meters; assumptions by source category are given in Table 16-15)

 T_S = the temperature of the stack gases (${}^{O}K$; assumptions by source category are given in Table 16-15)

 T_a = the ambient temperature (${}^{\circ}$ K; assumed to be 298 for all sources)

 β , a, b = coefficients, assigned as follows (EPA, 1995f):

	xf	$x_f > x$	
coefficient	Fb < 55	Fb ≥ 55	
β	21.425	38.71	1.60
a	0.75	0.60	0.33

²¹See note 18 above.

b	0.00	0.00	0.67
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 x_f = the x-axis distance from the source to the "final rise" -- the point at which Δh is reached (meters):

$$x_f = 49 \cdot Fb^{0.625}$$
 if $Fb < 55$
 $x_f = 119 \cdot Fb^{0.40}$ if $Fb \ge 55$

Note that we have, and use, actual statistics on v_s , d_s , and T_s for major point sources in five categories: i) fuel combustion, electric utilities; ii) fuel combustion, industrial; iii) fuel combustion, other (mainly residential wood combustion); iv) chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes; and v) solvent utilization, storage and transport, and waste disposal and recycling (Table 16-17; from EPA [1995d]).

Our minor modification to the EPA's (1995f) formula is to account for the ground speed of the source in the calculation of the effect of horizontal wind on the plume rise. In general, the plume rise is related to the horizontal velocity of the stack gases, which is the sum of the velocity vector due to the wind (w) and the velocity vector imparted by the movement of the source. If the emissions source is stationary, the horizontal velocity of the stack gases is due entirely to -- and equal to -- the wind velocity: $v_g = 0$, and the denominator of the Δh equation reduces to w, the atmospheric wind speed. (This is how the equation is presented by the EPA [1995f], which is concerned with stationary sources only.)

To estimate the horizontal velocity of the stack gases, as the sum of the vector due to the wind and the vector due to the momentum imparted by the movement of the source, one must estimate the magnitude of the two vectors, and the angle between them. The wind speed will remain more or less constant in magnitude (w) over the horizontal scales of interest. However, the speed imparted by the movement of the source will diminish rapidly with distance, from v_g, the source speed, at immediate release, to near zero in a relatively short distance, as the exhaust gases dissipate their forward momentum in collisions with ambient gases. At the point of final rise of the plume, the speed due to the initial momentum from the moving source might well be essentially zero. Ideally, one would estimate a weighted-average speed due to source momentum, as the area under the speed-distance curve divided by the distance. However, for simplicity, we simply assume that the average speed of the gases due to the movement of the source is one-quarter of the initial speed of the gases, which is the speed v_g of the source itself. Although this is a crude assumption, it is not important, because the calculated results for DN_i are not sensitive to plausible variations in the value of vg.

Regarding the angle between the two vectors (w and v_g), if vehicle travel is random with respect to the wind direction, then, on average, the wind vector and the

source travel vector will be perpendicular, and the sum of the two -- the stack-gas horizontal velocity -- will be the hypotenuse of the resulting right triangle.

Note that the ground speed of the source is relevant only here, in the calculation of the effect of wind on the plume rise; it is not relevant to other effects of wind on dispersion, because after a short distance the gas speed imparted by the movement of the source itself drops to zero.

Note, too, that the Δh formula results in very small absolute values of Δh for motor-vehicle source, consistent with the statement by Hanna et al. (1982), mentioned above, that for motor vehicles and some building vents, Δh is close to zero.

The calculated results for DN_i are somewhat sensitive to plausible variations in the values for T_s , V_s , and d_s . Lower values for these variables result in higher DN_i and hence lower costs to motor vehicles. Our low and high values for these parameters for the 13 source categories are shown in Table 16-15.

Receptor height ($z_{\underline{r}}$). In our analysis, the receptors are the ambient air-quality monitors, which are the sources of the pollution data for use in the air-pollution damage functions. Consequently, we need to represent the height of ambient air-quality monitors.

We assume that most monitors are at or near ground level, about 3 meters off of the ground. The calculated results for DN_i are only moderately sensitive to plausible variations (from about 2 to 5 meters) in the height of the ground-level receptor.

<u>Mean wind velocity at source height h (w).</u> Generally, the mean wind speed increases with height. Because the Gaussian equation requires the wind speed at the effective source height, we estimate the wind speed at height h by scaling wind speed w' at reference height h' according to the power-law formulation:

$$w = w' \left(\frac{h}{h'}\right)^p$$

w = the mean wind speed at the effective source height h (m/sec)

w' = the mean wind speed at the reference height h' (discussed below)

h = the effective source height (0.5 m < h < 200 m; discussed above)

h' = the reference height, at which the reference wind speed is measured (10 m)

p = power-law exponent (ranges from about 0.10 for moderately unstable conditions to 0.35 for slightly stable conditions, where stability is defined according to the Pasquill six-category system mentioned described above [EPA, 1995f; Hanna et al., 1982])

In this formula, the effective height h cannot be less than 0.5 m or more than 200 m, because the formula does not give reasonable results for heights outside of these bounds.

Reference wind speed and height (w' and h'). The average annual wind speed (measured at airports) varies surprisingly little throughout the U.S.: from 6.3 mph in

Charleston, West Virginia and Phoenix, Arizona to 13.0 mph in Cheyenne, Wyoming (Bureau of the Census, *Statistical Abstract of the United States* 1992, 1992). In most cities in the U.S., the average annual wind speed is around 9 mph (about 4 meters/second). We assume an average of 3-5 meters/second everywhere²².

The *Statistical Abstract* does not give the reference height for these measurements, but Hanna et al. (1982) use a reference height of 10 meters, Ahrens (1985) states that surface winds should be measured at a height of 10 meters, and Pasquill (1974) states that surface speeds conventionally are given at 10 meters. Therefore, we assume that h' = 10 m.

Sensitivity analyses indicate that the higher wind speed results in higher $\mathrm{DN_i}$ and hence lower costs. Hence, we use 5 m/sec in our low-cost case, and 3 m/sec in our high-cost case.

In the model, the wind speed w is not allowed to be less than 0.50 m/sec, at any height. There are two reasons for this. First, it is not physically realistic to extend the power-law formula down to near-zero ground heights. Hanna et al. (1982) note that rarely is the wind speed truly zero, and that "calm winds are defined as 0.5 m/s". Second, a zero wind speed generates infinite travel time from source to receptor in the Gaussian equation.

Horizontal and vertical diffusion parameters (σ_y and σ_z). The estimation of the horizontal and vertical diffusion parameters is perhaps the biggest challenge in Gaussian dispersion modeling. Generally, these parameters are represented by curves fit to data generated from field tests under a variety of conditions. We have chosen to use Draxler's (1976) simple empirical formulas, which are a fit to dispersion data from experiments on both ground-level sources and elevated sources, because they do distinguish between ground-level and elevated sources:

$$\sigma_{y} = \frac{\sigma_{\theta} \cdot x}{1 + \beta_{y} \cdot T^{\alpha_{y}}}$$
 ground - level and elevated sources

$$\sigma_z = \frac{\sigma_\phi \cdot x}{1 + \beta_{ze} \cdot T^{\alpha_{ze}}}$$
 elevated sources

$$\sigma_z = \sigma_\phi \cdot x \cdot \left(\beta_{zg} \cdot \left(T - c_{zg}\right)^{\alpha_{zg}} + d_{zg}\right)^{j_{zg}}$$
 ground - level sources

where:

²²In their analysis of the effect of location on the damage cost of emissions from transportation, Eyre et al. (1997) assume 7.5 m/sec for Great Britain.

- σ_y = the horizontal diffusion parameter: the standard deviation of the distribution of the concentration C in the direction perpendicular to the wind (meters)
- σ_Z = the vertical diffusion parameter: the standard deviation of the distribution of C in the vertical direction (meters)
- x =the x-axis distance from source to receptor (meters)
- T =the travel time = x/w (sec)
- σ_{θ} = the standard deviation of the horizontal wind angle θ (radians; discussed below)
- σ_{φ} = the standard deviation of the vertical wind angle φ (radians; discussed below)
- Coefficients for stable and unstable conditions²³ estimated as follows (from Draxler, 1976, except as noted):

	Stable	Unstable
σy Ground level (0-20 m)		
β _V T≤550	0.052	0.052
β _y T>550	28.5	0.052
α _V T≤550	0.500	0.500
α _V T>550	-0.500	0.500
σ_V Elevated (above 20 m)		
$eta_{ m V}$	0.028	0.028
$lpha_{ m V}$	0.500	0.500
σ_z Ground level (0-20 m)		
$\beta_{ m Zg}$	0.127	0.001875*
$c_{\mathbf{Z}\mathbf{g}}$	0	0*
$\alpha_{ m Zg}$	0.5	1.31*
dzg	1	0.7
jzg	-1	1
σz Elevated (above 20 m)		
$\beta_{ m ze}$	0.023	0.040
$\alpha_{ m ze}$	0.806	0.500

^{*}Different from Draxler's (1976) original values, as explained below

We assume that ground-level sources are from 0 to 20 m, and that "elevated" sources are above 20 m. Draxler (1976) did not actually specify the range, but in the "ground-level" experiments that provided the data from which his equations are derived, the heights ranged from 1 meter to 17 meters.

52

²³See note 17 above.

Note that the values for β_{Zg} , c_{Zg} , and α_{Zg} , in the equation for σ_{Z} for ground-level sources in unstable conditions, are different from Draxler's (1976) original values of 0.0001875, 40, and 2, respectively. We set c_{Zg} = 0 to prevent the possibility of taking root of a negative number (which would have occurred if T < 40 and c_{Zg} = 40). We increased β_{Zg} (by a factor of 10) and decreased α_{Zg} to make the pattern of results fit more closely with those reported by Griffiths (1994) and EPA (1995f) for σ_{Z} for unstable conditions²⁴.

Finally, recall that if the $\sigma_Z > a \cdot z_i$, where z_i is the mixing height, then the concentration is assumed to be homogeneous vertically, and a different equation is used to calculate the concentration (equation 8b rather than 8a).

The standard deviation of θ and ϕ . Ideally, these parameters should be calculated from local data on wind variation. Verrall and Williams (1982) and Hanna et al. (1982) supply several formulas for calculating σ_{θ} , but we cannot obtain the necessary input data for every county, and so cannot use them. Consequently, we simply must assume low and high bounds for national averages.

 σ_{θ} : Several sources provide data on typical values of σ_{θ} :

- Benson (1984) assumes that σ_{θ} = 10° for "the standard input value for the sensitivity analysis" (p. 73) for the highway/line-source dispersion model CALINE4.
- Nokes and Benson (1985) report "worst-case" one-hour values for σ_{θ} that range from 5° to 30°. For longer averaging times, the values are much higher.
- Hanna et al. (1982) shows a table from Cramer in which σ_{θ} at 10 meters varies from 3°0 in extremely stable conditions to 30°0 in extremely unstable conditions (stability here defined according to Cramer's own system), and another table, from Gifford, in which σ_{θ} at 10 meters varies from 2.5°0 in moderately stable conditions to 25°0 in very unstable conditions (stability here defined according to Pasquill's categories, discussed above).
- Pasquill (1974) cites studies in England and Nebraska in which σ_{θ} varies from 2^{o} to 6^{o} at 16 m over open grassland under neutral conditions. In unstable conditions, the values are 2-3 times higher.

These data suggest that at 10 meters, σ_{θ} ranges from 5° (slightly stable) to 20° (moderately unstable). However, equations in Hanna et al. (1982) indicate that σ_{θ} declines with height. Our analysis of those equation suggests that the relationship between the change in source height h and the change in σ_{θ} is approximately: σ_{θ} = 5· $(10/h)^{0.33}$ for stable conditions, and σ_{θ} = 5· $(10/h)^{0.41}$ for unstable conditions.

 σ_{φ} : Data from Cramer (in Hanna et al., 1982) indicate that at 10 meters:

53

²⁴Draxler (1976) estimated a bulk Richardson number for each experiment. If this number was greater than 0, the conditions were said to be stable; if less than zero, then unstable. This classification appears to correspond reasonably well with Pasquill's stability classes. In the experiments described by Draxler (1976), the release time was on the order of 30 to 60 minutes.

$$\sigma_{\phi} = 0.33 \cdot \sigma_{\theta}$$

Other formula in Hanna et al. (1982) indicate that σ_{ϕ} also declines with height. With these formula, we estimate that σ_{ϕ} changes with the 0.33 power of the change in height in stable conditions, and with the 0.25 power in unstable conditions.

Thus, on the basis of the data and formulas presented or mentioned above, we assume the following:

	σ _θ (degrees)		σ_{ϕ} (degrees)	
	Stable	Unstable	Stable	Unstable
Ground-level (0-10m)	5	20	1.7	6.7
Elevated (above 10 m)	5· (10/h) ^{0.33}	5· (10/h) ^{0.41}	5· (10/h) ^{0.33}	$5 \cdot (10/h)^{0.25}$

where h is the effective source height, defined above.

Low-high weights on stability parameters. We ran tests to determine which set of parameters -- stable or unstable -- gave high $\mathrm{DN}_{p',i}$ and hence low costs attributable to motor vehicles, and which gave the high-cost results. It turns out that the parameters for "unstable" conditions result in high $\mathrm{DN}_{p',i}$ and hence low cost for motor vehicles. Therefore, in our low-cost case we give more weight to unstable conditions than we do in our high-cost case.

In both the low-cost and the high-cost case, we give more weight to the parameters for unstable conditions than to the parameters for stable conditions. We have two reasons for this. First, we assume that neutral conditions, which Draxler (1976) did not model, are represented better by the parameters for unstable conditions than by the parameters for stable conditions. Second, the parameters for unstable conditions give more reasonable results.

Therefore, in our low-cost case we assign a weight of 75% to the parameters for unstable conditions, and 25% to the parameters for stable conditions. In the high-cost case, we assign a weight of 60% to the parameters for unstable conditions, and 40% to the parameters for stable conditions.

The final calculated DNi results turn out to be sensitive to these weights. If the weight on the parameters for unstable conditions is more than 98% or less than 50%, the estimated DNi are quite different from the range used here. Because our grounds for selecting the weights of 75% and 60% (see above) are relatively weak, this sensitivity is defect of the model.

Adjustments for particle settling and deposition (D1, D2). Particles can settle out of a pollution plume on account of gravity, and small particles and some gases can be deposited on surfaces as a result of diffusion and Brownian motion (Hanna et al., 1982). This settling and deposition depletes the pollution plume, and reduces the

pollutant concentration at the receptor. The parameters D1 and D2 adjust the standard Gaussian plume model to account for this depletion.

Note that only particles "settle", but that some gases as well as particles can be deposited on surfaces.

The rate of settling or deposition is a function of the settling velocity, the deposition velocity, and the vertical diffusivity, which in turn are functions of the particle size and other factor. Ermak (1977) derives the following expressions²⁵ for D1 and D2:

$$D1 = e^{\left(\frac{-W \cdot (z_r - h)}{2 \cdot K} - \frac{W^2 \cdot \sigma_z^2}{8 \cdot K^2}\right)}$$

$$D2 = -\frac{2.51 \cdot V_1 \cdot \sigma_z}{K} \cdot e^{\left(\frac{V_1 \cdot (z_r + h)}{K} + \frac{V_1^2 \cdot \sigma_z^2}{2 \cdot K}\right)} \cdot erfc(M)$$

$$M = \frac{V_1 \cdot \sigma_z}{1.414 \cdot K} + \frac{z_r + h}{1.414 \cdot \sigma_z}$$

$$V1 = V - \frac{W}{2}$$

where:

 z_r , h, and σ_z are as defined above

W = the gravitational settling velocity (m/sec; discussed below)

V = the deposition velocity (m/sec; discussed below)

K =the vertical eddy diffusivity ($m^2/$ sec; discussed below)

erfc(M) = the complementary error function of M (discussed below)

The settling velocity (W). The gravitational settling velocity is estimated on the basis of Stoke's law for the terminal settling speed of spherical particles, and the relationship between the aerodynamic diameter of a particle and the diameter of the sphere with the same settling velocity. First, we express represent the settling velocity of a spherical particle, as a function of the particle size and density (Altshuller et al., 1996):

 $^{^{25}}$ The formula given on page 233 of Ermak's (1977) article shows that the z_r +h term in the expression for D2 is squared. This is a misprint. The derivation of the expression given in the Appendix to his article, and an examination of the units (the D2 expression must be unitless), show that the z_r +h should be as shown here.

$$W_{sp} = \frac{\left(\rho_{sp} - \rho_{air}\right) \cdot g \cdot d_{sp}^{2} \cdot SL}{18 \cdot \eta_{air}} \cdot c$$

where:

 W_{SD} = the gravitational settling velocity of a spherical particle (m/sec)

 ρ_{sp} = the density of the spherical particle (g/cm³)

 ρ_{air} = the density of air (0.00129 g/cm³)

g = the gravitational constant (9.81 m/sec²)

 d_{Sp} = the diameter of the spherical particle (μm)

 η_{air} = the absolute viscosity of air (1.81 · 10⁻⁴ g/cm/sec)

c = constant to convert units to m/sec $(10^6 \text{ cm}^3/\text{m}^3 \cdot 10^{-12} \text{ m}^2/\mu\text{m}^2 \cdot (10^2 \text{ cm/m})^{-1} = 10^{-8} \text{ cm}^2/\mu\text{m}^2)$

SL = the slip correction factor for the spherical particle (unitless; discussed below)

Now, we need to work from this to an expression for the settling velocity for non-spherical particles, because most actual ambient pollutant particles have irregular shapes. To do this, we note that the aerodynamic diameter of a particle, which is the particulate measure usually reported, and which we in fact know for different emission sources, is *defined* as the diameter of the spherical particle that has the same settling velocity W as the actual particle, but a material density of 1 g/cm^3 . Thus:

$$W_{a} = W_{sp} \{d_{a}, \rho_{o}\}$$

$$W_{a} = \frac{(\rho_{o} - \rho_{air}) \cdot g \cdot d_{a}^{2} \cdot SL_{a}}{18 \cdot \eta_{air}} \cdot c$$
(10a)

where:

 W_a = the gravitational settling velocity of the actual particle (m/sec)

 d_a = the aerodynamic diameter of the particle (μ m; values for different emissions sources given in Table 16-15)

 ρ_0 = the unit density of the spherical particle with aerodynamic diameter d_a and the same settling velocity of the actual particle (1.0 g/cm³)

 SL_a = the slip correction factor for the spherical particle evaluated at the aerodynamic diameter d_a (unitless; discussed below)

Substituting the values for the constants ρ_0 , ρ_{air} , g, c, and η_{air} results in the following simple expression, without any additional approximations:

$$W_a = 0.00003 \cdot d_a^2 \cdot SL_a$$

The slip correction factor is given by (Altshuller et al., 1996):

$$SL_a = 1 + \frac{\lambda}{d_a} \cdot \left(2.514 + 0.800 \cdot e^{\left(\frac{-0.55 \cdot d_a}{\lambda}\right)} \right)$$

where:

da is the aerodynamic diameter, as above

 λ = the mean free path of air molecules (0.0653 µm; Altshuller et al., 1996)

The deposition velocity (V). Particles and gases mix and diffuse toward the surface of the earth, where chemical absorption, impaction, photosynthesis, and other biological, chemical, and physical processes cause the material to be retained at the surface (Hanna et al., 1982). The rate of deposition depends on the characteristics of the atmosphere, particles and gases, and surface (Hanna et al., 1982; Altshuller et al., 1996). Reactive gases, such as ozone, deposit much more readily than non-reactive gases such as CO. Surfaces such as forest canopies and mosses give rise to relatively high deposition rates (Hanna et al., 1982). Finally, precipitation and clouds change the deposition rates: for example, precipitation can substantially increase the deposition rate of particulate matter.

The deposition velocity V (m/sec) is defined as an empirical function of the observed deposition flux F (g/ m^2/s) and the concentration near the surface Co (g/ m^3) (usually measured at 1.0 m) (Altshuller et al., 1996; Hanna et al., 1982):

$$V = F/Co$$

We will estimate the deposition velocity of particles, and of reactive and non-reactive gases.

<u>Particles:</u> The dry deposition velocity of particles is a function of the size of the particle and other factors. To estimate this relationship, we fit equations to the data of Lin et al. (1994), who measured the deposition velocity of particles as a function of particle aerodynamic diameter and wind speed, and of Lin et al. (1993), who show deposition velocity versus particle size as calculated by four models. The following equations fit the measured data and the model results reasonably well:

$$d_a < 0.3 \mu m$$
: V = 0.001 cm/sec = 0.00001 m/sec (10b)

$$d_a \ge 0.3 \ \mu m: \log(V) = -4.01 + 1.89 \cdot \log(d_a)$$
 (10c)

where d_a is in μm and V is in m/sec.

<u>Gases</u>: The dry deposition of velocity of non-reactive gases, such as CO, is very low -- on the order of 10^{-3} to 10^{-4} cm/sec (Hanna et al., 1982) (Eyre et al., 1997, assume that it is "negligible".) By contrast, the dry deposition velocity of more reactive gases, such as SO₂ and O₃, can be much higher. For example, the deposition velocity of ozone ranges from 0.02 to 1.4 cm/sec, depending on the surface, with an average, according to Hanna et al. (1982), of 0.5 cm/sec.

Table 16-18a summarizes several estimates and assumptions of deposition velocities. Note that the deposition velocities in Langner and Rodhe (1991) and in Dastoor and Pudykiewicz (1996) apply at 1.0 m height. Both articles present formulas to estimate the deposition velocity at other heights. We ignore this correction.

Most of the estimates in Table 16-18a are in reasonable agreement. Given these estimates and data, our assumptions and calculations regarding settling and deposition velocity are shown in Table 16-18.

The vertical eddy diffusivity (K). Ermak's (1977) deposition model, used above, defines the standard deviation of the concentration in the vertical direction, σ_Z , in terms of the vertical eddy diffusivity²⁶:

$$\sigma_z^2 = \frac{2}{w} \cdot \int_0^x K(x') dx'$$

Thus:
$$K = \frac{\sigma_z^2 \cdot w}{2 \cdot x}$$

where all the terms are as defined above (lower-case w is the wind speed at the effective source height)

The complementary error function (erfc (M)). The complementary error function is defined as:

$$erfc(M) = \frac{2}{\sqrt{\pi}} \cdot \int_{M}^{\infty} e^{-t^2} dt$$

There is no analytical solution to this integral. However, several good approximations are available. We use the one employed in CALINE4, a highway line-source dispersion model (Benson, 1984):

$$erfc(M) = \left(\frac{0.3480242}{1 + 0.47047 \cdot M} - \frac{0.0958798}{\left(1 + 0.47047 \cdot M\right)^2} + \frac{0.7478556}{\left(1 + 0.47047 \cdot M\right)^3}\right) \cdot e^{-M^2}$$

²⁶This is the form used in CALINE4, a highway line-source air quality model (Benson, 1984).

Adjustment for removal by chemical reaction (D3). The concentration of a pollutant can be reduced by chemical transformation as well as by deposition and settling. For example, as discussed below, NO_X reacts with ammonia to form nitrate particulates. The more NO_X reacted, the less the ambient concentration of NO_X (and the higher the ambient concentration of nitrate particulate). Similarly, CO reacts with the OH^{\bullet} radical to form CO_2 , albeit relatively slowly. The more CO reacted, the lower the ambient concentration of CO. Consequently, for those ambient pollutants whose concentration we model (CO, NO_2 , O_3 , and PM), we need to account for any reduction in the ambient concentration due to chemical transformation.

Recall from section 16.1.2 that we consider the following pathways from emitted pollutants to ambient pollutants:

- 1) CO --> CO
- 2) NO_X --> NO₂
- 3) $NO_X --> PM_{10}$, $PM_{2.5}$
- 4) $NO_X --> O_3$
- 5) VOCs --> O3
- 6) VOCs --> PM2.5
- 7) SO₂ --> PM₁₀, PM_{2.5}
- 8) NH3 --> PM10, PM2.5
- 9) PM_{2.5-10} (also called "coarse" PM₁₀) --> PM₁₀
- 10) PM_{2.5} --> PM_{2.5}, PM₁₀

The CO-->CO pathway (#1). As mentioned above, O reacts with the OH • radical to form CO₂. The further the source from the monitor, the longer the time for CO to react with OH •, and hence the lower the CO concentration at the monitor. Consequently, we will model the removal of CO by chemical reaction (term D3 in equations 8 and 9) simply as the amount reacted per hour:

$$D3 = \left(1 - \frac{RR}{100}\right)^{\frac{T}{3600}}$$

where:

D3 = the adjustment for removal by chemical reaction, in equations 8 and 9

RR = the reaction removal rate (% of pollutant removed per hour; discussed below)

T = the travel time from emission source to receptor (seconds; calculated above as the downwind distance x divided by the wind speed w)

3600 = seconds/hour

The EPA (1992) reports that CO has a lifetime of 1 to 4 months, with an average of about 2 months, or 1460 hours. This implies a removal rate of 0.0475%/hour, which results in 50% of CO remaining after 1460 hours. In their analysis of the effect of location on the damage cost of emissions from transportation, Eyre et al. (1997) assume 0.054%/hour. Therefore, we assume that for CO, RR = 0.05%/hr (Table 16-18).

The NO_X pathways (#s 2, 3, and 4). Nitrogen oxides are involved in several complex reaction pathways. Ideally, we would model these paths dynamically, as they occur in the plume, and keep track of the reactants and the products. However, this is beyond our scope. Instead, we will make three simplifying assumptions. First, we will assume that the NO_X --> O3 pathway (#4) does not deplete NO_X , but rather shifts the equilibrium of NO and NO_2 . Thus, we will assume that this pathway does not reduce the amount of NO_X that can end up as ambient NO_2 (#2) or as nitrate (#3). Second, we will assume that NO_X that is converted to nitrate (#3) is in fact removed from the ozone chemistry pathway (#4) and the ambient NO_2 pathway (#2). Third, we will assume that NO_X --> nitrate is the only chemical removal route for NO_X . (Depostion is treated separately, above.)

With these assumptions, we need only to calculate the amount of NOx that is converted to nitrate, and then subtract that amount from the amount that can become ambient NO₂ (#2) or participate in ozone chemistry (#4). As explained below, we assume that 5-7% of the nitrogen in NO_X is converted to nitrogen in nitrate. We therefore deduct 5-7% of NO_X emissions from the amount available for paths #2 (NO₂) and #4 (O₃)²⁷.

The VOC pathways (#s 5 and 6). VOCs participate in ozone formation, and also can form organic aerosols. However, the transformations of VOCs in ozone chemistry do not necessarily remove VOCs from particulate chemistry, and the transformations of VOCs in particulate chemistry do not necessarily remove VOCs from ozone chemistry (in the way that the conversion of NO_X to particulate nitrate removes NO_X from ozone chemistry). For example, some organic aerosols (particulate VOCs) are reactive, and hence can participate in ozone chemistry (Winer and Busby, 1995). Thus, there is no basis for simply assuming that VOCs involved in ozone chemistry deplete the amount of VOCs available to form particulate nitrates, or vice versa. The chemistry in fact is much more complex than this, and well beyond the scope of our modeling efforts here. If we are to make a simple assumption, the most reasonable is that ozone chemistry does not remove VOCs from particulate chemistry, and particulate chemistry does not

60

 $^{^{27}}$ As discussed below, most of the nitrate, formed from 5% to 7% of NO_X, will be nuetralized by ammonia to form ammonium nitrate, which we count as particulate matter. However, in some instances there will not be enough ammonia to fully nuetralize all of the nitrate. We ignore any such un-nuetralized nitrate: we do not count it as particulate matter, do not have a separate dose-response function for it, and exclude it from ozone chemistry.

remove VOCs from ozone chemistry. Consequently, we assume that for VOCs, the term D3 in equations 8 and 9 is equal to 1.0

The SO_X and NH_3 pathways (#s 7 and 8). We assume that these pollutants react to form particulates (particulate sulfate and particulate nitrate). However, we model this reaction as occurring instantaneously at the monitor; we do not model the reaction dynamically, as a function of time or distance. Moreover, we do not care about the concentration of SO_X and NH_3 per se, because we do not have any dose response functions for these pollutants. As a result, we do not need to model the removal (or conversion) of SO_X or NH_3 over time; we simply calculate the conversion to particulates at the monitor, as described below. Thus, for these paths and pollutants, the term D3 = 1.0

The PM pathways (#s 9 and 10). We assume that PM emitted as such is inert, and is not removed in appreciable amounts by chemical reactions (apart from deposition). Thus, for PM, term D3 = 1.0.

Adjustment for multiple reflections (S1). The Gaussian model assumes that molecules are reflected perfectly, off of the ground, and off of the underside of the inversion layer. We follow the EPA (1995f), and use the method of multiple "virtual" stacks to model multiple "bounces" off of the ground and the inversion layer as the molecules move downwind. In this method, the first bounce off of the ground is modeled as an emission from the inverted image of the actual effective stack (with a height of -h). The first bounce off of the underside of the inversion layer is modeled as an emission from a virtual effective stack that sticks as far above the inversion layer as the actual effective stack is below (with a height of $2 \cdot z_i$ -h). The second bounce off the ground, which follows the first bounce off of the inversion layer, is modeled as an emission from an inverted virtual effective stack of the same height as the virtual effective stack that generated the first bounce off of the underside of the inversion layer (-($2 \cdot z_i$ -h)). This continues, with ever higher virtual effective stacks and virtual inverted effective stacks, until the desired number of bounces have been modeled. Formally, we model 50 bounces, with the following model (EPA, 1995f):

$$S1 = \sum_{j=1}^{50} e^{0.5 \cdot \left(\frac{H1}{\sigma_z}\right)^2} + e^{0.5 \cdot \left(\frac{H2}{\sigma_z}\right)^2} + e^{0.5 \cdot \left(\frac{H3}{\sigma_z}\right)^2} + e^{0.5 \cdot \left(\frac{H4}{\sigma_z}\right)^2}$$

$$H1 = z_r - \left(2 \cdot j \cdot z_i - h\right)$$

$$H2 = z_r + \left(2 \cdot j \cdot z_i - h\right)$$

$$H3 = z_r - \left(2 \cdot j \cdot z_i + h\right)$$

$$H4 = z_r + \left(2 \cdot j \cdot z_i + h\right)$$

where:

 z_r = the height of the receptor (m; discussed above)

 z_i = the height of the mixing layer (m; discussed below)

h = the effective source height (m; discussed above)

 σ_Z = the vertical diffusion parameter (m; discussed above)

<u>The effect of the mixing height (zi)</u>. Note that the concentration C at the receptor is determined by one of three different formulas (8a, 8b, or 8c), depending on the relationship between source height h and mixing height z_i , and between σ_z and z_i . In this section, we discuss the mixing height z_i its role in determining the pollutant concentration.

The mixing height is the height of the well-mixed layer of the atmosphere. It defines the region in which pollutants emitted near the surface can mix. Usually, above the mixed layer there is a temperature inversion, which prevents mixing of the air above the inversion with air below the inversion. Thus, the height of mixing layer limits the vertical spread of pollutants, and thereby strongly influences the concentration at the ground.

Pollutants emitted below the mixing height -- below the inversion -- are effectively capped by the inversion. If the inversion layer/mixing height is relatively high, the vertical spread of pollution can be large, and the ground concentration relatively low. However, if there is a low inversion, pollutants emitted at ground level will be trapped and in high concentration near the ground. Such are the conditions during particularly bad pollution episodes.

Conversely, pollutants emitted above an inversion will not mix with the pollutants below, in the air near the ground. In this case, the concentration at the ground will be zero.

We thus have two effects to model: the inversion layer as a cap on pollution, and the inversion layer as a floor on pollution.

Inversion layer as a cap on pollution. The inversion layer limits the vertical spread of pollution and hence the standard deviation of concentration in the vertical direction. Close to the emissions source, the atmospheric mixing height generally will have little effect on the plume, because few pollution molecules will have reached the mixing height (unless the stack height is very close to the mixing height). That is, if the time T and distance x and hence σ_z are relatively small, then we generally we can ignore the effects of z_i .

However, as the plume progresses downwind, more and more pollution will run up against the underside of the inversion layer. This pollution will be reflected off of the inversion layer towards the surface, increasing the concentration at the surface but also increasing the homogeneity -- reducing the standard deviation -- of the vertical concentration. Far enough downwind, one can assume that the pollution simply is uniformly mixed between the ground and the top of the mixing layer.

Consequently, we handle this effect of the mixing height with a simple dichotomous model. We establish a value of σ_z , the vertical standard deviation of concentration, as a fraction of z_i ($\sigma_z = a \cdot z_i$), such that:

- below this value $(a \cdot z_i)$ the concentration is calculated on the basis of a Gaussian distribution in the vertical but with multiple reflections of the plume off of the underside of the inversion layer (term S1 in equation 8a).
- \bullet above this value (a·z_i), the pollution is assumed to be distributed uniformly throughout the mixed layer.

The EPA's ISC model makes similar assumptions, and adopts a value of a = 1.6. We adopt assume that a=20.0 for unstable conditions, and 75.0 for stable conditions. When the pollution is assumed to be uniformly distributed in the vertical, the form of the model is as shown equation 8b.

Of course, this dichotomous modeling does have the disadvantage of creating a discontinuity in the concentration at the point ($\sigma_z = a \cdot z_i$).

Inversion layer as floor on pollution. To model the case in which the pollutants are emitted above the inversion layer and cannot reach the air ground level, we simply make C = 0 if $h > z_i$. (In our model, this actually is handled by setting the term D1 equal to zero if $h > z_i$.)

The mixing height. Holzworth (1972; presented in Anthes et al., 1975) shows contours of the mean annual morning and afternoon mixing depths across the U.S. In the morning, when conditions are relatively stable, the depths typically range from 600 to 900 meters along the coasts, to 300 to 600 meters inland. In the afternoon, when conditions are more unstable, the depths are greater -- from 800 to 2600 meters. (There is much more diurnal variation inland than on the coasts because the diurnal variation in ground temperature is much more than the diurnal variation in ocean temperature.)

These data suggest that during stable conditions, z_i is around 400 meters, and during unstable conditions, around 1500 meters. We adopt these values here. Pasquill (1974) suggests that during stable conditions, material released from the ground will not spread above 300 m.

16.3.3 The results of the model

We can use the simple dispersion model presented above to estimate the contribution to ambient air pollution (per kg of emissions) of any source relative to the contribution of light-duty vehicles. This contribution is represented by the parameter $DN_{p',i}$ in equations 6, 7, and 9. We will estimate this parameter for the 13 general emission sources categories shown in Table 16-15, and for five different pollutant categories: fine PM (PM2.5, which we assume includes secondary organic aerosols); coarse PM (less than 10 μ m but greater than 2.5 μ m), CO, NO_X (we assume that the DN_i for NH_3 are the same as the DN_i for NO_X), VOCs, and SO_X . Thus, we have 65 pollutant and source combinations. We estimate 12 sets of results: 6 for monitors in urban areas, and 6 for monitors in agricultural areas. Each of these six sets consists of two sets (low

and high) for in-county emissions, two sets (low and high) for out-of-county emissions in small AQCRs, and two sets (low and high) for out-of-county emissions in large AQCRs. Thus, we have $5 \times 13 \times 12 = 780$ separate estimates of $DN_{p',i}$.

Table 16-15 shows, for each emission source, the assumptions for the input parameters that can vary by emissions source. The calculated $\mathrm{DN_{i}}$, by pollutant and emission source-category, for in-county and out-of-county emissions, are shown in Tables 16-19a to 16-19l. Tables 16-19a through 16-19f show the results for urban monitors, and 16-19g through 16-19l show the results for agricultural monitors.

With our model and assumptions, nearly all in-county emissions sources contribute less to ambient air pollution, per kg of emissions, than do light-duty vehicles. The contribution of fuel-combustion by electric utilities is quite small (perhaps too small), mainly on account of the great distance and height of power-plant stacks. The contribution of out-of-county emissions is more uniform, mainly because of our assumption that all out-of-county sources are located at the same distance.

Of course, the results of Table 16-19 are quite uncertain -- even more uncertain than is indicated by our low-high range -- because they are based on a simplified representation of complex phenomenon. The model is especially sensitive to the weight given to the parameters for unstable atmospheric as opposed to stable atmospheric conditions. At one extreme, one can specify sets of parameter values that make motor vehicles responsible for essentially all of the ambient pollution (DNp', i = 0), and at the other extreme, values that make motor vehicles responsible for essentially non of the pollution (DNp', i = 20 or more). The model behaves this way because it is highly nonlinear, and very sensitive to small changes in some of the key parameters.

16.3.4 Comparison with other estimates

The application of the model summarized above gives us some idea of the "average" magnitude of the parameter $\mathrm{DN}_{p',i}$ in equations 6, 7, and 9. However, the results are so sensitive to plausible changes in input parameters that it is important to check them against other estimates of $\mathrm{DN}_{p',i}$.

The EPA (1994b) has used a model similar to but somewhat more sophisticated than ours to estimate exposure to emissions of particulate matter. They estimated tons of "effective" PM emissions, which consist of direct emissions plus secondary ammonium sulfate and ammonium nitrate PM formed from NO_X , SO_2 , and NH_3 emissions, and then used a Gaussian dispersion model to estimate exposure in terms of persons- $\mu g/m^3$. If we take the ratio of exposure to emissions for each source i, and then compare each of these ratios to the exposure:emissions ratio for motor-vehicles, we have an estimate of the equivalent of $DN_{PM,i}$ in our equations 6, 7, and 9^{28} . The results of this exercise are shown in Table 16-20. The lower the ratio relative to motor-vehicles - the lower the values of the last two columns -- the lower the exposure to one unit of PM emissions from source i compared to one unit of PM emissions from motor vehicles. Thus, the EPA's (1994b) estimates indicate that if fuel combustion and motor-vehicles produced the same amount of emissions, the contribution of fuel combustion to ambient PM at an air-quality monitor would be 1/4 to 1/3 the contribution of motor vehicles.

As explained in the notes to Table 16-20, the EPA estimated exposure to transportation pollution in general, but not to motor-vehicle pollution specifically. We have disaggregated their general transportation category into highway and off-highway sources, under two different assumptions regarding the share of highway sources. The last two columns of Table 16-20 show the normalized exposure or dispersion estimates under the two different assumptions. We believe that the results of the higher motor-vehicle-exposure scenario ("highway @ 300") are the more accurate.

In general, the estimates of $\mathrm{DN_{PM,i}}$ derived from the EPA modeling (Table 16-20) are broadly similar to our own estimates of $\mathrm{DN_{PM,i}}$ in Tables 16-19a and 16-19b. (Note that the EPA's category "fuel combustion" comprises our three fuel-combustion categories, and that the EPA category "manufacturing" comprises our categories "chemicals..." and "solvents...") We estimate somewhat lower $\mathrm{DN_{PM}}$ from fuel combustion and manufacturing, and a considerably lower $\mathrm{DN_{PM}}$ from construction, than does the EPA. We expect that we have underestimated the $\mathrm{DN_{PM}}$ for electric utilities, but that the EPA has overestimated the $\mathrm{DN_{PM}}$ for construction. We do not believe that exposure to construction dust is substantially greater than exposure to motor-vehicle PM: in major metropolitan areas, in which most exposure occurs, people must be exposed at least as much to motor vehicles as to construction.

The results of Tables 16-19 and 16-20 also can be compared with the results of Cass and Gray's (1995) analysis of the contribution of diesel engines to particulate air pollution in Los Angeles, shown in Table 16-21. Their results are consistent with ours, except that they estimate a higher DN_{PM} for railroads.

65

²⁸There is a minor difference between the particulate-matter normalized-dispersion term (DN) derived from the EPA (1994b) modeling and our own: theirs pertains to secondary (indirect) as well as primary (direct) particulate matter, whereas as ours pertains to primary (direct) PM only. (We account for the formation of secondary particulate matter after all of the precursors have been transported to the site of the air-quality monitor.)

16.3.5 Long-range transport

As mentioned in section 16.1, our analysis ignores the transport of pollution from one AQCR to another. Of course, we know that in some regions, emissions from one AQCR can have a significant effect on ambient air quality in another. Indeed, in many parts of the U.S., especially in the east, local air quality is significantly affected by pollutants that have been carried hundreds of miles by the prevailing winds, across many counties. But we are not aware of any systematic body of work that quantifies the contribution and sources of long-range pollution's effect on different areas in the U.S., and hence do not formally model long-range transport of pollution.

16.4 ATMOSPHERIC CHEMISTRY: THE CONTRIBUTION OF MOTOR VEHICLES TO OZONE

16.4.1 Background

Ozone is not emitted as such by motor vehicles or any other source, but rather forms in the atmosphere from a series of photochemical reactions that involve NO_X , VOCs, and other compounds. The reaction rate and equilibrium depends on the relative abundance of the reactants, temperature, atmospheric mixing, and other factors (National Research Council, 1991). The reactions are complex and highly nonlinear, and there is no simple, universal formula for determining the marginal contribution of each emission source or each precursor pollutant to ozone.

The National Research Council (1991) provides a good summary of the ozone formation process. First, reactive organic compounds (RH), emitted from a variety of sources, react with hydroxyl radicals (OH) to form organic radicals (R):

$$RH + OH --> R + H_2O$$
 O1

The organic radicals combine with oxygen in the presence of an inert third body M to form peroxy radicals (RO₂):

$$R \cdot + O_2 - M - > RO_2 \cdot$$

The peroxy radicals react with nitric oxide, which is emitted from combustion and other sources, to form nitrogen dioxide:

$$RO_{2}$$
 + NO_{--} > NO_{2} + RO_{-} O3

Nitrogen dioxide is photo-dissociated by high-energy solar radiation (hv, Planck's constant h multiplied by frequency v):

$$NO_2 + hv --> NO + O$$

The oxygen atoms then combine with molecular oxygen to form ozone (O₃):

$$O + O_2 - M - > O_3$$

The ozone can be photo-dissociated back to O and O₂, and the single oxygen O can react with water vapor to form two hydroxyl radicals (OH).

In this simplified representation of the chemistry, there are two main precursor pollutant emissions: reactive hydrocarbons, which also are called volatile organic compounds (VOCs), and nitrogen oxides (NO and NO2). As one can infer from even the simplified chemistry, the relationship between emissions of VOCs and NO_X and ozone formation is highly nonlinear.

The most accurate way to estimate the contribution of each precursor or set of precursors to ozone -- i.e., to estimate $C_{p'-p}$ in equations 6 and 7 above -- is to run photochemical grid models with and without the precursor emissions from specific sources and estimate the change in the ozone level. But obviously this is very costly to do for the entire U.S. In the following sections, we briefly discuss three simpler but less accurate ways to estimate $C_{p'-p}$: ozone isopleths, statistical models, and simple nonlinear formulas. In the end, we assume a simple nonlinear relationship between VOCs (weighted according to their ozone-formation potential), NO_X , and ozone (the third method). Although this is a crude basis for apportioning ozone damages, especially given the sophistication of regional ozone modeling, it almost certainly is not likely to be so much in error as to have a significant effect on our results.

16.4.2 Alternative simple methods for estimating the contribution of precursors to ozone formation

<u>Ozone Isopleths</u>.. Rather than run an air quality model for each region, we could estimate average ambient VOC and NO_X levels in each region and use a regional ozone isopleth (which relates ozone levels to NO_X and VOC levels) to determine the relative contribution of VOCs and NO_X (from motor-vehicles and other sources) to the formation of ozone.

Unfortunately, there are serious difficulties with this method. Ozone isopleths, and positions on ozone isopleths, vary from city, but are available for only a few cities in the U.S. We would have to make up isopleths for the majority of cities. And in every city, we would have to estimate the relationship between ambient NO_X and VOC and emissions of NO_X and VOC. As a result, this method certainly is more difficult but not necessarily much more accurate than the method that we adopt, below.

<u>Statistical models (backcasting).</u> Rather than estimate contributions to ozone on the basis of region-specific data, one could estimate a universal, statistical relationship between ozone and some of the variables that determine it, such as ambient VOC, ambient NO_X , temperature and sunlight. One then would apportion ozone damages to

VOCs and NO_X on the basis of the coefficients on NO_X and VOC emissions in the regression equation. This method has been referred to as "backcasting".

However, there are several difficulties with this approach: i) it is data intensive; ii) it is unclear how to group spatially and temporally the data from air quality monitors, because the spatial and temporal relationship between ozone and ozone precursors is complex; iii) the estimated coefficients would be valid only over the range of conditions used in the estimation, and hence might not apply to large reductions in ozone. We do not believe that this can be done is such a way as to have the extra accuracy justify the considerable extra analytical effort.

<u>Simple nonlinear relationship (method adopted here)</u>. The simplest way to model the nonlinear ozone formation process is to assume a universal nonlinear relationship between ozone levels and VOC and NO_X emissions:

$$Oxidant = k \cdot (Hydrocarbons)^A \cdot (NO_x)^B$$
 (11)

This form has been used by others. For example, Schwing et al. (1980) used the following equation, taken from Merz et al. (1972), to estimate ozone formation in Los Angeles:

$$Oxidant = k \cdot (Hydrocarbons)^{0.15} \cdot (NO_x)^{0.54}$$

Schwing et al. (1980) assumed that the estimated functional form stayed constant over all pollution levels and that it was generalizable to the rest of the cities in the U.S.

We will use equation 11 to estimate the contribution to ambient ozone of VOC and NO_X emissions from motor vehicles and other sources. Like Schwing et al. (1980), we will use a single equation for all regions and conditions in the U.S. However, we doubt that Schwing et al.'s (1980) estimates of the exponents A and B (0.15 and 0.40), developed many years ago for Los Angeles, apply to all cities in the U.S. today.

But how then to estimate the exponents A and B, which in effect weight the contribution of VOCs and NO_X to ozone? It of course is difficult to generalize about the relative importance of VOCs and NO_X emissions in ozone formation. As the NRC (1991) notes, the sensitivity of ozone levels to VOC and NO_X emissions varies from one region to another. Still, we can define some reasonable bounds for the exponents A and B in our ozone formation equation 11.

First, it is likely that, on average, ozone levels are slightly more sensitive to VOC emissions than to NO_X emissions. This is because in some cases a decrease in NO_X emissions will cause a significant increase in ozone, whereas a decrease in VOCs never will cause a significant increase in ozone (NRC, 1991). This suggests that A>B.

However, it is quite clear that ozone levels do not depend on VOC emission alone²⁹. As the NRC notes, "there are many areas where control of VOCs is either ineffective or does not bring an area into compliance...hence NO_X control probably will be necessary in addition or instead of VOC control.." (p. 377). Thus, we know that $B \neq 0$.

Third, on the basis of the information reviewed next, it appears that ozone sensitivity to VOC or NO_X emissions -- defined formally as the percent change in ozone divided by the percent change in emissions of VOCs or NO_X -- typically ranges between 0.2 and 0.7. Chang et al. (1989) used the EPA's ozone trajectory model, EKMA (Empirical Kinetic Modeling Approach) to estimate the sensitivity of ozone levels to changes in VOC emissions from light duty vehicles. In 20 cities in Ohio, Georgia, Massachusetts, North Carolina, Texas, Indiana, Tennessee, Florida, Pennsylvania, Maine, Virginia, Missouri, and Washington, D. C., the ozone sensitivity (the percent change in ozone concentration divided by the percent change in light-duty VOC emissions) ranged from 0.43 to 1.45, and in most cities was between 0.45 and 0.65. The average value was 0.62. The National Research Council (1991) reports that another study obtained similar results.

In some areas of the country, ozone is more sensitive to NO_X emissions than to VOC emissions. The NRC (1991) summarizes estimates from the Regional Oxidant Model (ROM; a 3-dimensional airshed model), of the response of ozone in the Northeast to VOC and NO_X controls. In Washington, Philadelphia, Rhode Island, Boston, Pittsburgh, and Detroit, the modeled ozone sensitivity to NO_X exceeded the modeled sensitivity to VOCs. The sensitivity to VOCs ranged from 0.09 to 0.63, with an average of 0.21. The sensitivity to NO_X ranged from -0.17 (i.e., a decrease in NO_X emissions increased ozone levels, in New York) to 0.43, with an average of 0.25 (including the negative value).

On the basis of these considerations, we choose A = 0.55, and B = 0.40, which results in an ozone sensitivity to VOC of around 0.6, and an ozone sensitivity to NO_X of slightly less. The final form of our equation is therefore:

$$Ozone = (VOCs)^{0.55} \cdot (NO_x)^{0.40}$$

Table 16-22 shows the ozone sensitivities to VOC and to NO_X predicted by this equation for different amounts of emissions reductions. As shown in the notes to Table 16-22, the ozone sensitivity, given an ozone-formation equation of the form of equation 11, is a function only of the percentage reduction in precursor emissions; it is independent of the units or magnitude of emissions. As shown in Table 16-22, the

²⁹We raise this possibility for three reasons. First, it is appealing because it is so simple: one assumes that ozone damages simply are proportional to VOC emissions. Second, until recently, most air-quality planners aimed to reduce ozone levels exclusively by reducing VOC levels. Third, others, such as the Office of Technology Assessment (1989), doing analyses somewhat like ours, have assumed that ozone levels are proportional to VOC emissions.

estimated sensitivity to VOC is broadly consistent with the modeling results discussed above.

<u>Reactivity-weighted VOC emissions</u>. In the "first" step of the ozone formation process, emissions of VOCs react with the hydroxyl radical (equation O1 above). The rate of this reaction depends on the specific type of organic compound involved: some compounds, such as methane, react relatively slowly; others, such as some alkenes, react quite rapidly -- about two orders of magnitude more rapidly than does methane. Beyond this, the rates and equilibria of other reactions in the atmospheric chemistry of ozone also are determined by the specific mix of organic compounds involved. Thus, overall, the amount of ozone formed from VOC and NO_X emissions depends very much on the specific mix of individual organic compounds within the broad class "VOCs" (NRC, 1991).

Different emission-source categories emit very different mixes of organic compounds. For example, motor vehicles emit lots of relatively reactive alkenes, whereas natural-gas pipelines leak mainly unreactive alkanes. Because the mix of VOC emissions varies from source to source, and the ozone-creation potential of different VOC mixes varies widely, it is important to account for the different ozone-creation potential of different emission source categories. We will do that in this section.

In order to estimate the ozone-creation potential of different VOC-emission sources, one must: a) define a measure of ozone-creation potential; b) estimate the ozone formation potential of individual organic compounds; and c) estimate emissions of individual organic compounds from each source category. This can be a tall order, but fortunately for us, Derwent et al. (1996) have essentially done this already. They estimated the photochemical ozone-creation potential (POCP) of a large number of reactive hydrocarbons, under European conditions; estimated emissions of individual VOCs in each source category in the United Kingdom's emissions inventory; and then multiplied emissions of each compound by its POCP and summed over all VOC emissions within a source category, to produce an overall POCP-weighted VOC emission for each of the source category in the United Kingdom's emissions inventory.

The ratio of POCP-weighted VOC emissions to unweighted VOC emissions gives a POCP adjustment factor for each source category, which we can apply to raw or "unweighted" VOC emissions in the U. S. emissions inventory. (Derwent et al., 1996, refer to the adjustment factor as the "sector mean POCP"). Thus, formally, we estimate reactivity-weighted, or POCP-weighted VOC emissions:

$$VOC_{i-POCP} = VOC_{i} \cdot POCP_{i}$$

$$\sum_{c} VOC_{UK-i-c} \cdot POCP_{c}$$

$$VOC_{UK-i}$$
(12)

where:

VOC_{i-POCP} = VOC emissions from source i in the U. S., weighted according to ozone-creation potential

 VOC_i = VOC emissions from source i in the U. S.

POCP_i = adjustment factor to account for differences in photochemical ozonecreation potential (Derwent et al., 1996; Table 16-23 here).

VOC_{UK-i-C}photochemical ozone-creation potential of organic compound C under European conditions

 VOC_{UK-i} = Emissions of VOCs from source S in the U. K.

The results of the Derwent et al. (1996) analysis, which we use here, are summarized in Table 16-23. We assume that POCPs estimated for European conditions are similar to POCPs for U. S. conditions, and that the mix of VOCs in each source category in the U. K. inventory is similar to the mix in the corresponding category in the U. S. emissions inventory. In our analysis of ozone pollution, whenever we refer to VOC pollution, we mean POCP-weighted, or reactivity-weighted, VOC emissions as estimated by equation 12.

<u>Estimating ozone on the basis of our nonlinear ozone equation.</u> Recall from the introduction to this report (Section 16.1) that our objective is to estimate ozone levels after a hypothetical change in emissions of ozone precursors. Specifically, we will use equation 11 to model three pollution-reduction scenarios: I) eliminate all anthropogenic pollution; and II) eliminate 10% (IIA) and 100% (IIB) of motor-vehicle related pollution. These scenarios will be used in the analysis of the health costs of pollution (Report #11), and the analysis of the agricultural cost of pollution (Report #12).

In all cases, the initial or baseline level is taken to be actual measured ambient levels (in 1988, 1989, 1990, or 1991). These data, from ambient air-quality monitors, are discussed in Reports 11 and 12. What remains to be done, here, is to estimate what ozone levels would be were all anthropogenic pollution (case I) or 10% (case IIA) or 100% (case IIB) of motor-vehicle-related pollution eliminated.

Equation 11 expresses the relationship between the precursor pollutants P' (VOCs and NO_X) and the ambient pollutant P (ozone); put another way, it is the explicit functional form of the general functional relationship $PI_{p',C}^* = C_{p'-p}$ (E...) expressed by 6. Formally, applying equation 11 in equation 1b, we have, for case IIB (elimination of motor-vehicle-related pollutants; the other cases are analogous):

$$PP = PI \cdot \frac{PP *}{PI *} \rightarrow$$

$$Ozone(DF)_{NO - MVs} = Ozone(DF)_{total - A} \cdot \frac{Ozone(EQ11*)_{NO - MVs}}{Ozone(EQ11*)_{total}} =$$

$$Ozone(DF)_{total - A} \cdot \frac{\left(VOC(EQ6*)_{NO - MVs}\right)^{A} \cdot \left(NO_{x}(EQ6*)_{NO - MVs}\right)^{B}}{\left(VOC(EQ6*)_{total}\right)^{A} \cdot \left(NO_{x}(EQ6*)_{total}\right)^{B}}$$
(13)

where:

- PP = the estimated actual pollution level, without motor-vehicle-related emissions
- PI = the actual total ambient pollution level (data from air-quality monitors; discussed in Reports 11, 12, and 13)
- PP* = the modeled level of pollution, without motor-vehicle related emissions PI* = the modeled level of total ambient pollution
- Ozone(DF) $_{NO-MVs}$ = the estimated ambient level of ozone after motor-vehicle-related pollution is eliminated; an input in the ozone damage functions (DF)
- Ozone(DF) $_{total-A}$ = the measured ambient level of ozone (from ambient airquality data; see Reports #11 and 12)
- Ozone(EQ11*) $_{
 m NO-MVs}$ = the level of ozone after motor-vehicle--related pollution is eliminated, modeled by equation, 11
- Ozone(EQ11*) $_{total}$ = total ozone modeled by equation 11
- VOC(EQ6*)_{NO-MVs} = the ambient level of reactivity-weighted VOC pollution after motor-vehicle-related VOC emissions are eliminated, modeled by equation 6
- $NO_X(EQ6^*)_{NO-MVs}$ = the ambient level of NO_X pollution after motor-vehiclerelated VOC emissions are eliminated, modeled by equation 6
- VOC(EQ6*)_{total} = the ambient level of total reactivity-weighted VOC pollution (anthropogenic plus biogenic), modeled by equation 6
- $NO_X(EQ6^*)_{total}$ = the ambient level of total NO_X pollution (anthropogenic plus biogenic), modeled by equation 6.

In all cases, "pollution" (e.g., "all anthropogenic NO_X pollution") refers to official emissions [OEI in equation 6] multiplied by our emissions-correction factor [EC $_{p',i}$ in equation 6] multiplied by the normalized dispersion term [DN $_{p',i}$ in equation 6], and, in the case of VOCs, multiplied by the POCP adjustment factor (Table 16-23).

Note that the results of this equation are independent of the scale of the units of VOC and NO_X .

We emphasize that, for case II (10% or 100% of motor-vehicle emissions removed), there is no coherent alternative to estimating the incremental contribution of motor vehicles to ozone, as we do here. For example, suppose that one estimated total anthropogenic ozone, and then apportioned a part of this total to motor vehicles on the basis of some weighting of emissions of precursors from motor-vehicles. What exactly would this apportioning tell us? What specific scenario would this apportioning correspond, and how would we interpret the results? The results of this "average" analysis would not tell us the effect of eliminating motor-vehicle emissions first, or last, or anywhere in between (except fortuitously), because we would not have estimated

those specific scenarios. And it will not do to answer that such an "average" tells us the contribution of motor vehicles as part of a program to eliminate all pollution, because if the program is to eliminate all pollution, then we can speak only of the effects of eliminating all pollution, and nothing more³⁰.

The point, in short, is that because ozone is nonlinear in formation, one must model specific scenarios.

The incremental contribution of specific precursor emissions. Because regulators control individual pollutants, and because damage estimates often are expressed per ton of pollutant emitted, it will be useful to estimate the incremental contribution to ozone of VOC and NO_X emissions. Again, though, this can be done only for specific increments; there is no meaningful "average" *individual* contributions of VOC and NO_X to ozone, because these pollutants jointly produce ozone. For example, given an estimate of the total ozone damages due to all motor vehicle pollution, such as we make here, there is no way to estimate the separate effects of the precursors, because *ipso facto* we have estimated the joint effect of all of the precursors. We may assign the total ozone cost to VOCs and NO_X combined, and estimate a \$-ozone cost per ton of VOCs+ NO_X , but this \$/combined-kg cost is valid only for the specific scenario estimated -- for the specific quantities of VOCs and NO_X involved.

Of course, one can use equation 11 or 13, or a sophisticated model, to estimate the effects of changing only VOCs or only NO_X , but the resultant \$ or \$/kg damages cannot apply to any scenario of jointly changed emissions. Also, one can derive from equation 11 the rate of change of ozone with respect to a change in VOCs, and with respect to a change in NO_X , as a function of VOC and NO_X pollution:

$$\frac{\partial O_3}{\partial VOC} = A \cdot VOC^{A-1} \cdot NO_x^B$$

$$\frac{\partial O_3}{\partial NO_x} = VOC^A \cdot B \cdot NO_x^{B-1}$$

where:

VOC = the level of VOC pollution at which the rate of change of ozone is calculated

 $^{^{30}}$ Suppose, as a further example, that we reduce NO_X and VOCs one at a time in small, equal-percentage increments, estimate the change in ozone and health effects at each step, and continue until motor-vehicle NO_X and VOC emissions are eliminated. Can we add up the changes estimated at each step, and call the sum for each precursor the share of the total attributable to each precursor? No. The shares thusly determined will not represent what will happen if we eliminate all VOC or all NO_X emissions all at once, and will not tell us what will happen if we make marginal changes. The only thing we can do is add the shares together and state that the total is what you get if you eliminate both precursors -- and that, of course, obviates the whole exercise of determining separate "average" contributions.

 NO_X = the level of NO_X pollution at which the rate of change of ozone w is calculated

A = exponent A from equation 11

B =exponent Bfrom equation 11.

From these two expressions, one can derive a simple but useful metric, the ratio of the ozone-VOC sensitivity to the ozone-NO $_X$ sensitivity:

$$\frac{\frac{\partial O_3}{\partial VOC_{MV}}}{\frac{\partial O_3}{\partial NO_{x_{MV}}}} = \frac{A \cdot VOC^{A-1} \cdot NO_x^B}{VOC^A \cdot B \cdot NO_x^{B-1}} = \frac{A}{B} \cdot \frac{NO_x}{VOC}$$

Note that "pollution" here always means official emissions multiplied by the emissions correction factors multiplied by normalized dispersion -- $OEI_{p',i} \cdot EC_{p',i} \cdot DN_{p',i}$, from equation 6 - and, in the case of VOCs, multiplied by the ozone-creation potential, POCP.

The incremental contribution of specific vehicle types or emissions sources. As written above, equations 11 and 13 model the effect of eliminating all motor-vehicle related emissions, but obviously they can be applied easily to estimate the effect of eliminating only direct motor-vehicle emissions, or only indirect motor-vehicle emissions, or only direct emissions from a particular class of motor vehicles. Essentially, all one has to do is change the parameter MSp', i in equation 6 to represent whatever incremental emissions source one wants to model. If one wishes to estimate the effect of eliminating only direct emissions from light-duty vehicles, then MSp, i is the fraction of total emissions, in each source category , that is direct emissions from light-duty vehicles. In this case, MSp', i will be zero for every emission source category except light-duty vehicles, for which it will be 1.0.

In our own presentation of results (in Reports 11, 12, and 13), we estimate ozone damages attributable to each of six individual vehicle classes, to each of two aggregated vehicle classes (gasoline vehicles, and diesel vehicles), and to indirect motor-vehicle related sources. Note, though, that because equation 13 is nonlinear, the sum of damages estimated for each vehicle class considered separately will not equal the sum of damages from all vehicles considered at once.

16.5 ATMOSPHERIC CHEMISTRY: THE FORMATION OF SECONDARY SULFATE AND NITRATE PARTICULATES FROM EMISSIONS OF NOX, SO₂, AND NH₃

16.5.1 Background

Emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_X), and ammonia (NH_3) interact with water vapor, hydrocarbons, dust, and other carbons to form particles of ammonium sulfate and ammonium nitrate. Because this "secondary" particulate matter can constitute a sizable fraction of the total ambient particulate matter measured at air quality monitors (Tables 16-9 and 16-10), and because emissions of the precursors can vary substantially from source to source, it is important to have at least a simple model of the formation of secondary particulate matter from emissions of SO_2 , NO_X , and NH_3 .

Unfortunately, but not surprisingly, it is not easy to model secondary aerosol chemistry. According to Herrick and Kulp (1987):

Reactions of the precursors with other chemical entities, often formed from photolysis, begin immediately on emission, and depending on emission rate, weather, and air concentration of all reactants may proceed at different rates. Some reactions will take place in minutes, others in days. In the meantime, the pollutants and their products are being transported, diluted, deposited, and augmented by new emissions along their path. (p. I-4).

Nevertheless, in the following sections, we will develop a simple model of the formation of secondary sulfate and nitrate particles from emissions of NO_X , SO_2 , and NH_3 . We ignore the effects of weather, relative concentrations, and emission rates. As discussed above, we assume that all precursors first disperse from the source to the receptor site (the site of the air-quality monitor), and there undergo simple chemical reactions. We consider as precursors only SO_2 , NO_X , and NH_3 ; we do include dust, water vapor, or other compounds.

In each case, we first consider the general chemistry of the formation of the secondary particulates from the precursor emissions. Given this general background, we then analyze the relationship between emissions of the precursors, and formation of secondary particulate compounds. Our goal is to develop simple formulas that predict secondary particulates given only emissions of precursors.

16.5.2 Formation of ammonium sulfate from SO2 and NH3 emissions

General chemistry

The conversion of sulfur in fuel to sulfur in particulate sulfates proceeds in several steps (Eatough et al., 1994; Watson et al., 1994a; McHenry and Dennis, 1994; Pilinis and Farber, 1991; Herrick and Kulp, 1987). First, sulfur in fuel, which is the main source of anthropogenic sulfur in the atmosphere, is burned with air to sulfur dioxide:

$$S [fuel] + O_2 [air] \longrightarrow SO_2$$

The resulting sulfur dioxide is converted to sulfuric acid via gas-phase and aqueous-phase reactions. In the dominant gas-phase reaction, sulfur dioxide reacts with the hydroxyl radicals in the atmosphere to form hydrogen sulfite, which then reacts

quickly with oxygen and small amounts of water vapor to become sulfuric acid (Watson et al., 1994a):

$$SO_2 + OH + H_2O + O_2 --> H_2SO_4 + HO_2$$
 (gas phase) S2

The transformation rate in this gas-phase pathway is controlled more by the concentration of hydroxyl radicals than by the concentration of sulfur dioxide, and hence is highest during the daytime, when hydroxyl radicals are produced by photochemical processes. According to Herrick and Kulp (1987), the transformation to sulfuric acid is linear in the daytime, and proceeds at about 0.5% per hour in rural air in the summertime. McHenry and Dennis (1994) report an estimate that sulfur dioxide converts to sulfate in the gas phase at the rate of about 5% per hour, and Eatough et al. (1994) calculate a rate of 5.5% from previous studies. Watson et al. (1994a) cite a range of 0.01% to 5%/hour, and Eatough et al. (1994) cite a range of less than 1% to 10% per hour, the latter occurring at high temperature and humidity. The EPA (1994b) assumes a rate of 0.2+0.02P per hour, where P is the annual precipitation rate, in inches.

In this gas-phase pathway, the sulfuric acid produced is a gas initially. However, sulfuric acid has a low vapor pressure, and hence readily forms sulfuric acid droplets, or condenses on existing particles, such as dust particles.

In the aqueous phase, in clouds or fog, sulfur dioxide can be dissolved in water droplets, and then react very quickly with any hydrogen peroxide dissolved in the droplet:

This aqueous reaction rate is controlled by the solubility of the precursor gases, and generally is 10 to 100 times higher than the gas-phase reaction rate (Watson et al., 1994a; Eatough et al., 1994).

At this point we have droplets of liquid sulfuric acid, or sulfuric acid condensed on particles such as dust, or sulfuric acid dissolved in water droplets. The dissolved sulfuric acid can be neutralized by any ammonia that also is dissolved in the water droplet, and the condensed sulfuric acid can be neutralized by ammonia that reacts on the surface of the particle. Depending on the amount of ammonia, the sulfuric acid can be partly neutralized to ammonium bisulfate, or fully neutralized to ammonium sulfate:

Very close to major SO₂ sources, only a relatively minor amount of sulfuric acid will have been neutralized, depending on the amount of ammonia in the immediate area. As the SO₂ and sulfuric acid disperse further from the emissions source, more of the sulfate will be neutralized to ammonium bisulfate and ammonium sulfate, as the

sulfates come into contact with more ammonia. Within a 100 km of major SO₂ sources, most of the sulfuric acid will have been at least partially neutralized to ammonium bisulfate. On a regional scale, nearly all of the sulfate will be neutralized fully by ammonia to ammonium sulfate. In general, sulfuric acid is neutralized relatively easily and hence is relatively rare (Waldman et al., 1995) and usually converted to ammonium sulfate.

Note that in the aqueous-phase pathway, the ammonium sulfate or bisulfate is produced initially within a droplet. That is, initially, the sulfate particle is a droplet with a small portion of liquid water (Watson et al., 1994a). However, as the relative humidity drops below 70%, the water evaporates and a small, solid sulfate particle remains.

Quantitative relationship between precursor emissions and secondary ammonium sulfate

We analyze the formation of secondary particulates in two steps: the formation of sulfate (as sulfuric acid) from SO₂, and the formation of ammonium sulfate or bisulfate from sulfuric acid (S4, S5).

Langner and Rodhe (1991) have developed a three-dimensional model of the global sulfur cycle. They model the conversion of sulfur in SO₂ to sulfur in sulfate under two scenarios: fast in-cloud oxidation (case I), and slow in-cloud oxidation (case II). For case I, the model estimates that, globally, 8% of sulfur in SO₂ is converted to sulfur in SO_4^{2-} by gas-phase oxidation by OH (route S2 above) and that 44% is converted by oxidation in clouds (S3 above). For case II, the respective percentages are 13% and 24%.

The Langner and Rodhe (1991) model thus predicts that 52% (case I) or 37% (case II) of sulfur in SO₂ is converted to sulfate, depending on whether aqueous-phase oxidation is relatively rapid or relatively slow. Results from the Regional Acid Deposition Model (RADM), which is "a comprehensive Eulerian model designed to incorporate known major atmospheric physical and chemical processes related to acidic deposition" (McHenry and Dennis, 1994, p. 892), suggest that Langner and Rodhe's case II is more realistic. A version of the RADM, called the Comprehensive Sulfate Tracking Model (COMSTM), predicts that in the Eastern United States, gas-phase reactions (mostly S2) contribute 36% of the total sulfate, and aqueous-phase reactions 64% (with reaction S3 above by itself contributing 50%) (McHenry and Dennis, 1994). This is a predicted ratio of gas-phase formation to aqueous-phase formation of 0.56, which is very close to the ratio of 0.54 predicted in case II in Langner and Rodhe (1991). Thus, the Langner and Rodhe (1991) work indicates that 37% of sulfur in SO₂ becomes sulfate.

Dastoor and Pudykiewicz (1996) also have developed a global meteorological sulfur transport model. Their model includes cloud processes, dry and aqueous-phase chemical processes for sulfur, dry deposition, and the precipitation scavenging of sulfur. The model simulates that, in the regions where the sulfur is emitted, the surface concentration (in ng/m^3) of SO₂ is about 7 times the concentration of SO₄, which implies that about 10% of the emitted SO₂ has been converted to SO₄, relatively

quickly. However, the oxidation of SO_2 continues as the emissions are transported away from the source regions, so that by the time the sulfur reaches the arctic, the simulated surface concentration (in ng/m^3) of SO_2 is 2-2.5 times the simulated concentration of SO_4 . Thus, far from the source, 20-25% of the emitted SO_2 has been converted to SO_4 .

Other studies have found or assumed conversion percentages on the order of 20%. Lioy and Waldman (1989) report that in the U. S. in 1979 and 1978, the ratio of sulfur in SO_4^{2-} to sulfur in $SO_2 + SO_4^{2-}$ varied from 5 to 50%, and generally was between 10% and 30%. Altshuler (1984) reports that at six sites in Saint Louis in 1976, sulfur in particulates was 19% of total sulfur measured. Given that some particulate sulfates are emitted directly, the implied secondary rate of conversion of sulfur in SO_2 to sulfur in particulate sulfate is less than 19%. In an earlier study, Altshuler estimated the following linear relationship between SO_4 concentration and SO_2 concentration: $SO_4 = 4.92 + 0.144 \cdot SO_2$ [r = 0.82], valid up to $SO_2 = 80 \, \mu g/m^3$, above which level SO_4 was independent of SO_2 (reported in Barnes et al., 1983). Assuming that the units of concentration in this equation are ppm, the equation implies that 14.4% of sulfur in SO_2 becomes sulfur in SO_4 .

In its 1990 interim emissions inventory, the EPA (1995d) assumes that 10% of the sulfur in SO₂ converts to sulfur in ammonium sulfate. In its documentation to the particulate emission-factor model, PART5, the EPA (1995c) states that nationally, 12% of the sulfur in SO₂ converts to sulfur in ammonium sulfate.

The studies reviewed above indicate that 10% to 40% of sulfur in SO₂ is converted to sulfur in sulfate. Most likely, the conversion percentage is higher when aqueous-phase conversion, which is much faster than gas-phase conversion, is the main route. The extent of aqueous-phase conversion, in turn, depends in large part on the relative humidity and cloud cover. Because the eastern U. S. is cloudier and more humid than the western U.S., we expect more sulfate to form via aqueous-phase chemistry -- and hence more sulfur to be converted to sulfate -- in the east than in the west. There is some evidence in support of this. Burton et al. report that 65% of PM_{2.5} (50% of PM₁₀) in Philadelphia is particulate sulfate, and Eatough et al. (1994) remark that particulate sulfate contributes up to half of fine particulate matter in the eastern U.S. throughout the year -- a contribution much higher than reported in the CMB studies done in the west (most of the studies of Table 16-9). The few CMB source-apportionment studies that have been done in the East (Illinois, Ohio and Philadelphia) have indeed found a relatively large share for sulfate particulate (Table 16-9).

On the basis of these studies, we assume that in the Western U. S., 25% to 15% (low-cost to high-cost)³¹ of the sulfur in SO₂ is converted to sulfur in SO₄²⁻, and that in the Eastern U. S., 35% to 25% (low-cost to high-cost) is converted³².

Next, we assume that the sulfate is neutralized by NH3 emissions. Ammonia reacts with sulfate before it reacts with nitrate. Watson et al. (1994a) report that significant amounts of ammonium nitrate form only when there are twice as many moles of ammonia as sulfate; i.e., that reactions N9 or N10 do not go until reaction S5 is completed.

Thus, we assume that first, sulfate is neutralized to ammonium bisulfate by NH3 emitted within the air basin (actually, the AQCR). If there is more than enough NH3 to convert all of the sulfate to ammonium bisulfate, then we assume that bisulfate is further neutralized to ammonium sulfate, NH3 permitting. If there is more than enough NH3 to fully neutralize the ammonium bisulfate to ammonium sulfate, we assume that the NH3 then begins to neutralize nitrates.

The remainder of the sulfur in SO₂ -- i.e., the sulfur that does not convert to sulfate -- precipitates in water ("wet deposition") or settles out or deposits on surfaces ("dry deposition). Languer and Rodhe (1991) estimate that 32% (case I) or 38% (case II) of sulfur in SO₂ deposits as a gas, and that 15% (case I) or 25% (case II) precipitates in water. Hegg (1985) states that "an appreciable fraction" of the sulfur emissions are

 $^{^{31}}$ We experimented with different low and high SO_X -conversion percentages, and found that the higher the SO_X conversion percentage, the lower the particulate damage costs attributable to motor vehicles. (Keep in mind that the ultimate purpose of this conversion percentage is to allocate ambient particulate concentrations to different emission sources.) The higher the conversion percentage, the greater the share of ambient ammonium sulfate (from all sources) out of all ambient particulate matter. The greater the ammonium-sulfate share, the lower the share of primary particulate matter from any source, including direct particulate matter from motor vehicles. Thus, a high SO_X conversion percentage downweights the contribution of direct motor-vehicle emissions, and so tends to reduce damages due to motor vehicles. Now, at the same time, a high conversion percentage also increases the share of motor-vehicle related SO_X emissions. However, this increase is relatively minor, because motor-vehicles are a minor source of sulfur emissions. Thus, in the end, the higher SO_X conversion percentage increases the share of motor-vehicle- SO_X emissions (a minor effect), increases the share of non-motor-vehicle SO_X emissions (a major effect), and decreases the share of motor-vehicle and other direct PM emissions (a major effect), with the net effect being a decrease in the contribution of direct and indirect (primary and secondary) motor-vehicle particulate matter.

³²The true relationship between sulfur emissions and sulfate formation might be nonlinear. In support of this, Herrick and Kulp (1987) note that a "reduction in the emissions of sulfur dioxide in the northeastern quadrant of the United States in winter is unlikely to result in proportional decrease in the formation and subsequent deposition of sulfuric acid over the northeastern United States" (p. I-8). Barnes et al. (1983) make similar observations. However, the aerosol trajectory model (ATM) used by Pilinis and Farber (1991) predicts that in the South Coast Air Basin, if sulfate is produced only in the gas phase by oxidation of SO₂, then sulfate levels decrease linearly with SO₂ emissions.

deposited. We will assume that this deposited SO₂ does not become a liquid or solid particle that can be measured by an air-quality monitor.

Finally, for simplicity, we assume that the sulfate particles measured at the ambient air-quality monitors are pure ammonium sulfate, with no water or organic matter. To the extent that measured ambient sulfates do include water or dust material, we will have underestimated the mass of secondary particulate matter formed. In this respect, we note that Dzubay et al. (1988) found that the weight fraction of sulfur in ambient sulfates was less than the weight fraction of sulfur in pure ammonium sulfate, which implies that water and possibly organic matter are retained on the particle during laboratory analysis³³.

16.5.3 Formation of ammonium nitrate from NO_X and NH_3 emissions General chemistry

The conversion of di-nitrogen in the air to nitrogen in particulate nitrates also proceeds in several steps (Watson et al., 1994a; Zhang et al., 1994; NRC, 1991). First, nitrogen oxide is formed from combustion:

$$N_2 [air] + O_2 [air] --> 2NO$$
 N1

The nitrogen oxide is oxidized to NO₂, NO₃, and N₂O₅, and other species:

Beyond this, we ignore any water that is inherent in liquid aerosols, even though this water probably is (and certainly should be) counted in the actual measurements.

³³Ideally, we would model the formation of particulates as they actually end up being measured at airquality monitors. This, however, is impossible, because nobody knows *exactly* what the air quality monitors measure.

Appel (1993a) describes the official procedure for sampling particulate air pollution: "PM-10 samples for monitoring compliance with national ambient air quality standards are routinely collected on 8" x 10" quartz filters with high-volume samplers equipped with PM-10 inlets. They are weighed before and after sample collection to the nearest mg or 0.1 mg...Following sampling, the weight increase is used to calculate the suspended particle mass concentration...The procedures for PM-10 mass measurement specify equilibration at 20-45 ± 5% RH [relative humidity] at 15-30° C for 24 h, both before and after filter loading" (p. 238 and p. 239; brackets added). If this process were perfect, it would measure every liquid or solid particle except water vapor suspended in the atmosphere. But of course, the process is not perfect: some particles that should not be counted are, and some that should be counted are not. For example, quartz filters can retain gaseous nitric acid, which can form particulate nitrate in the filter and thus end up being counted as particulate matter. But this "artifactual" particulate nitrate, formed from a gas, should not be counted. Other filters can retain gaseous SO2, and form artifactual sulfates. Cellulose filters and hygroscopic particles themselves can absorb water vapor, and thereby confound accurate measurement of the ambient particle mass (Appel, 1993a; Lee and Ramamurthi, 1993). Conversely, semivolatile nitrate and organic particulates, which coexist in the condensed phase and the gas phase, can volatilize during collection (Appel, 1993a), and so not be counted properly. The nature and extent of these errors depends on the type of filter and lab protocol, which can vary widely. Consequently, we cannot possibly model particulates as they actually are measured.

These oxides of nitrogen are converted to nitric acid via two principle pathways, one dominant during the day, the other dominant at night. During the day, nitric acid is formed by reaction with the same hydroxyl radical that reacts with sulfur dioxide in the gas phase (Watson et al, 1994a; Zhang et al., 1994; Herrick and Kulp, 1987; NRC, 1991):

Herrick and Kulp (1987) state that calculated oxidation rate of NO_X to HNO3 by OH is about 8% per hour in the summer, which results in nearly complete conversion in one day (p. I-20). The EPA (1994b) assumes that nitrate forms at 2% per hour.

At night, the dominant production pathway is (Zhang et al., 1994; Herrick and Kulp, 1987; NRC, 1991):

$$N_2O_5 + H_2O --> 2HNO_3$$
 N6

In a detailed simulation of atmospheric aerosol chemistry, Zhang et al. (1994) found that N5 accounts for 96% of total nitrate formation during the day, and that N6 and gas-to-particle conversion of NO3 accounts for 80% of total nitrate formation at night. They also estimated that 30% more particulate nitrate is formed during the day than the night.

There are other nitric-acid production pathways, of relatively minor importance:

Finally, the nitric acid can be neutralized by ammonia:

Because HNO₂ photo-dissociates quickly to HO and NO (National Research Council, 1991), there probably is considerably more HNO₃ than HNO₂ in the atmosphere, and hence considerably more NH4NO₃ than NH4NO₂. However, NH4NO₃ is not especially stable itself. It exists in an equilibrium, influenced by temperature and relative humidity, with gaseous ammonia and nitric acid. Solomon et al. (1992) describe the equilibrium between HNO₃, NH4NO₃, and NH₃ in Los Angeles:

The NH₃-HNO₃-NH₄NO₃ equilibrium condition is very sensitive to temperature, with greatly increased ambient HNO₃ concentration predicted to be in the gas phase at higher ambient temperatures..Aerosol NH₄NO₃ formation also is sensitive to the absolute magnitude of concurrently observed NH₃ concentrations...the increased fine-particulate nitrate levels observed at most sites during the winter most likely result from the lower winter NH₃ levels which shift the NH₄NO₃-HNO₃-NH₃ equilibrium toward the aerosol phase (p. 1600).

They also summarize the formation, transport, and reactions of nitric acid and particulate nitrate in Los Angeles:

...the highest NO₂ concentrations accumulate near the coast in the western portion of the air basin overnight and during the early morning hours. As the day proceeds, NO and NO₂ typically are advected eastward across the air basin; NO₂ is oxidized to form nitric acid, and high nitric acid concentrations are predicted to occur in the middle portion of the air basin...As this nitric acid-laden air mass passes over the Chino dairy area...very large amounts of ammonia are injected into the atmosphere from livestock waste decomposition and from other agricultural activities...The available nitric acid reacts to form large amounts of nitrate aerosol, resulting in the extremely high aerosol nitrate concentrations and low HNO₃ levels measured farther downwind at Rubidoux (p. 1599).

Quantitative relationship between precursor emissions and secondary ammonium nitrate

We will analyze the formation of secondary particulate nitrate in two steps: the formation of nitrate (as nitric acid) from NO_X (N5 - N8), and the formation of ammonium nitrate from nitric acid (N9).

Watson et al. (1994a) use a secondary aerosol equilibrium model, SEQUILIB, to evaluate the relationship between emissions of NO $_{\rm X}$ and NH3 and the concentration of HNO3 and NH4NO3. (The SEQUILIB model also is used within the Aerosol Trajectory Model, described next.) They find that nitrate levels are proportional to emissions of NO $_{\rm X}$ but not NH3, because the former is limiting. However, the aerosol trajectory model (ATM) used by Pilinis and Farber (1991) predicts that in the South Coast Air Basin nitrate levels decrease nonlinearly with NO $_{\rm X}$ and NH3 emissions. Pilinis and Farber (1991) also note that the total aerosol (nitrate + sulfate + SOA) does not decrease linearly with decreases in all of the emissions. One reason is that when sulfates are reduced, more ammonia is available to react with nitric acid and form particulate nitrates. Also, the reduction in organic emissions reduces the formation of peroxyacetylnitrate, which again makes more nitric acid available to form particulate nitrate.

Nevertheless, we will assume that a fixed percentage of nitrogen in NO₂ is converted to nitrogen in nitric acid. To set an upper bound to this percentage, we note that reaction N5 (the day time production of nitric acid) is similar to reaction S2 (the gas-phase production of sulfuric acid) in two respects: in both, oxidation occurs via the hydroxyl radical, and the rate of oxidation is on the order of 5%/hour. This suggests that as much as 10% to 15% of the nitrogen in NO_X converts to nitrogen in nitric acid

(see the discussion above regarding the conversion of SO₂ to sulfate). To set a lower bound to this percentage, we note that the EPA (1994b) assumes that 5% of the N in NO_X is converted to N in ammonium nitrate. If all of nitrate is neutralized to ammonium nitrate, then the EPA's (1994b) assumption implies that 5% of the N in NO_X is converted to nitrate; otherwise, if some nitrate is not neutralized, then more than 5% of the N in NO_X must be converted to nitrate.

On the basis of these considerations, we assume that 5% (low-cost) to 7% (high-cost)³⁴ of the N in NO_X is converted to N in nitrate. Although somewhat higher, this assumption is not inconsistent with the EPA's (1994b) assumption that 5% of the N in NO_X is converted to N in ammonium nitrate, because not all nitric acid will be neutralized to ammonium nitrate. We assume that the higher conversion rate results in the higher motor-vehicle cost because it means that more motor-vehicle NO_X emissions are converted to particulate nitrates.

Finally, we assume that any NH3 that remains after sulfuric acid is fully neutralized is available to neutralize nitric acid to ammonium nitrate via reaction N9. In most (but not necessarily all) places, there is enough NH3 to fully neutralize the nitric acid. For example, in the Denver "brown cloud" study, there was enough ammonia to neutralize all of the nitric and sulfuric acid (Watson et al. 1988b). Lipfert et al. (1989) report that the average $\mathrm{NH_4}^+/\mathrm{SO_4}^{2^-}$ ratio "tends to remain constant over a large range in $\mathrm{SO_4}^{2^-}$ and site locations -- other factors, such as season, remaining constant -- [which] implies that the ammonia supply generally is not the limiting factor at any of the sites" (p. 1318). In the Southern California Air Quality Study, there generally was enough ammonium ion to react with all of the available nitrate and sulfate ions (Chow et al, 1994c). At San Carlos Street in San Jose, during the daytime, there was enough ammonia to neutralize all of the sulfate to NH4HSO4 and all of the nitrate, or all of the sulfate to (NH4)2SO4, and nearly all of the nitrate (Chow et al, 1995)³⁵. However, at Santa Barbara, it appears that there was enough ammonia to neutralize all of the free

 $^{^{34}}$ Footnote 29 discusses how we determined the low-cost and the high-cost conversion percentage for SO_X emissions. That discussion applies here to the NO_X conversion percentage, with one significant difference: motor-vehicles are such a large source of NO_X emissions that the higher NO_X conversion factor results in higher motor-vehicles particulate damages. Thus, in the end, the higher NO_X conversion percentage increases the share of motor-vehicle- NO_X emissions (a major effect), increases the share of non-motor-vehicle NO_X emissions (a major effect), and decreases the share of motor-vehicle and other direct PM emissions (a major effect), with the net effect being an *increase* in the contribution of direct and indirect (primary and secondary) motor-vehicle particulate matter.

 $^{^{35}}$ Chow et al. (1995) report average concentrations of NH₄ $^+$ (4.01 μg/m³), SO₄ $^=$ (2.14 μg/m³), and NO₃ $^-$ (11.78 μg/m³), which we convert to μmoles/m³ (0.223, 0.0223, 0.190).

nitrate and all of the sulfate to NH4HSO4, or about 3/4 of the sulfate to (NH4)2SO4 and none of the nitrate (Chow et al., 1996)³⁶.

16.5.4 Other contributors to secondary particulate formation

We have considered SO₂, NO_X, and NH₃ emissions only, as precursors to ammonium sulfate and ammonium nitrate only. However, there are other precursors to ammonium nitrate and ammonium sulfate. For example, Zhang et al. (1994) have found that dust particles can be an important surface for particulate nitrate formation. They used a detailed, coupled aerosol/gas-phase chemistry model to study the influence of dust on the tropospheric photochemical oxidant cycle, given dust loadings and other ambient conditions representative of East Asia, and found that 1.5-11.5 μ g/m³ of particulate nitrate formed on dust particles. These levels are consistent with concentrations measured in East Asia (Zhang et a., 1994), and, for that matter, with concentrations observed in the Western U. S. (Table 16-9). However, Zhang et al. (1994) found that under all simulation conditions, particulate nitrate levels decreased with increasing dust levels. They also found that ozone levels decreased with increasing dust levels at 500 μ g/m³ dust were about 25% lower than ozone levels with no dust. Dust particles in the range of 0.5 - 1.5 μ m were the most important.

There also are secondary particles other than ammonium nitrate, ammonium sulfate, and secondary organic aerosols (discussed below). For example, sodium chloride, from sea salt or road salt, can react with nitrate to from coarse particles of sodium nitrate (Watson et al., 1994a).

Solomon et al. (1992) mention both dust and sea salt in their discussion of the formation of coarse particulate nitrate in Los Angeles:

..the coarse-particle nitrates are largely composed of the nonvolatile reaction products of HNO3 with sea salt or soil dust, while the fine-particle nitrates consist largely of NH4NO3 which may dissociate to release HNO3 and NH3...coarse-particle formation is limited by HNO3 diffusion to an existing coarse-particle surface; coarse-particle nitrate formation is driven by the availability of HNO3 in the gas phase (p. 1600).

Ideally, one would use a detailed model of aerosol and oxidant chemistry to quantify the effects on particulate levels of eliminating motor-vehicle pollution. For example, the Aerosol Trajectory Model (Pilinis and Farber, 1991) assumes that the following constituents may occur:

gas phase: NH3, HCl, HNO3, H2O

liquid phase: H₂O, NH₄⁺, ,SO₄²⁻, HSO₄⁻, H⁺, ⁻NO₃⁻, Cl⁻, Na⁺, and H₂SO₄

³⁶Chow et al. (1996) report average concentrations of NH₄⁺ (0.8 μg/m³), SO₄⁼ (2.8 μg/m³), and free NO₃⁻ (0.83 μg/m³), which we convert to μmoles/m³ (0.0444, 0.0292, 0.0134).

solid phase: Na₂SO₄, NaHSO₄, NaCl, NaNO₃, NH₄Cl, NH₄NO₃, (NH₄)₂SO₄, NH₄HSO₄, and (NH₄)₃H(SO₄)₂.

Unfortunately, this level of detail is beyond our scope³⁷. Consequently, we ignore the role of dust, sodium chloride, water vapor, and other compounds in the formation of secondary sulfate and nitrate particles.

16.5.5 Secondary organic aerosols (SOA)

Some organic particulate matter is emitted directly from vehicles and other sources, and some is formed in the atmosphere from emissions of gaseous organic compounds and other compounds. It appears that levels of SOAs are linearly related to emissions of reactive hydrocarbons (Pilinis and Farber, 1991). In any event, the contribution of SOA to total ambient particulate levels generally is less than the contribution of secondary sulfates and nitrates (Table 16-9). To estimate emissions, we use the anthropogenic SOA emission estimates from the EPA (1995d), however, the data we obtained did not have biogenic SOA emission estimates. Fortunately, the EPA (1994a: Table II-13) provides sufficient information for us to estimate SOA emissions from our biogenic VOC emission inventory (EPA, 1995e).

The EPA (1994a) assumes that the formation of SOAs depends on the reactivity of the emissions of organic compounds. An organic compound that is more reactive is assumed to be more likely to form SOAs, and thus is given a higher "fractional aerosol coefficient," or FAC. The FAC multiplied by the mass of the organic compound released gives the mass of SOA formed. The EPA assigned FACs to each organic compound from a given source, such as "oak forest," and then multiplied each FAC by the fraction of the total VOC from that source. The product of the FAC and the fraction of total VOC were summed for all compounds from that source to give a source-specific FAC.

Table 16-25 summarizes the source-specific FACs for eight land cover types. The FACs range from 5% to 18%, and average 11%. Alternatively, using the EPA's (1994a) national summary of biogenic emissions³⁸, we find an average FAC of 12.8%, by simply dividing total biogenic SOA emissions by biogenic VOC emissions. We feel that the 12.8% estimate is better because presumably it takes into account vegetation types across the nation.

³⁸Presumably, EPA (1994a) based the national summary of biogenic SOA on county-level data. However, we were able to obtain only the national summary. We estimated county-level biogenic SOA on the basis of county-level VOC emissions (EPA, 1995e).

³⁷The development of complete aerosol models apparently lags the development of complete photochemical ozone models. According to Cass (1995), "model components have been developed that can track the transport of particles from sources, the production of low vapor pressure materials by chemical reaction in the atmosphere, growth of airborne particles by condensation and coagulation, and the dry deposition of particles at the earth's surface," and "many investigators are presently in the process of integrating descriptions of each of these steps into complete models..." (p. 767).

16.5.6 Size distribution of ammonium sulfate, ammonium nitrate, and organic aerosols

As we discuss in Report #11 of this social-cost series (see the list at the beginning of this report), we distinguish between fine particles (less than 2.5 μ m in aerodynamic diameter) and coarse particles (between 2.5 and 10 μ m in diameter) because there is some evidence that the fine particles are more dangerous. Because we make this distinction, we must estimate the fraction of secondary sulfate and secondary nitrate particles that are less than 2.5 μ m. Below, we review a number of studies of the size distribution of ammonium sulfate, ammonium nitrate, and SOAs. It appears that essentially all secondary particulate matter is PM10, and that most but not all is PM2.5.

To some extent, the size of a particle is determined by the way in which it was formed. In general, particles can form by homogeneous nucleation, accumulation, or mechanical abrasion. Homogeneous nucleation is the growth of a single compound on a nucleus or "seed" particle. This growth is relatively rapid (a few milliseconds in combustion, a few minutes in the atmosphere [Flagan, 1993]), and typically results in particles that are less than $0.1~\mu m$, and drop out of the atmosphere relatively slowly. Accumulation is the reaction of gaseous pollutants, such as sulfates and ammonia, on other particles to form secondary aerosols. In the atmosphere, particles accumulate in a few minutes to a few hours, and typically end up between $0.1~and~1.0~\mu m$ in size (Flagan, 1993). Mechanical particles are ground up pieces of minerals or organic material. They typically are larger than $1.0~\mu m$, and drop out of the atmosphere relatively rapidly. Dust comprises mechanically generated particles.

Because ammonium sulfate and ammonium nitrate form by accumulation, we might expect that most of them are between 0.1 and 1.0 μ m in size. It turns out that most sulfate particles indeed are between 0.1 and 1.0 μ m, but that nitrate particles typically are somewhat larger³⁹. Essentially all SOAs are less than 1.0 μ m. About 5% of sulfates, and at least 10% of nitrates, are larger than 2.5 μ m, which is the size threshold that we care about.

- 1). Size distributions graphed in Waldman et al. (1995) indicate that the majority of sulfates are between 0.2 and 1.0 μ m, with a minor amount between 1.0 and 2.5 μ m. (It is not clear if they sampled particles larger than 2.5 μ m, however.) They state that particles larger than 2 μ m contain wind–blown minerals but little sulfate.
- 2). Sioutas et al. (1995) cite a study that found that sulfate particles vary between 0.2 and 1.0 μ m, and that particulate sulfate, nitrate, and ammonium ions had a median size of 0.7 μ m.
- 3). Cahill and Wakabayashi (1993) show a graph in which sulfur, collected in drum impactors on the roof of Davis California in 1983, and "present largely in the form

86

³⁹Note, though, that it is difficult to measure the size and abundance of particulate nitrates, because nitric acid, particulate nitrates, and particulate carbon are "fragile" and readily dissociate, react, form, or stick during sampling and storage (Appel, 1993).

of ammonium sulfate" (p. 212), is distributed mainly between 0.05 and 2.0 μ m, with apparently less than 5% greater than 2.0 μ m.

- 4). Dzubay et al. (1988) found that 3% of the sulfate in aerosol in Philadelphia was greater than $2.5 \mu m$.
- 5). Lundgren and Burton (1995) report that 25% of sulfate aerosols, 50% of nitrate aerosols, and about 10% of organic aerosols are larger than 1.0 μ m, and that 10% of nitrate aerosols and 1-5% of organic aerosols are greater than 2.5 μ m.
- 6). Allen (1995) measured the size distribution of aerosols in Los Angeles between 0.05 μm and 4.0 μm , and reported that:
- i) most sulfate particles were between 0.1 and 1.0 μm , but at least 10% were between 2.0 and 4.0 μm ; and
- ii) virtually all ambient nitrate particles were between 0.5 and 4.0 μ m, with 50-75% between 2.0 and 4.0 μ m;
- iii) all organic aerosols (carbonyl particles, aliphatic carbon particles, and organonitrate particles) were $1.0 \mu m$ or less in size.

Note, however, that Allen (1995) did not sample above $4.0~\mu m$. It is possible that some sulfate and nitrate particles are larger than $4.0~\mu m$.

7). Appel (1993) presents a table of the following distribution between fine and coarse particles in Houston, Texas during the daytime:

	0 - 2.5 μm ng/m ³	2.5 - 15.0 μm	
		ng/m ³	fraction
SO_4^{2-}	16,700	1100	0.06
NO_3	250	1800	0.88
NH_4^+	4300	<190	< 0.04

The fraction of nitrates above 2.5 μm is unusually large; perhaps it was the result of unusually high humidity.

On the basis of the data presented above, we will assume that 5% of sulfate particulates, and 20% of nitrate particles, are larger than $2.5~\mu m$.

16.5.7 Formal model of ambient particulate levels after a change in emissions

With the foregoing, we can develop a formal model of ambient particulate levels after a change in emissions. We will show here the model for case IIB, the elimination of all motor-vehicle related pollution. The models for case I, the elimination of anthropogenic pollution, and case IIA, the elimination of 10% of motor-vehicle-related pollution, of course are analogous.

We have:

$$\begin{split} &PP = PI \cdot \frac{PP *}{PI *} \rightarrow PMX \ (DF)_{NO-MVs} = PMX \ (DF)_{total-A} \cdot \frac{PMX *_{NO-MVs}}{PMX *_{total}} \\ &= PMX \ (DF)_{total-A} \cdot \frac{PMXd *_{NO-MVs} + PMXs *_{NO-MVs}}{PMXd *_{total} + PMXs *_{total}} \\ &= PMX \ (DF)_{total-A} \cdot \frac{PMXd *_{total} - PMXd *_{MVs} + PMXs *_{total} - PMXs *_{MVs}}{PMXd *_{total} + PMXs *_{total}} = \\ &= PMX \ (DF)_{total-A} \cdot \left(1 - \frac{PMXd *_{MVs} + PMXs *_{MVs}}{PMXd *_{total} + PMXs *_{total}}\right) \\ &= PMX \ (DF)_{total-A} \cdot \left(1 - \frac{PMXd *_{MVs} + PMXs *_{MVs}}{PMXd *_{total} + PMXs *_{total}}\right) \\ &= PMX \ (DF)_{total-A} \cdot \left(1 - \frac{PMXd *_{MVs} + SOAX *_{MVs} + AMNITX *_{MVs} + AMSULX *_{MVs}}{PMXd *_{total} + SOAX *_{total} + AMNITX *_{total} + AMSULX *_{total}}\right) \\ &= PMX \ (DF)_{total-A} \times \\ \left(1 - \frac{PMXd *_{MVs} + T1 \cdot SOA *_{MVs} + T2 \cdot F1 \cdot SO2 *_{MVs} \cdot F2 \cdot M1 + T3 \cdot F3 \cdot NOX *_{MVs} \cdot F4 \cdot M2}{PMXd *_{total} + T1 \cdot SOA *_{total} + T2 \cdot F1 \cdot SO2 *_{total} \cdot F2 \cdot M1 + T3 \cdot F3 \cdot NOX *_{total} \cdot F4 \cdot M2}\right) \\ &= PMX \ (DF)_{total-A} \times \\ &= PMX \ (DF)_{total-A} \times \\ &= PMX \ (DF)_{total-A} \times PMXd *_{total} + PMXs + PMXs *_{total} + PMXs *_{tota$$

where:

 $PMX(DF)_{NO-MVs}$ = the estimated ambient level of particulate matter (PM) of size class X after motor-vehicle-related PM pollution of size class X is eliminated; an input in the PM damage functions (DF)

X = size classes of PM: PM_{2.5} (less than 2.5 μ m) and coarse PM₁₀ (between 2.5 and 10 μ m)

 $PMX(DF)_{total-A}$ = the measured ambient level of PM of size class X (from ambient air-quality data; see Reports #11 and 12)

PMX*_{NO-MVs} = the modeled level of PM of size class X after motor-vehiclerelated PM pollution of size class X is eliminated

 PMX^*_{total} = the modeled level of total PM pollution of size class X

PMXd*_{NO-MVs} = the modeled level of PM of size class X after direct motorvehicle-related PM pollution of size class X is eliminated

PMXs*_{NO-MVs} = the modeled level of PM of size class X after secondary motor-vehicle--related PM pollution of size class X is eliminated

PMXd*_{total} = the modeled level of PM of size class X due to all direct PM pollution of size class X

- PMXs*_{total} = the modeled level of PM of size class X due to all secondary PM pollution of size class X
- $PMXd*_{MVs}$ = modeled direct PM pollution of size class X from motor vehicles
- PMXs*_{MVs} = modeled secondary PM pollution of size class X from motorvehicles
- $SOAX^*_{MVs}$ = modeled secondary organic aerosols of size class X due to motor vehicles
- AMNITX*_{MVs} = modeled ammonium nitrate of size class X due to motor vehicles
- $AMSUL_{MVs}^*$ = modeled ammonium sulfate or ammonium bisulfate of size class X due to motor vehicles
- SOAX*_{total} = modeled total secondary organic aerosols of size class X
- AMNITX*_{total} = modeled total ammonium nitrate of size class X
- AMSUL*_{total} = modeled total ammonium sulfate or ammonium bisulfate of size class X
- T1 = the fraction of all SOAs that fall within size class X (T1= 1.0 for PM_{2.5}, T1 = 0.0 for coarse PM₁₀)
- T2 = the fraction of all ammonium sulfate or ammonium bisulfate that falls within size class X (T2= 0.95 for PM_{2.5}, T2 = 0.05 for coarse PM₁₀)
- T3 = the fraction of all ammonium nitrate that falls within size class X (T3 = 0.80 for PM_{2.5}, T3 = 0.20 for coarse PM₁₀)
- F1 = the fraction of sulfur in SO₂ pollution that is converted to sulfur in sulfate (0.15 in the Western U. S., 0.25 in the more humid Eastern U.S.)
- F2= the fraction of sulfate that is neutralized to ammonium bisulfate or ammonium sulfate (depends on the availability of NH3 emissions within the AQCR)
- F3 = the fraction of nitrogen in NO_X pollution that is converted to nitrogen in nitric acid (0.15)
- F4 = the faction of nitric acid that is neutralized to ammonium nitrate (depends on the availability of NH₃ emissions after sulfuric acid has been fully neutralized)
- M1 = mass enhancement factor: S in SO₂ to S in (NH₄)HSO₄ (1.80) or (NH₄)₂SO₄ (2.06)
- M2 = mass enhancement factor: N in NO_X (as NO_2) to N in NH4NO3 (1.74)
- SOA*_{MVs} = modeled secondary organic aerosols due to motor vehicles
- SO2*_{MVs} = modeled SO₂ pollution due to motor vehicles
- NOx^*_{MVs} = modeled NO_X pollution due to motor vehicles
- SOA*_{total} = modeled total secondary organic aerosols
- $SO2*_{total}$ = modeled total SO_2 pollution
- NOx^*_{total} = modeled total NO_X pollution.

In all cases, "pollution" (e.g., "all direct PM pollution," "all secondary PM pollution," "VOC pollution from motor vehicles") refers to official emissions [OEI in equation 6] multiplied by our emissions-correction factor [EC $_{p',i}$ in equation 6] multiplied by the normalized dispersion term [DN $_{p',i}$ in equation 6]. For simplicity, for the purpose of estimating \$-damages/kg-emitted, we will attribute all secondary ammonium sulfate to SO2 emissions, and all secondary ammonium nitrate to NO $_{x}$ emissions. This is not terribly unreasonable, because as noted above ammonia generally is not the limiting factor in the formation of secondary sulfate or nitrate PM.

16.6 COMPARISON OF OUR MODELING RESULTS WITH THE SOURCE-APPORTIONMENTS FROM CHEMICAL MASS-BALANCE STUDIES

How do our model results compare with the results of other ways of estimating the contribution of various sources to ambient air pollution? As discussed above, another way to estimate the contribution of motor-vehicles to ambient particulate pollution is to examine the chemical composition of particulate matter captured at airquality monitors, and relate the chemical profile of different emissions sources to the chemical profile of the ambient pollutant. This statistical "chemical mass-balance" (CMB) relationship results in weights, or source-apportionments, for the different emission sources. These CMB source apportionments are analogous to the pollutant shares -- PP*/PI*, from equation 1 above -- calculated by our model.

Table 16-9, reviewed above, presents CMB results for 21 counties, mostly in the western U.S., and 10 sources of particulate matter: primary geologic (PG), primary construction (PC), primary motor vehicle (PMV), primary vegetative burning PV), secondary ammonium sulfate (SAS), secondary ammonium nitrate (SAN), and four miscellaneous categories (M1 to M4). In order to compare the CMB results with our model results, we group the CMB studies of Table 16-9 by county and state (because we have emissions data — a key part of our modeling — by county), and chose from the ten CMB source categories four that match reasonably closely to source categories in our model. Thus, in Table 16-26, we compare the CMB and model estimates of pollutant shares for road dust ("primary geologic" in the CMB studies), motor vehicles, and secondary ammonium nitrate and sulfate. For the CMB studies, we show the low and the high source-apportionment share for each county and emissions source. For our modeling results, we show the low-cost and high-cost cases (see the notes to Table 16-26).

For three out of four source categories -- road dust/geologic, secondary ammonium sulfate, and secondary ammonium nitrate -- the model results and the CMB results agree reasonably well, although the CMB results generally are more variable. There are two explanations for the greater variability of the CMB studies. First, the CMB studies often look at a relatively short interval, say, one month, whereas our modeling results are based on annual emissions. Second, the CMB studies capture

pollution at single spot, often in downtowns where the motor vehicle contribution should be relatively high, whereas we model shares on the basis of emissions throughout the entire county or air basin.

Our estimates of the contribution of road dust agree reasonably well with the CMB estimates of the contribution of primary geologic matter. The CMB estimates probably include some geologic material other than road dust, but the amount most likely is small, and in any case our model results look to be slightly less than the CMB results on average. Most importantly, our model results (which include substantial corrections to the emissions inventory, as well as corrections for particle settling and dispersion) and the CMB results agree that road dust constitutes a major fraction of ambient particulate air pollution in urban areas.

The comparison for the nitrate contribution also is favorable, particularly for California. Outside of California, CMB studies report low nitrate levels -- typically lower than what we estimate, although our estimates themselves are relatively low. Perhaps our assumption that 5% to 7% of NO_X converts to nitrate (see above) is too high for some parts of the country.

Our modeled estimates of the contribution of sulfate are within the range found in the CMB studies about half of the time, and are either above or below the range the rest. It is encouraging to note that both the CMB studies and our modeled results find a large contribution from sulfates in Stuebenville, Ohio, probably due to power plants in the region.

However, the CMB studies estimate a much larger direct contribution from motor vehicles than does our model. Our model estimates that direct, primary PM emissions from motor vehicles contribute 1% to 9% of ambient PM; the CMB studies estimate that motor vehicles contribute about 3 times as much, although again there is considerable variability in the CMB results⁴⁰.

We can explain at least some of this difference. For example, the CMB estimates of motor-vehicle emissions probably include PM from non-motor-vehicle diesel combustion. On the basis of the data in Tables 16-21 and 16-24, we might expect that in the CMB studies some 25% of the PM attributed to motor vehicles actually comes from other diesel sources, such as off-road engines and trains, whose PM emissions have the same profile as do the PM emissions from heavy-duty diesel trucks. Another explanation of the difference is that CMB studies often sample at times and places of especially high motor-vehicle contributions. Even so, the difference between our model results and the CMB results is conspicuous, and we certainly cannot rule out the possibility that our model is significantly underestimating the direct contribution of motor vehicles to ambient PM, either because it underestimates motor-vehicle emissions or overestimates the contribution of other emission sources.

91

 $^{^{40}}$ As we noted, part of the variability is due to the short time-span common in CMB studies. To avoid this temporal problem for at least one area -- Riverside County -- we compared our results with those of Chow et al. (1992b: Figure 5), who estimate that motor vehicle contribute 7-17% of the *annual* average PM_{10} at three sites around Riverside, California. Our estimate of 5-7% for this particular area still is much less than the CMB estimate.

Overall, we find that our relatively simple model of emissions, dispersion, and atmospheric chemistry compares with reasonably well with the results of the CMB studies, especially in light of the limitations of the latter.

16.7 REFERENCES

- J. P. Adhikari, G. K. Sen, B. K. Tewary, A. Banerjee, R. N. Mukherjee, D. P. Rajwar and B. Singh, "Modification and Testing of a Gaussian Dispersion Model for Particulate Matter in the Respirable Size Range," *Atmospheric Environment* **24A**: 1647-1651 (1990).
- C. D. Ahrens, Meteorology Today, West Publishing Company, St. Paul, Minnesota (1985).
- D. T. Allen, "Loadings, Size Distributions, and Sources of Compound Classes in Los Angeles Aerosol," *Inhalation Toxicology* **7**: 723-734 (1995).
- A. P. Altshuler, "Atmospheric Particle Sulfur and Sulfur Dioxide Relationships at Urban and Nonurban Locations," *Atmospheric Environment* **18**: 1421-1431 (1984).
- P. Altshuller et al., "Physics and Chemistry of Particulate Matter," in *Air Quality Criteria for Particulate Matter*, Vol. I of III, EPA/600/P-95/001aF, U. S. Environmental Protection Agency, Office of Research and Development, Washington, D. C., pp. 3-1 to 3-243 (1996).
- R. A. Anthes, H. A. Panofsky, J. J. Cahir and A. Rango, *The Atmosphere*, Charles E. Merril Company, Columbus, Ohio (1975).
- B. R. Appel, "Atmospheric Sample Analysis and Sampling Artifacts," in *Aerosol Measurement: Principles, Techniques, and Applications*, ed. by K. Willeke and P. A. Baron, Van Nostrand Reinhold, New York, New York, pp. 233-259 (1993).

Automotive Handbook, 3rd edition, Robert Bosch GmbH, Germany (distributed by the Society of Automotive Engineers, Warrendale, Pennsylvania), 1993.

- Auto/Oil Air Quality Improvement Research Program, Real World Automotive Emissions -- Results of Studies in the Fort McHenry and Tuscarora Mountain Tunnels, Technical Bulletin No. 14, Coordinating Research Council, Atlanta, Georgia, June (1995).
- M. Balogh, T. Larson, and F. Mannering, "Analysis of Fine Particulate Matter near Urban Roadways," *Transportation Research Record* **1416**: 25-32 (1993).
- W. Barnard, "Assessment of Highway Particulate Impacts", presented at *Transportation Research Board, Air Quality Committee*, Washington, D. C., January 10 (1996).
- R. A. Barnes, G. S. Parkinson and A. E. Smith, "The Costs and Benefits of Sulphur Oxide Control," *Journal of the Air Pollution Control Association* **33**: 737-741 (1983).

- P. Benson, *CALINE4 -- A Dispersion Model for Predicting Air Pollutant Concentrations Near Roadways*, FHWA/CA/TL-84/15, Federal Highway Administration and California Department of Transportation, Washington, D. C., November (1984).
- F. Black, W. Ray, F. King, W. Karches, R. Bradow, N. Perry, J. Duncan and W. Crews, "Emissions from In-use Heavy-Duty Gasoline Trucks", Paper #841356, *SAE Technical Paper Series*, Society of Automotive Engineers, Warrendale, Pennsylvania (1984).

Bureau of the Census, *USA Counties. A Statistical Abstract Supplement*, CD-ROM, C3.134/6: vol. 1994, U.S. Department of Commerce, Economics and Statistics Administration, Washington, D. C. (1994).

- R.M. Burton, H. H. Suh, and P. Koutrakis, "Spatial Variation in Particulate Concentrations within Metropolitan Philadelphia," Environmental Science and Technology **30**: 400-407 (1996).
- T. A. Cahill, D. Sperling, D. P. Chang, E. Gearhart, O. Carbacho and L. Ashbaugh, *PM*₁₀ *Aerosols from California Roads*, University of California, Davis, California, March 2 (1994).
- T. A. Cahill and P. Wakabayashi, "Compositional Analysis of Size-Segregated Aerosol Samples," in *Measurement Challenges in Atmospheric Chemistry*, ed. by L. Newman, American Chemical Society, Washington, D. C., pp. 211-228 (1993).

California Air Resources Board, Public Workshop Announcement, Motor-Vehicle Emission-Inventory 7G (MVEI7G) Update, December (1995).

- P. M. Carey, *Air Toxics Emissions From Motor Vehicles.*, EPA-AA-TSS-PA-86-5, U.S. Environmental Protection Agency, Office of Mobile Sources, Ann Arbor, Michigan, September (1987).
- G. R. Cass, "Comments on Sources, Atmospheric Levels, and Characterization of Airborne Particulate Matter," *Inhalation Toxicology* **7**: 765-768 (1995).
- G. R. Cass and H. A. Gray, "Regional Emissions and Atmospheric Concentrations of Diesel Engine Particulates," in *Diesel Exhaust: A Critical Analysis of Emissions, Exposure, and Health Effects*, Health Effects Institute, Cambridge, Massachusetts, pp. 125-137 (1995).
- S. Cha, P. Carter, and R. L. Bradow, "Simulation of Automobile Brake Wear Dynamics and Estimation of Emissions," SAE Technical Paper Series, #831036, Society of Automotive Engineers, Warrendale, Pennsylvania (1983).

- T. Y. Chang, S. J. Rudy, G. Kuntasal, and R. A. Gorse Jr., "Impact of Methanol Vehicles on Ozone Air Quality," *Atmospheric Environment* **23**: 1629-1644 (1989).
- J. C. Chow, J. G. Watson, D. H. Lowenthal, and R. J. Countess, "Sources and Chemistry of PM₁₀ Aerosol in Santa Barabara County, California," *Atmospheric Environment* **30**: 1489-1499 (1996).
- J. C. Chow, D. Fairley, J. G. Watson, R. DeMandel, E. M. Fujita, D. H. Lowenthal, Z. Lu, C. A. Frazier, G. Long and J. Cordova, "Source Apportionment of Wintertime PM₁₀ at San Jose California," *Journal of Environmental Engineering*, May, pp. 378-387 (1995).
- J. C. Chow, J. G. Watson, J. E. Houck, L. C. Pritchett, C. F. Rogers, C. A. Frazier, R. T. Egami, and B. M. Ball, "A Laboratory Resuspension Chamber to Measure Fugitive Dust Size Distributions and Chemical Compositions," *Atmospheric Environment* **28**: 3463-3481 (1994a).
- J. C. Chow, J. G. Watson, E. M. Fujita, Z. Lu, D. R. Lawson and L. Ashbaugh, "Temporal and Spatial Variations of $PM_{2.5}$ and PM_{10} Aerosol in the Southern California Air Quality Study," *Atmospheric Environment* **28**: 2061-2080 (1994c).
- J. C. Chow, J. G. Watson, D. M. Ono, and C. V. Mathai, "PM10 Standards and Nontraditional Particulate Source Controls: A Summary of the A&WMA/EPA International Specialty Conference," *Journal of the Air and Waste Management Association* **43**: 74-84 (1993).
- J. C. Chow, J. G. Watson, D. H. Lowenthal, P. A. Solomon, K. L. Magliano, S. D. Ziman, and L. W. Richards, "PM10 Source Apportionment in California's San Joaquin Valley," *Atmospheric Environment.*, **26A**: 3335-3354 (1992a).
- J. C. Chow, C. S. Liu, J. Cassmassi, J. G. Watson, Z. Lu, and L. C. Pritchett, "A Neighborhood-Scale Study of PM10 Source Contributions in Rubidoux, California," *Atmospheric Environment*, **26A**: 693-706 (1992b).
- J. C. Chow, J. G. Watson, L. W. Richards, D. L. Haase, C. McDade, D. L. Dietrich, D. Moon and C. Sloane, *The 1989-90 Phoenix PM*₁₀ *Study Volume II: Source Apportionment*, Final Report, DRI Document No. 8931.6F1, Desert Research Institute, Energy and Environmental Engineering Center, Reno, Nevada, April (1991).
- J. C. Chow, J. G. Watson, C. A. Frazier, A. G. R. T. Egami and C. Ralph, "Spatial and Temporal Source Contributions to PM_{10} and $PM_{2.5}$ in Reno, NV," in PM_{10} : Implementation of Standards, Transactions TR-13, ed. by C. V. Mathai and D. H.

- Stonefield, Air and Waste Management Association, Pittsburgh, Pennsylvania, pp. 438-457 (1988).
- C. Cowherd, Midwest Research Institute, Kansas City, Missouri, personal communication, October 4 (1995a).
- C. Cowherd, Midwest Research Institute, Kansas City, Missouri, personal communication, August 31 (1995b).
- R. G. Derwent, M. E. Jenkin, and S. M. Saunders, "Photochemical Ozone Creation Potentials for a Large Number of Reactive Hydrocarbons Under European Conditions," *Atmospheric Environment* **30**: 181-199 (1996).
- H. E. Dietzmann, M. A. Parness and R. L. Bradow, "Emissions from Trucks by Chassis Version of 1983 Transient Procedure", Paper #801371, *SAE Technical Paper Series*, Society of Automotive Engineers, Warrendale, Pennsylvania (1980).
- R. R. Draxler, "Determination of Atmospheric Diffusion Parameters," *Atmospheric Environment* **10**: 99-105 (1976).
- A. L. Dresser and B. K. Baird, "A Dispersion and Receptor Model Analysis of the Wintertime PM₁₀ Problem in Telluride, Colorado," in *PM*₁₀: *Implementation of Standards*, ed. by C. V. Mathai and D. H. Stonefield, Air and Waste Management Association, Pittsburgh, Pennsylvania, p. 458 (1988).
- T. G. Dzubay, R. K. Stevens, G. E. Gordon, I. Olmez, A. E. Sheffeld and W. J. Courtney, "A Composite Receptor Method Applied to Philadelphia Aerosol," *Environmental Science and Technology* **22**: 46-52 (1988).
- D. J. Eatough, F. M. Caka and R. J. Farber, "The Conversion of SO₂ to Sulfate in the Atmosphere," *Israel Journal of Chemistry* **34**: 301-314 (1994).

Energy Information Administration, Fuel Oil and Kerosene Sales 1994, DOE/EIA-0535(94), U. S. Department of Energy, Washington, D. C., September (1995).

Environmental Protection Agency (EPA), Office of Air Quality Planning and Standards, *Compilation of Air Pollutant Emission Factors, Volume I: Stationary Sources*, AP-42, fifth edition, Research Triangle Park, North Carolina, January (1995a).

Environmental Protection Agency (EPA), Office of Mobile Sources, *Highway Vehicle Emission Estimates -- II*, white paper, Ann Arbor, Michigan, May (1995b).

Environmental Protection Agency, Office of Mobile Sources, *Draft User's Guide to PART5: A Program for Calculating Particulate Emissions from Motor Vehicles*, EPA-AA-AQAB-94-2, Ann Arbor, Michigan, February (1995c).

Environmental Protection Agency (EPA), Office of Policy, Planning, and Evaluation (OPPE), computer transmission of data file containing estimate of emissions (excluding VOCs from plants and NO_X from soil), in every county in the U. S. in 1990, by E. H. Pechan Associates, Springfield, Virginia, for the EPA OPPE, Washington, D.C. (1995d).

Environmental Protection Agency (EPA), Office of Air Quality Planning and Standards (OAQPS), Emissions Factor and Inventory Group, computer transmission of data file containing estimate of emissions of VOCs from plants and NO_X from soil, in every county in the continental U. S. in 1990, Research Triangle Park, North Carolina (1995e).

Environmental Protection Agency, Office of Air Quality Planning and Standards, *User's Guide for the Industrial Source Complex (ISC3) Dispersion Models*, EPA-454/B-95-003b, Vol. II, Research Triangle Park, North Carolina, September (1995f).

Environmental Protection Agency (EPA), Office of Policy, Planning and Evaluation/Office of Policy Analysis, *Emissions Inventory for the National Particulate Matter Study, Final Draft*, EPA 68-D3005, Washington, D. C., July (1994a).

Environmental Protection Agency (EPA), Office of Policy, Planning and Evaluation/Office of Policy Analysis, *Development of the OPPE Particulate Programs Implementation Evaluation System, Final,* EPA 68-D3005, Washington, D. C., September (1994b).

Environmental Protection Agency (EPA), Office of Health and Environmental Assessment, *Air Quality Criteria for Carbon Monoxide*, EPA/600/8-90/045F, Washington, D. C., June (1992).

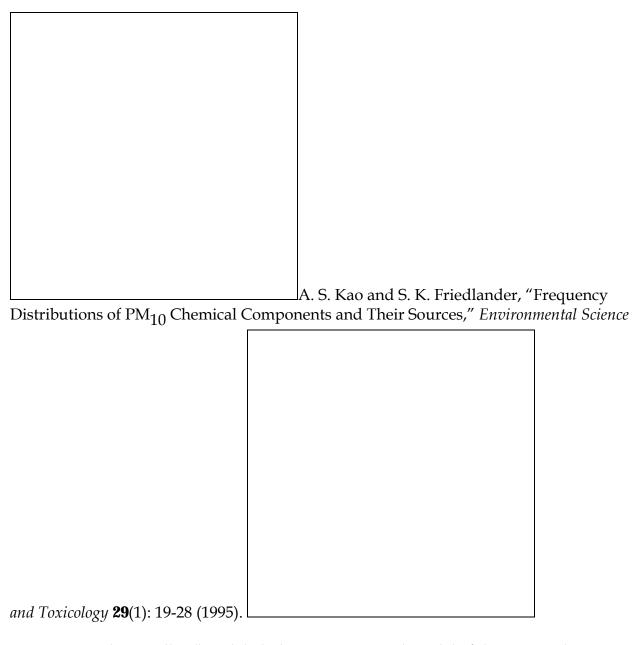
Environmental Protection Agency, National Air Data Branch, magnetic data tape of ambient air quality measurements of CO, NO_X , O3, $PM_{2.5}$, PM_{10} and TSP at all available monitoring sites in the United States 1988-1991, Research Triangle Park, North Carolina (1993).

Environmental Protection Agency (EPA), Office of Air Quality Planning and Standards, *Air Emissions Species Manual, Volume II, Particulate Matter Species Profiles*, EPA-450/2-90-001b, second edition, Research Triangle Park, North Carolina, January (1990).

Environmental Protection Agency (EPA), Office of Mobile Sources, *Compilation of Air Pollutant Emission Factors, Volume II: Mobile Sources*, AP-42, fourth edition, Ann Arbor, Michigan (1985).

- J. J. Erbink, "A Practical Model for the Calculation of σ_y and σ_z for Use in an On-Line Gaussian Dispersion Model for Tall Stacks, Based on Wind Fluctuations," *Atmospheric Environment* **25A**: 277-283 (1991).
- D. L. Ermak, "An Analytical Model for Air Pollutant Transport and Deposition from a Point Source," *Atmospheric Environment* **11**: 231-237 (1977).
- N. J. Eyre, E. Ozdemiroglu, D. W. Pearce, and P. Steele, "Fuel and Location Effects on the Damage Costs of Transport Emissions," *Journal of Transport Economics and Policy*, January: 5-24 (1997).
- Federal Highway Administration, *Highway Statistics* 1990, FHWA-PL-91-003, U. S. Department of Transportation, Washington, D. C. (1991).
- R. C. Flagan, "Probing the Chemical Dynamics of Aerosols," in *Measurement Challenges in Atmospheric Chemistry*, ed. by L. Newman, American Chemical Society, Washington, D. C., pp. 185-210 (1993).
- E. M. Fujita, J. G. Watson, J. C. Chow and K. L. Magliano, "Receptor Model and Emissions Inventory Source Apportionment of Nonmethane Organic Gases in California's San Joaquin Valley and San Francisco Bay Area," *Atmospheric Environment* **29**: 3019-3035 (1995).
- E. M. Fujita et al., "Comparison of Emission Inventory and Ambient Concentration Ratios of CO, NMOG, and NOx in Cailfornia's South Coast Air Basin," *Journal of the Air and Waste Management Association* **42**:264-276 (1992).
- J. German, "Off-Cycle Emission and Fuel Efficiency Considerations," presented at the 1995 Conference on Sustainable Transportation Energy Strategies, Asilomar, California, July 31-August 3 (1995).
- A. W. Gertler, J. C. Sagebiel, W. R. Pierson, C. Atkinson, and N. Clark, *On Road and Chassis Dynamometer Measurements of Heavy-Duty Vehicle Emission Factors*, Desert Research Institute, Reno, Nevada (1995).
- R. F. Griffiths, "Errors in the Use the Briggs Parameterization for Atmospheric Dispersion Coefficients," *Atmospheric Environment* **28**: 2861-2865 (1994).
- R. Guensler, D. Sperling and P. Jovanis, *Uncertainty in the Emission Inventory for Heavy-Duty Diesel-Powered Trucks*, UCD-ITS-RR-91-02, Institute of Transportation Studies, University of California, Davis, California, June (1991).

- S. R. Hanna, G. A. Briggs and R. P. H. Jr., *Handbook on Atmospheric Diffusion*, Technical Information Center, U. S. Department of Energy, Washington, D. C. (1982).
- A. D. A. Hanson and H. Rosen, "Individual Measurements of the Emission Factor of Aerosol Black Carbon in Automobile Plumes," *Journal of the Air and Waste Management Association* **40**: 1654-1657 (1990).
- D. Hegg, "The Importance of Liquid-Phase Oxidation of SO₂ in the Troposphere," *Journal of Geophysical Research* **90**(D2): 3773-3779 (1985).
- C. N. Herrick and J. L. Kulp, *Interim Assessment: The Causes and Effects of Acidic Deposition*, Volume 1, Executive Summary, National Acid Precipitation Assessment Program, Office of the Director of Research, Washington, D. C. (1987).
- G. Holzworth, *Mixing Heights, Wind Speed, and Potential for Urban Air Pollution Throughout the Contiguous United States,* AP-101, U.S. Environmental Protection Agency, Research Triangle Park, North Carolina, January (1972).
- J. L. Horowitz, *Air Quality Analysis for Urban Transportation Planning*, MIT Press, Cambridge, Massachusetts (1982).



J. Langner and H. Rodhe, "A Global Three-Dimensional Model of the Tropospheric Sulfur Cycle," *Journal of Atmospheric Chemistry* **13**: 225-263 (1991).

K. W. Lee and M. Ramamurthi, "Filter Collection," in *Aerosol Measurement: Principles, Techniques, and Applications*, ed. by K. Willeke and P. A. Baron, Van Nostrand Reinhold, New York, New York, pp. 179-205 (1993).

J. J. Lin, K. E. Noll and T. M. Holsen, "Dry Deposition Velocities as a Function of Particle Size in the Ambient Atmosphere," *Aerosol Science and Technology* **20**: 239-252 (1994).

- J. Lin, G. Fang, T. M. Holsen and K. E. Noll, "A Comparison of the Dry Deposition Modeled from Size Distribution Data and Measured with a Smooth Surface for Total Particle Mass, Lead and Calcium in Chicago," *Atmospheric Environment* **27A**: 1131-1138 (1993).
- F. W. Lipfert, S. C. Morris and R. E. Wyzga, "Acid Aerosols: The Next Criteria Air Pollutant," *Environmental Science and Technology* **23**: 1316-1322 (1989).
- P. J. Lioy and J. Waldman, "Acidic Sulfate Aerosols: Characterization and Exposure," *Environmental Health Perspectives* **79**: 15-34 (1989).
- D. A. Lundgren and R. M. Burton, "Effect of Particle Size Distribution on the Cut Point Between Fine and Coarse Ambient Mass Fractions," *Inhalation Toxicology* **7**: 131-148 (1995).
- K. L. Magliano, F. W. Lurmann and J. G. Watson, "Reconciliation of Emissions Inventory and Ambient Data in the San Joaquin Valley and San Francisco Bay Area," Paper 93-WP-100.08, presented at *Air and Waste Management Association 86th Annual Meeting & Exhibition*, Denver, Colorado, June 13-18 (1993).
- J. N. McHenry and R. L. Dennis, "The Relative Importance of Oxidation Pathways and Clouds to Atmospheric Ambient Sulfate Production as Predicted by the Regional Acid Deposition Model," *Journal of Applied Meteorology* **33**: 890-905 (1994).
- P. M. Merz, L. J. Painter, and P. R. Ryason, "Aerometric Data Analysis-Time Series Analysis and Forecast and an Atmospheric Smog Diagram," *Atmospheric Environment*: 319-342 (1972).

Midwest Research Institute, *Emission Factor Documentation for AP-42, Sections* 11.2.5 and 11.2.6, *Paved Roads*, Prepared for the Emission Inventory Branch, Office of Air Quality Standards and Planning, U. S. Environmental Protection Agency, Research Triangle Park, North Carolina, March 8 (1993).

National Research Council, Committee on Tropospheric Ozone Formation and Measurement, *Rethinking the Ozone Problem in Urban and Regional Air Pollution*, National Academy Press, Washington, D. C. (1991).

- K. W. Nicholson, J. R. Branson, P. Giess, and R. J. Cannell, "The Effects of Vehicle Activity on Particle Resuspension," *Journal of Aerosol Science* **20**: 1425-1428 (1989).
- W. W. Nokes and P. E. Benson, *Development of Worst Case Meteorology Criteria*, FHWA/CA/TL-85/14, California Department of Transportation, Sacramento, California, November (1985).

- Office of Technology Assessment, *Urban Ozone and the Clean Air Act: Problems and Proposals for Change*, U. S. Congress, Washington, D. C., April. (1988). See also: Office of Technology Assessment, *Catching our Breath, Next Steps for Reducing Urban Ozone*, U. S. Congress, Washington, D. C., July, (1989).
- S. J. Ostria, "Assessing the Emissions Contribution of Intercity Trucking", Paper 960513, presented at Transportation Research Board 75th Annual Meeting, Washington, D. C., January 7-11 (1996).
- W. R. Pierson and W. W. Brachaczek, "Particulate Matter Associated with Vehicles on the Road. II," *Aerosol Science and Technology* **2**: 1-40 (1983).
- C. Pilinis and R. J. Farber, "Evaluation of the Effects of Emission Reductions on Secondary Particulate Matter in the South Coast Air Basin of California," *Journal of the Air and Waste Management Association* **41**: 702-709 (1991).
- J. P. Pinto et al., "Sources and Emissions of Atmospheric Particles," in *Air Quality Criteria for Particulate Matter*, Vol. I of III, EPA/600/P-95/001aF, U. S. Environmental Protection Agency, Office of Research and Development, Washington, D. C., pp. 3-1 to 3-243 (1996).
- W. F. Rogge, L. M. Hildemann, M. A. Mazurek, G. R. Cass and B. T. Simoneit, "Quantifying the Contribution of Sources of Organic Aerosols in Atmospheric Samples," *Inhalation Toxicology* **7**: 796-798 (1995).
- M. Ross, "Automotive Fuel Consumption and Emissions: Effects of Vehicle and Driving Characteristics," *Annual Review of Energy and Environment* **19**: 75-112 (1994).
- M. Ross, R. Goodwin, R. Watkins, M. Q. Wang and T. Wenzel, *Real-World Emissions from Model Year* 1993, 2000 and 2010 Passenger Cars, American Council for an Energy Efficient Economy, Washington, D. C., November (1995).
- R. D. Rowe, L. G. Chestnut, C. M. Lang, S. S. Bernow and D. E. White, "The New York Environmental Externalities Cost Study: Summary of Approach and Results", presented at *External Costs of Energy*, Brussels, Belgium, January 30-31, 1995 (1995).
- W. M. Ryan, C. R. Badgett-West, D. R. Holtz, T. A. Peters, J. A. Cooper and D. Ono, "Reconciliation of the Receptor and Dispersion Modeling Impacts of PM₁₀ in Hayden, AZ," in *PM*₁₀: *Implementation of Standards*, Vol. TR-13, ed. by C. V. Mathai and D. H. Stonefield, Air Pollution Control Association, Pittsburgh, Pennsylvania, pp. 419-429 (1988).

- J. Sagebiel, data transmittal, Desert Research Institute, Reno, Nevada, November 6 (1995).
- R. C. Schwing, B. W. Southworth, C. R. von Buseck, and C. J. Jackson, "Benefit-Cost Analysis of Automotive Emission Reductions," *Journal of Environmental Economics and Management* **7**: 44-64 (1980).
- C. Sioutas, P. Koutrakis, S. T. Ferguson and R. M. Burton, "Development and Evaluation of a Prototype Ambient Particle Concentrator for Inhalation Exposure Studies," *Inhalation Toxicology* **7**: 633-644 (1995).
- P. A. Solomon, L. G. Salmon, T. Fall and G. R. Cass, "Spatial and Temporal Distribution of Atmospheric Nitric Acid and Particulate Nitrate Concentrations in the Los Angeles Area," *Environmental Science and Technology* **26**: 1594-1601 (1992).
- D. Solomon, E. H. Pechan Associates, Raleigh, North Carolina, personal communication, October 4 (1995).
- K. A. Verrall and R. L. Williams, "A Method for Estimating the Standard Deviation of Wind Directions," *Journal of Applied Meteorology* **21**: 1922-1925 (1982).
- J. M. Waldman, P. Koutrakis, G. Allen, G. D. Thurston, R. M. Burton and W. E. Wilson, "Human Exposure to Particle Strong Acidity," *Inhalation Toxicology* **7**: 657-699 (1995).
- M. Walsh, transportation consultant, Arlington, Virginia, personal communication, August 19 (1995).
- J. G. Watson, J. C. Chow, F. W. Lurmann and S. P. Musarra, "Ammonium Nitrate, Nitric Acid, and Ammonia Equilibrium in Wintertime Phoenix, Arizona," *Journal of the Air and Waste Management Association* **44**: 405-412 (1994a).
- J. G. Watson, J. C. Chow, Z. Lu, E. M. Fujita, D. H. Lowenthal, D. R. Lawson, and L. L. Ashbaugh, "Chemical Mas Balance Source Apportionment of PM₁₀ During the Southern California Air Quality Study," *Aerosol Science and Technology* **21**: 1-36 (1994b).
- J. G. Watson, J. C. Chow, and C. V. Mathai, "Receptor Models in Air Resources Management: A Summary of the APCA International Specialty Conference," *Journal of the Air Pollution Control Association* **39**: 419-426 (1989).
- J. G. Watson, J. C. Chow, R. T. Egami, C. A. Frazier, A. Goodrich and C. Ralph, *PM*₁₀ Source Apportionment the Truckee Meadows, Nevada, for State Implementation Plan Development, Volume 1: Modeling Methods and Results, Final Report, DRI Document

- 8086.2F, Desert Research Institute, Energy and Environmental Engineering Center, Reno, Nevada, February (1988a).
- J. G. Watson, J. C. Chow, L. W. Richards, W. D. Neff, S. R. Andersen, D. L. Dietrich, J. E. Houck and I. Omez, *The 1987-88 Metro Denver Brown Cloud Study Volume II: Measurements*, Final Report, DRI Document No. 8810 1F2, Desert Research Institute, Reno, Nevada, November 8 (1988b).
- D. N. Whittorf, A. W. Gertler, J. C. Chow, W. R. Barnard, and H. A. Jongedyk, "The Impact of Diesel Particulate Emissions on Ambient Particulate Loadings," paper 94-WP91.01, presented at the 87th Annual Meeting & Exhibition of the Air & Waste Management Association, Cincinnati, Ohio, June 19-24 (1994).
- P. B. Williams, M. P. Buhr, R. W. Weber, M. A. Volz, J. W. Koepke and J. C. Selner, "Latex Allergen in Respirable Particulate Air Pollution," *Journal of Allergy and Clinical Immunology* **95**: 88-95 (1995).
- W. E. Wilson, Environmental Protection Agency, Research Triangle Park, North Carolina, memo on "Emission Inventory Factors" to C. A. Knoppes, Environmental Protection Agency, Office of Policy Analysis, Washington, D. C., February 7 (1995).
- L. B. Wiman, M. H. Unsworth, S. E. Lindberg, B. Bergkvist, R. Jaenicke, and H. C. Hansson, "Perspectives on Aerosol Deposition to Natural Surfaces: Interactions Between Aerosol Residence Times, Removal Processes, the Biosphere and Global Environmental Change," *Journal of Aerosol Science* 21: 313-338 (1990).
- A. M. Winer and W. F. Busby, "Atmospheric Transport and Transformation of Diesel Emissions," in *Diesel Exhaust: A Critical Analysis of Emissions, Exposure, and Health Effects*, Health Effects Institute, Cambridge, Massachusetts, pp. 83-106 (1995).
- Y. Zhang, Y. Sunwoo, V. Kotamarthi and G. R. Carmichael, "Photochemical Oxidant Processes in the Presence of Dust: An Evaluation of the Impact of Dust on Particulate Nitrate and Ozone Formation," *Journal of Applied Meteorology* **33**: 813-824 (1994).

ABBREVIATIONS USED IN THE TABLES IN THIS REPORT

Vehicles

LDGV = light-duty gasoline vehicle (passenger vehicles, including station wagons and motorcycles)

LDGT = light-duty gasoline truck (light-duty gasoline trucks (trucks, vans, minivans, jeeps, and utility vehicles, that have a gross vehicle weight rating of 8,500 lbs or less and a curb weight of 6,000 lbs or less)

LDGT1 = LDGT with a weight rating of 6,000 lbs or less

LDGT2 = LDGT with a weight rating of 6,001 to 8,500 lbs

HDGT = heavy-duty gasoline truck (all other gasoline trucks, and buses)

LDDV = light-duty diesel vehicle (passenger vehicles, including station wagons)

LDDT = light-duty diesel truck (trucks, vans, minivans, jeeps, and utility vehicles, that have a gross vehicle weight rating of 8,500 lbs or less and a curb weight of 6,000 lbs or less)

HDDT = heavy-duty diesel truck (all other diesel trucks, and buses)

LDV = light-duty vehicle (LDGV + LDDV)

HDV = heavy-duty vehicle (HDGV + HDDV)

VMT = vehicle miles traveled

Pollutants

CO = carbon monoxide

HC = hydrocarbons

NO₂ = nitrogen dioxide

 NO_X = nitrogen oxides (including but not limited to NO_2)

NH3 = ammonia

 $O_3 = ozone$

PM = particulate matter

 PM_{10} = particulate matter with a diameter of 10 microns or less

 $PM_{2.5}$ = particulate matter with a diameter of 2.5 microns or less

Coarse PM₁₀ = particulate matter with a diameter between 2.5 and 10 microns

 SO_2 = sulfur dioxide

 SO_X = sulfur oxides

SOA = secondary organic aerosols

TSP = total suspended particulates

VOCs = volatile organic compounds

Emissions tests

HDTC = Heavy-Duty Transient Cycle

DRR = Durham Road Route

FTP = Federal Test Procedure

TABLE 16-1. CORRECTIONS TO THE EMISSIONS INVENTORY: THE RATIO OF OUR ESTIMATE OF EMISSIONS TO THE EPA'S (1995D) OFFICIAL ESTIMATES

Emission source	VC	Cs	C	O	N	O _X	PN	110	PM	[2.5	SO	$O_{\mathbf{X}}$
	low	high	low	high	low	high	low	high	low	high	low	high
LDGV and LDGT	1.1	1.3	1.5	1.8	1.2	1.4	1.5	2.0	1.5	2.0	1.0	1.0
All other vehicle classes	1.0	1.0	1.0	1.0	1.0	1.0	1.5	2.0	1.5	2.0	1.0	1.0
Road dust, paved roads ^a	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.3	0.8	0.07	0.57	n.a.	n.a
Road dust, unpaved roads ^b	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1.0	1.0	0.30	0.95	n.a.	n.a.
Construction (except road) ^C	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.5	0.1	5.0	1.0	n.a.	n.a.
Road construction ^C	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	0.1	0.5	1.0	5.0	n.a.	n.a.
Wind erosion ^d	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1.2	1.1	1.2	1.1	n.a.	n.a.

n.a. = not applicable.

Each entry is equal to the ratio of our estimate of emissions to the EPA's (1995d) estimate. Hence, we multiply the official emission-inventory estimates by these correction factors. See the text for details.

Note that "low" and "high" refer to *motor-vehicle*-related costs. Thus, higher emissions from sources, such as wind erosion and construction (except road construction), that are unrelated to motor-vehicle use result in a lower pollution share for motor vehicles and hence a lower cost.

^aIn the lower bound we assume that 10% of PM₁₀ from paved roads is PM_{2.5}; in the upper bound we assume 30%. We discuss this in the text.

^bIn the lower bound we assume that 8% of PM₁₀ from unpaved roads is PM_{2.5}; in the upper bound we assume 25%. We discuss this in the text.

^CEmissions from all construction, including road construction, are estimated with a single emission-factor, discussed in the text. However, we assume that the errors in the estimation of emissions from road construction are independent of the errors in the estimation of emissions from other construction, so that it is possible to have the low-value correction factor for road construction with the high-value factor for other construction.

 $^{
m d}$ The EPA (1995d) has not accounted for all natural sources of dust. We have increased emissions from wind erosion slightly to account for this

TABLE 16-2. PM AND OTHER EXHAUST EMISSIONS FROM HIGH-MILEAGE, IN-USE LIGHT-DUTY GASOLINE VEHICLES COMPARED TO PART5 MODEL EMISSIONS

		Exhaust emissions (g/mi)				
	Miles	PM	НС	СО	$NO_{\mathcal{X}}$	
Average of all 23 vehicles ^a	105,691	0.18	3.52	45.03	1.72	
Average of 6 smoking vehicles ^a	119,925	0.56	6.18	63.47	1.57	
Average of 17 non-smoking vehicles ^a	100,667	0.05	2.59	38.52	1.78	
PART5 Model ^b	n.a.	0.020	n.a.	n.a.	n.a.	

n.a. = not applicable.

bSagabiel reported results from vehicles made from 1976 to 1990, and tested in Nevada; our assumptions for PART5 try to replicate these conditions as closely as possible. In the PART5 estimate, we specify a 1989 fleet, transient cycle, speed of 19.6 mph, low altitude, no inspection and maintenance, no reformulated gasoline, and PM30. Note two things: 1) presumably Sagabiel used a standardized drive cycle called the Federal Test Procedure (FTP). We can not specify the FTP when using the PART5 model; however, the drive cycle and speed make no difference in the PART5 estimates, so specifying the transient cycle and a speed of 19.6 mph does not affect the results; and 2) Sagabiel reported only "PM" without indicating a size cutoff. When running PART5, we chose PM30, although PM10 would have given the same result, since all motor vehicle exhaust PM is in the PM10 size range.

Seven of the 23 vehicles were light-duty gasoline trucks (LDGT1) and the rest were light-duty gasoline vehicles (LDGV), so we estimated emissions for both vehicle types and calculated a weighted average. We found 0.018 g/mi for LDGVs and 0.026 g/mi for LDGT1s, which gives a weighted average of 0.02 g/mi. We report exhaust emissions only, and exclude tirewear, brakewear and indirect sulfates.

^aFrom IM240 test results reported by Sagabiel (1995).

TABLE 16-3. PM EXHAUST EMISSIONS FROM IN-USE HEAVY-DUTY VEHICLES TESTED OVER A TRANSIENT CYCLE ON A CHASSIS DYNAMOMETER

Vehicle	Mileage	PM exhaust emissions (g/mi)
<u>Diesel</u> ^a		
1979 Caterpillar 3208	7,000	1.0
1979 Mack ENDT 676	69,000	1.9
1979 Cummins Formula 290	26,000	1.6
1977 Detroit Diesel 8V-71	60,000	2.7
PART5 prediction ^b	Calendar years 1979-1984	2.1
<u>Gasoline</u> ^C		
1973 International Harvester Stake-Bed	105,000	0.3
1975 General Motors Stake-Bed	35,000	0.5
1980 General Motors Ryder Van	<10,000	0.3
1979 Ford Van	<10,000	2.1
1979 Ford Stake Bed (same engine as above)	<10,000	0.5
PART5 prediction ^b	Calendar years 1979-1984	0.3 - 0.4

^aFrom Dietzmann et al. (1980).

bWe run the PART5 model for two years: 1979 and 1984. The assumptions used in the model for both years are: transient cycle, speed of 19.6 mph, low altitude, no inspection and maintenance, no reformulated gasoline, and PM₃₀. We report exhaust emissions only, and exclude tirewear, brakewear and indirect sulfates. For HDGVs, we got 0.33 g/mi for 1984 and 0.44 for 1979 (which we rounded to 0.3 to 0.4); for HDDV we got 2.1 g/mi for both years.

^cFrom Black et al. (1984). For each vehicle, Black et al. measured emissions at two test weights (about half of gross-vehicle weight, and about 3/4 of gross vehicle weight), and over two test cycles, the Heavy-Duty Transient Cycle (HDTC) and the Durham Road Route (DRR). We have reported the results for the heavier of the two vehicle weights, because it seemed more realistic, and for the HDTC, which was the official EPA test cycle. The DRR always produced lower PM emissions than did the HDTC, and in most cases the lighter configuration produced lower PM emissions than did the heavier configuration.

We have excluded results for a 1976 Ford with a gross vehicle weight of only 9,000 lbs.

TABLE 16-4. COMPARISON OF MOTOR VEHICLE PM EXHAUST EMISSIONS BACK-CALCULATED FROM FIELD STUDIES AND EMISSIONS CALCULATED BY THE PART5 MODEL (GRAMS/MILE)

	Pierso Brach (19	aczek		n et al. 93)		rf et al. 94)	
PM size measured	PN	110	PN.	12.5	PM ₁₀		
Gasoline vehicle emissionsa							
Study results (all PM)b	0.0	064	0.0	032	0.0)15	
Adjusted study results (exhaust) ^C	0.100		0.044		0.023		
PART5 model (exhaust) ^d	0.133		0.016		0.016		
Diesel heavy-duty vehicle emissions ^a							
Study results (all PM)b	1.4	40	1.29		0.67		
Adjusted study results (exhaust) ^C	2.7	18	2.	2.01		1.04	
PART5 model (exhaust) ^d	2.0	07	1.	63	1.	47	
Gasoline and diesel fleet ^e	20% HDDVs	7% HDDVs	6% buses	3% buses	30% HDDVs	7% HDDVs	
Study results (all PM) ^f	0.33	0.16	0.11	0.07	0.21	0.06	
Adjusted study results (exhaust) ^C	0.51	0.25	0.17	0.11	0.33	0.09	
PART5 model (exhaust)g	0.52	0.27	0.12	0.07	0.45	0.12	

^aSee the discussion of vehicle types in the notes to Table 16-5.

In all of the studies, the measured PM apparently excludes indirect or secondary PM, such as ammonium sulfate. Pierson and Brachaczek (1983: 1) state that they exclude "photochemical or 'secondary' material", and Whittorf et al. (1994) seemed to have followed the

^bThe values shown are the original authors' apportionment of *total* roadway PM emissions, including road dust and tirewear PM, to the two different vehicle classes. Generally, they did this by relating the variation in the measured PM level to the variation in the composition of the traffic.

method of Pierson and Brachaczek (1983). We suspect that this sampling method does not allow enough time for significant amounts of secondary material to form.

We assume all of the studies exclude brakewear PM, because the vehicles were cruising and hence rarely if ever braking.

Whittorf et al. (1994) measured PM_{10} , and Balogh et al. (1993) measured $PM_{2.5}$. Pierson and Brachaczek (1983) measured "airborne" PM, but it appears that some 95% of the total amount was PM_{10} .

The results in Whittorf et al. (1994) also are reported in Gertler et al. (1995).

^cTo make the three field-study estimates of total PM emissions during cruising comparable to PART5 estimates, which as discussed pertain to transient driving, we make two changes to the field-study estimates: 1) we increase the cruising emissions by 75% to make them comparable to transient emissions; and 2) we reduce total emissions by 11% to remove road dust and tirewear to make them comparable to exhaust emissions.

<u>Adjusting cruise-cycle emissions to transient-cycle emissions.</u> The objective here is to estimate what the vehicles in the three field studies would have emitted had they been following a transient cycle (as modeled in PART5) rather than cruising. To make this estimate, we first describe the transient test cycle upon which the PART5 estimates apparently are based, and then analyze the relationship between emissions during cruising, and emissions during transient driving.

Black et al. (1984) describe the heavy-duty transient cycle (HDTC) test. It is 1060 seconds with an average speed of 18.86 mph, and comprises the following three sub-cycles, one of which is repeated: i) NY non-freeway, 254 seconds, 7.56 mph average; ii) LA non-freeway, 285 seconds, 14.55 mph average; iii) LA freeway, 267 seconds, 44.93 mph average; iv) NY non-freeway again. A substantial amount of time -- over 300 seconds -- is spent at or near zero mph. (It thus appears that the HDTC is meant to be an "average" cycle.)

To adjust cruising emissions to transient emissions, we can compare emissions from the LA freeway portion of the HDTC with emissions from the entire HDTC. Dietzmann et al. (1980) report PM emission for the LA freeway sub-cycle and for whole HDTC, for four heavy-duty engines. PM emissions over the transient cycle were 10% to 60% (mid value of about 40%) higher than emissions over the LA freeway sub-cycle.

Black et al. (1984) report that the four heavy-duty gasoline trucks emit 3.3 times more HCs over the NY non-freeway cycle than the LA freeway cycle, 2.6 times more HCs over the LA non-freeway than the LA freeway, and 1.78 times more HCs over the whole HDTC than over the LA freeway cycle. They do not report PM emissions over the different sub-cycles of the HDTC, but they do report PM emissions for the HDTC versus another completely different drive cycle, the RDD. The relationship between PM-HDTC and PM-RDD is the same as the relationship between HCs-HDTC and HCs-RDD. This suggests that PM emissions would have behaved over the HDTC sub-cycles the same way that HC emissions did. This means that PM emissions in LA freeway would be 1.78 times less than in the whole HDTC. This 78% increase is similar to 10-60% increase found above.

However, vehicles cruising at constant high speed, as in the three field studies, should emit even less PM than vehicles following the LA freeway sub-cycle, which has a few transients itself. Overall, we believe that the Black et al. (1984) data and Dietzmann et al. (1980) data imply that PM emissions (from normal vehicles) during transient driving are 50% to 100% higher than PM emissions during cruising. For super-emitters, which presumably

emit most of their "excess" emissions during transient driving, this ratio probably will be higher.

Finally, we note that Gertler et al. (1995) compared HC emissions from 5 heavy-duty diesel vehicles at steady 40 mph cruise and over a 5-peak drivecycle. Each peak had acceleration, steady cruise, deceleration, and idle. The HC emissions were 60% higher in the 5-peak cycle than at 40 mph cruise.

We infer from these studies that exhaust PM emissions over the transient cycle are 50% to 100% higher than exhaust emissions during cruising; we assume that they are 75% higher.

<u>Road dust and tirewear adjustment.</u> Because our purpose here is to check the accuracy of PART5's estimates of exhaust emissions, we must deduct road-dust and tirewear PM emissions from the total emissions measured in the field studies.

The Pierson and Brachaczek (1983) study allows us to calculate vehicle emissions excluding road dust (10% of total emissions) and tirewear (1% of total emissions). We assume the same percentages of road dust and tirewear apply to the Whittorf et al. (1994) and Balogh et al. (1993) studies.

^dIn order to compare the estimates of PART5 with the results of each field study, we specified the PART5 model to replicate the conditions of each study:

	Pierson & Br.(1983)	Balogh et al. (1993)	Whittorf et al. (1994)
Year	1977	1991	1993
PM size class	PM ₁₀	PM _{2.5}	PM ₁₀
Drive cycle	cruise	cruise	cruise
Vehicle speed (mph)	55.0	40.0	55.0
I & M	no	yes	yes
Reformulated gasoline	no	no	no

Year: The year in which the measurements were taken.

PM size class: Balogh et al. (1993) measured PM_{2.5}, and Whittorf et al. (1994) measured PM₁₀. Pierson and Brachaczek (1983) measured "airborne" PM, but because about 95% of the measured PM was PM₁₀, we specified PART5 for PM₁₀.

Drive cycle: PART5 offers two choices: "cruise," and "transient". Pierson and Brachaczek (1983), and Whittorf et al. (1994), measured PM along an expressway, along which vehicles obviously are "cruising." The study site of Balogh et al. (1993) was a two-lane road on a university campus, with a 2% grade. We assume that the vehicles were cruising at steady speed as they passed the monitors. (According to the PART5 users manual [EPA, 1995c], the drive cycle affects lead emissions only. Our runs of the model confirmed this: it gives the same results regardless of the cycle.)

Vehicle speed: Pierson and Brachaczek (1983) reported that vehicles approached the Allegheny and Tuscarora sampling sites at 55 mph, and went through the tunnel at 50 to 55 mph. We assume 55 mph. We also assume the normal expressway speed of 55 mph in the Whittorf et al. (1994) study. The vehicles at the campus study site of Balogh et al. (1993) probably were traveling at 30 to 35 mph, but up a 2% grade, which we assume is equivalent to 40 mph on flat ground. (The speeds in PART5 presumably are for level ground without a tailwind. However, in any case, the speed has almost no effect on emissions.)

Inspection & Maintenance, and reformulated gasoline: We have made assumptions that we believe are appropriate for the year of the study.

^ePM emissions from traffic depends on the mix of heavy-duty diesel vehicles (HDDVs) and gasoline vehicles. In the case of Pierson and Brachaczek (1993), we consider one case with 20% HDDVs, which was the average mix in their study, and one with 7% HDDVs, which is about the national average on all roads (Table 16-5). In the case of Balogh et al. (1993), we do not know the exact percentage of buses, and so consider two cases, one with 6%, and another with 3%. In the case of Whittorf et al. (1994), we consider one case with 30% HDDVs, which was the average in the study, and one with 7% HDDVs, which as just mentioned is about the national average on all roads.

fEqual to the HDDV or bus emission rate, from the original study, multiplied by the HDDV or bus fraction, plus the gasoline-vehicle emission rate from the original study multiplied by one minus the bus or HDDV fraction.

gEqual to the HDDV or bus emission rate, from the PART5 model, multiplied by the HDDV or bus fraction, plus the gasoline-vehicle emission rate from the PART5 model multiplied by one minus the bus or HDDV fraction.

TABLE 16-5. CALCULATION OF TRAVEL FRACTIONS AND AVERAGE VEHICLE WEIGHTS, FOR USE IN THE PART5 MODEL APPLIED IN TABLE 16-4 AND TABLE 16-6

	Gase	oline veh	icles		Diesel	vehicles	
	LDGVs	LDGTs	HDGVs	LDDVs	LDDTs	HDDVs	Buses
Vehicle travel (10 ⁹ VMT) ^a	1,525	439	24	18	13	154	in HDDVs
Weight-travel (10 ⁹ ton-miles) ^a	2,382	853	217	29	28	4,198	in HDDVs
Average vehicle weight (tons) ^b	1.562	1.943	9.042	1.611	2.154	27.260	17.5
Travel fractions (VMT/total VMT)							
National average ^C	0.702	0.202	0.011	0.008	0.006	0.071	in HDDVs
Pierson and Brachaczek (1983) ^d	0.604	0.174	0.010	0.007	0.005	0.2	in HDDVs
Balogh et al. (1993) (high buses) ^e	0.710	0.204	0.011	0.008	0.006	0.000	0.06
Balogh et al. (1993) (low buses) ^e	0.733	0.211	0.012	0.009	0.006	0.000	0.03
Whittorf et al. (1994) ^f	0.529	0.152	0.008	0.006	0.005	0.3	in HDVs

^aFrom Report #10 in this social-cost series (see the list at the beginning of this document).

^bEqual to ton-miles divided by miles. Note that the value for HDDVs is consistent with Pierson and Brachaczek's (1983) estimate that the HDDVs in their experiments weighed about 30 tons on average.

^cCalculated from the VMT data in the first row. Buses are included as HDDVs here.

^dPierson and Brachaczek (1983) distinguished between gasoline-powered vehicles, and heavy diesel trucks. Apparently, they counted light-duty diesel vehicles as heavy-duty diesel trucks. In the Tuscarora experiment, 84% of the vehicles were gasoline vehicles, and 16% were diesel trucks. In the Allegheny Mountain Tunnel experiment, 76% of the vehicles were gasoline vehicles, and 24% were diesel vehicles. We simply average the two experiment sites, and assume that 20% were HDDVs as classified here. We distribute the remaining 80% across all other vehicle categories in proportion to their share of national VMT.

^eBalogh et al. (1993) state that during peak periods, traffic volumes at the study site could exceed 500 vehicles and 30 buses per hour (Balogh et al., 1993). This implies that about 6% of the vehicles were buses. However, during 30 minutes of sampling during the peak period in July, they actually counted 7 buses and 1 diesel truck, or 16 HDDVs per hour. This implies that about 3% of the vehicles were buses; we ignore the relatively small percentage of heavyduty trucks. Thus, we calculate emissions for 3% buses, and for 6% buses.

fWhittorf et al. (1994) distinguish between spark-ignition vehicles (SIVs; cars, vans, pick-up trucks, motorcycles, and heavy-duty gasoline vehicles), and heavy-duty diesel vehicles (HDDVs), including buses. They do not explicitly classify light-duty diesel vehicles, but as these account for a tiny fraction of VMT, the omission is unimportant. They report that from July 12th to July 15th 1993, about 70% of the vehicles at the study site were SIVs, and 30% HDDVs. On this basis, we assign 30% of VMT to our HDDV category, and distribute the remaining 70% across all other vehicle categories in proportion to their share of national VMT.

TABLE 16-6. CALCULATION OF TOTAL PM EMISSIONS FROM TRAFFIC, USING PART5/AP-42

	Average weight (tons) ^a	K (size scalar) ^b	silt loading (g/m ²) ^c	Roadway emissions (g/mi) ^d
National average (expressway, PM10) ^e	3.55	7.30	0.02	0.47
Pierson & Brachaczek (1983) (expressway)	6.84	7.30	0.02	1.26
Balogh et al. (1993) (6% buses)	2.68	3.30	0.30	0.81
Balogh et al. (1993) (3% buses)	2.21	3.30	0.30	0.61
Whittorf et al. (1994) (expressway)	9.39	7.30	0.02	2.03

Note: PART5 uses the same equation as AP-42, so the two estimates are identical.

aCalculated as:

$$AW s = \sum_{v} TF_{v, s} \cdot VW_{v}$$

where:

 AW_S = the average vehicle weight in study S (tons)

 $TF_{V,S}$ = the travel fraction by vehicle type V in study S (Table 16-5)

 VW_V = the average weight of vehicle type V (Table 16-5)

^bThis is the factor "K" in the emission-factor equation from AP-42 (EPA, 1995a) -- our equation D2 in the text above. K scales the results to the particle size class of interest: K is 7.3 for PM₁₀, and 3.3 for PM_{2.5}.

^cAP-42 (EPA, 1995a) recommends using a value of 0.02 g/m² for expressways, and 0.30 for non-freeways with an average daily traffic that exceeds 5000 during the period July to December. In a calculation, Balogh et al. (1993, p. 31) use 480 vehicles/hour, which corresponds to 11,520 vehicles/day.

dCalculated using equation D2 from the text.

eThis case estimates PM₁₀ emissions from the national average mix of vehicles on freeways. We assume that the national average mix of vehicles on freeways is equal to the national average mix on all roads (Table 16-5). If one compares this case with the Pierson and Brachaczek (1983) case, and the Whittorf et al. (1994) case, one can see the effect of the different vehicle mix on emissions, because the only difference between these cases is the assumed vehicle mix (Table 16-4).

TABLE 16-7. COMPARISON OF EMFAC7F AND MOBILE5A ESTIMATES OF PM EMISSIONS

Model: pollutant source	Pollutant	LDGVs g/mi 1990	HDDVs g/mi 1990	Buses g/mi 1990
PART5: exhaust	PM ₁₀ , TSP	0.02	1.93	1.86
PART5: indirect sulfate	PM ₁₀ , TSP	0.03	0.79	0.77
PART5: brakewear	PM_{10} , TSP	0.01	0.01	0.01
PART5: tirewear	PM_{10}	0.01	0.04	0.01
PART5: tirewear	TSP	0.02	0.07	0.02
PART5: total	PM _{1O}	0.06	2.77	2.65
PART5: total	TSP	0.07	2.81	2.66
EMFAC7F: summer exhaust	TSP?	0.01a	3.51	4.67
EMFAC7F: summer tirewear	TSP?	0.40	1.32	1.32
EMFAC7F: summer total	TSP?	0.21	4.17	5.33

TSP = total suspended particulate.

EMFAC7F estimates only "exhaust particles" and "tirewear", without specifying the size of the estimated emissions. PART5 estimates emissions of PM10 or smaller, but nothing larger. We assumed that EMFAC7F is estimating TSP (about PM30), and converted the PART5 output to TSP for comparison. The conversion is straightforward, because according to EPA's AP-42 Volume II, Appendix L (1985), 100% of diesel exhaust PM, and 98% of brakewear PM, is less than PM10. Hence, for the exhaust, indirect sulfate, and brakewear factors above, the TSP emission rate equals the PM10 emission rate. PM from tirewear generally is larger than PM from brakewear; according to Williams et al. (1995: 89), 58.5% of tirewear particulates are PM10.

In the PART5 estimates we assume a transient cycle, 19.6 mph, no reformulated gasoline and no inspection and maintenance.

^aFor vehicles with a catalytic converter. Vehicles without a catalytic converter emit 0.04 g/mi.

TABLE 16-8. MOTOR-VEHICLE AND FUGITIVE-DUST EMISSIONS OF PM IN URBAN AREAS OF THE U.S. IN 1990, ACCORDING TO THE OFFICIAL EPA EMISSION INVENTORY (MILLION TONS)

	PM ₁₀	Coarse PM ₁₀	PM2.5
Motor vehicles	0.241	0.045	0.195
Fugitive dust			
Paved roads	4.108	2.386 (3.081)a	1.722 (1.027) ^a
Unpaved road	3.894	2.868	1.026
Wind erosion	0.630	0.385	0.246
Construction	7.749	7.591 (6.199) ^b	0.158 (1.550) ^b
Agricultural tillage	1.364	0.718	0.646
Agricultural livestock	0.116	0.058	0.058
Total fugitive dust	17.862	14.007	3.856

These estimates are from our analysis of data reported by the EPA (1995d). These are the uncorrected, official EPA estimates, not our corrected estimates (i.e., these are the emissions prior to the application of the correction factors of Table 16-1). Also, in the case of motor vehicles, they are direct emissions only; they do not include secondary particulates.

^aAfter the EPA produced the emissions inventory that we use here, it revised its estimate of the fraction of paved-road dust that is PM_{2.5} (Barnard, 1996). The values in parentheses show what the official emissions inventory that we use would have been had the EPA used the new PM_{2.5} fraction when it developed the inventory that we use here.

^bAfter the EPA produced the emissions inventory that we use in our apportioning analysis, it revised its estimate of the fraction of construction dust that is PM_{2.5} (Barnard, 1996). The values in parentheses show what the official emissions inventory that we use would have been had the EPA used the new PM_{2.5} fraction when it developed the inventory that we use here.

Table 16-9. Source contributions to ambient PM_{10} , as estimated by chemical mass-balance studies

		S	ource c	ontrib	ution (to PM ₁	0 conce	entratio	n
Site (reference)	PG	PC	PMV	PV	SAS	SAN	M1	M2	N
Arizona									
Corona de Tucson (Chow et al., 1993)	17.0	0.0	1.6	0.0	1.9	0.0	0.0	0.0	(
Craycroft (Chow et al., 1993)	13.0	0.0	8.3	0.0	0.7	0.6	1.2 ^a	0.0	(
Downtown Tucson (Chow et al., 1993)	26.0	5.1	14.0	0.0	1.0	0.2	1.3 ^a	0.0	(
Hayden 1 (Garfield) (Ryan et al., 1988)	5.0	2.0b	0.0	0.0	4.0	0.0	74.0 ^C	5.0d	1
Hayden 2 (jail) (Ryan et al., 1988)	21.0	4.0^{b}	0.0	0.0	4.0	0.0	28.0 ^C	0.0	1
Orange Grove (Chow et al., 1993)	20.0	0.0	15.0	0.0	0.4	0.4	0.0	0.0	(
Phoenix (central) (Chow et al., 1991)	33.0	0.0	25.0	2.3	0.2	2.8	0.0	0.0	0
Phoenix (west) (Chow et al., 1991)	30.0	0.0	25.0	10.0	0.4	3.1	0.0	0.0	0
Phoenix (Estrella Park) (Chow et al., 1991)	37.0	0.0	10.0	0.9	1.6	0.0	0.0	0.0	0
Phoenix (Gunnery Rg.) (Chow et al., 1991)	20.0	0.0	5.5	0.0	1.0	0.0	0.0	0.0	0
Phoenix (Pinnacle Pk.) (Chow et al., 1991)	7.0	0.0	2.9	1.0	0.9	0.0	0.0	0.0	0
Rillito (Chow et al., 1993)	42.7	13.8 ^b	1.2 ^f	0.0	0.0	0.0	11.6g	0.0	(
South Scottsdale (Chow et al., 1991)	25.0	0.0	19.0	7.4	0.6	3.6	0.0	0.0	0
California									
Anacapa Island (Chow et al., 1996)	2.2	0.0	4.9	0.0	3.4	1.0	9.6 ^h	0.0	0
Anaheim 1986 average (Kao and Friedlander, 1995)	27.4	0.0	14.7	0.0	4.75	9.56	0.91 ^j	1.51 ^h	0
Anaheim (Summer) (Watson et al., 1994b)	11.4	0.0	8.5	0.0	9.0	2.9	0.0j	6.5 ^h	0
Anaheim (Fall) (Watson et al., 1994b)	13.2	0.0	37.2	0.0	3.7	38.5	0.0j	3.1h	0
Azusa (Summer) (Watson et al., 1994b)	34.9	0.0	15.9	0.0	11.4	6.1	0.0j	5.7 ^h	0
Bakersfield (Watson et al., 1989)	27.4	3.0	5.5	9.6 ^l	4.1	0.0	0.5j	0.0	0
Bakersfield (Chow et al., 1992a)	42.9	1.6	7.7	6.5	5.5	12.7	$1.0^{\rm{m}}$	1.5 ⁿ	0
Burbank 1986 average (Kao and Friedlander, 1995)	21.3	0.0	17.7	0.0	4.19	10.0	1.02 ^j	1.00 ^h	0
Burbank (Summer) (Watson et al., 1994b)	14.0	0.0	17.0	0.0	12.4	6.5	0.0j	5.7 ^h	0
Burbank (Fall) (Watson et al., 1994b)	11.0	0.0	39.1	0.0	3.1	25.1	0.0j	1.9 ^h	0
Chula Vista 1 (Bayside) (Watson et al., 1989)	6.7	0.0	0.8	0.0	7.5	0.0	$0.4\dot{J}$	2.7 ^h	2
Chula Vista 2 (Del Ray) (Watson et al., 1989)	8.2	0.3	1.5	0.0	8.9	0.0	0.6j	1.8 ^h	0

TABLE 16-9 (CONTINUED).

		S	ource	contril	bution	to PM ₁₀	conce	ntrati
Site (reference)	PG	PC	PMV	PV	SAS	SAN	M1	M2
Chula Vista 3 (Watson et al., 1989)	9.7	0.3	1.4	0.0	8.2	0.0	0.6 ^j	1.7 ^h
Claremont (Summer) (Watson et al., 1994b)	19.4	0.0	14.4	0.0	9.5	6.3	0.0j	4.7 ^h
Crows Landing (Chow et al., 1992a)	32.2	0.0	2.2	3.4	2.8	6.5	0.5^{m}	1.5 ⁿ
Downtown L. A. 1986 average (Kao & Friedlander, 1995)	26.1	0.0	15.4	0.0	5.66	10.4	1.01 ^j	1.81 ^h
Downtown Los Angeles (Summer) (Watson et al., 1994b)	12.7	0.0	16.2	0.0	13.0	4.4	0.0 ^j	6.5 ^h
Downtown Los Angeles (Fall) (Watson et al., 1994b)	9.4	0.0	41.1	0.0	3.9	27.5	0.0j	1.8 ^h
Fellows (Chow et al., 1992a)	29.0	1.4	2.1	3.4	5.1	7.5	7.0 ^m	1.4 ⁿ
Fresno (Watson et al., 1989)	17.1	0.7	4.0	9.2 ^l	1.3	0.0	0.1 ^j	0.0
Fresno (Chow et al., 1992a)	31.8	0.0	6.8	5.1	3.6	10.4	0.3^{m}	1.0 ⁿ
Hawthorne 1986 average (Kao and Friedlander, 1995)	16.7	0.0	15.7	0.0	5.44	7.80	1.68 ^j	4.16 ^h
Hawthorne (Summer) (Watson et al., 1994b)	6.8	0.0	5.7	0.0	14.2	0.6	0.0 ^j	7.0 ^h
Hawthorne (Fall) (Watson et al., 1994b)	8.9	0.0	35.1	0.0	5.1	20.4	0.0 ^j	3.7h
Indio (Chow et al., 1993)	33.0	3.0	4.4	7.1	3.6	4.1	0.2 ^j	1.0 ^h
Kern Wildlife Refuge (Chow et al., 1992a)	15.1	2.0	2.2	4.0	3.3	1.5	$0.5^{\rm{m}}$	1.5 ⁿ
Lennox (Watson et al., 1989)	16.0	0.1	4.6^{i}	0.0	7.6	7.9	0.2j	3.1h
Long Beach 1986 average (Kao and Friedlander, 1995)	21.4	0.0	12.8	0.0	5.74	9.08	1.56 ^j	3.28 ^h
Long Beach (Summer) (Watson et al., 1994b)	11.1	0.0	6.3	0.0	10.9	0.8	0.1 ^j	6.2 ^h
Long Beach (Fall) (Watson et al., 1994b)	11.3	0.0	42.8	0.0	3.8	23.2	0.0 ^j	2.7h
Magnolia 1988 average (Chow et al., 1992b)	31.7	0.0	11.2	0.0	4.9	19.7	0.3j	1.2 ^h
Palm Springs (Chow et al., 1993)	16.4	1.4	2.3	5.1	3.7	4.2	0.1 ^j	0.5h
U. C. Riverside 1988 average (Chow et al., 1992b)	32.6	0.0	7.0	0.0	4.8	21.4	0.3j	1.3h
Rubidoux 1986 average (Kao and Friedlander, 1995)	42.5	0.0	11.0	0.0	5.54	20.4	0.23j	1.61 ^h
Rubidoux (Summer) (Watson et al., 1994b)	34.9	4.5	17.3	0.0	9.5	27.4	0.0j	5.1 ^h
Rubidoux (Fall) (Watson et al., 1994b)	19.2	16.1	30.3	0.0	2.1	31.6	0.0j	1.1 ^h
Rubidoux 1988 average (Chow et al., 1992b)	48.0	0.0	10.2	0.0	5.3	21.7	0.4j	1.5 ^h
San Jose (4th St.) 12/16/91 to 2/24/92 (Chow et al., 1995)	13.1	0.0	9.2	31.3	2.3	13.3	0.9h	0.0
San Jose (San Carlos St.) (Chow et al., 1995)	11.8	0.0	8.9	31.3	2.1	12.8	0.7 ^h	0.0
San Nicolas Island (Summer) (Watson et al., 1994b)	1.6	0.0	0.9	0.0	3.7	0.5	0.0 ^j	4.3h
Santa Barbara (Chow et al., 1996)	9.5	0.0	14.7	0.0	3.2	1.0	6.4 ^h	0.0

TABLE 16-9 (CONTINUED).

		S	ource o	ontrib	ution t	o PM ₁	0 conce	ntratio
Site (reference)	PG	PC	PMV	PV	SAS	SAN	M1	M2
Santa Barbara (Gaviota) (Chow et al., 1996)	3.2	0.0	5.1	0.0	2.8	0.5	6.3 ^h	0.0
Santa Maria (Chow et al., 1996)	7.4	0.0	7.6	0.0	3.1	1.4	5.7 ^h	0.0
Santa Ynez (Chow et al., 1996)	4.6	0.0	6.8	0.0	2.2	0.6	4.0h	0.0
Stockton (Chow et al., 1992a)	34.4	0.5	5.2	4.8	3.1	6.9	0.7^{m}	1.8 ⁿ
Upland (Watson et al., 1989)	25.4	0.4 ^j	4.1^{i}	0.0	6.4	14.5	0.6j	0.6 ^h
Vandenberg AFB (Watt Road) (Chow et al., 1996)	1.5	0.0	3.2	0.0	1.9	1.0	9.3h	0.0
Colorado								
Telluride 1 (Central) (Dresser & Baird, 1988)	32.0	0.0	0.0	98.7	0.0	0.0	61.3P	0.0
Telluride 2 (Society Turn) (Dresser & Baird, 1988)	12.1	0.0	0.0	7.3	0.0	0.0	7.3P	0.0
Idaho								
Pocatello (Chow et al., 1993)	8.3	7.59	0.1	0.0	0.0	0.0	0.0	0.0
Illinois								
South Chicago (Watson et al., 1989)	27.2	2.4	2.8	0.0	15.4 ^s	0.0	15.1 ^t	2.2 ^u
Southeast Chicago (Chow et al., 1993)	14.7^{V}	0.0	0.9 ^f	0.0	5.6	0.0	0.8^{t}	0.3h
Nevada								
Reno (Watson et al., 1988a)	14.9	0.0	10.0	1.9	1.3	0.6	0.0	0.0
Sparks (Watson et al., 1988a)	15.1	0.2	11.6	13.4	2.7	0.9	0.0	0.0
Verdi (Watson et al., 1988a)	7.8	0.0	4.0	1.1	0.9	0.1	0.0	0.0
Ohio								
Follansbee (Chow et al., 1993)	10.0	0.0	35.0	0.0	16.0		9.3t	0.0
Mingo (Chow et al., 1993)	12.0	0.0	14.0	4.1	15.0		3.4^{t}	11.0 ^X
Steubenville (Chow et al., 1993)	8.3	0.0	14.0	0.8	14.0		3.8 ^t	5.0 ^x
Pennsylvania								
Philadelphia 7/14/82 to 8/13/82 (Dzubay et al., 1988)	9.57	0	2.6	0.0	20.0	0.0	0.7У	$0.3^{\mathbf{Z}}$

PG = primary geological; PC = primary construction; PMV = primary motor-vehicle; PV = primary vegetative burning; SAS = secondary ammonium sulfate; SAN = secondary ammonium nitrate; M1 = miscellaneous source 1; M2 = miscellaneous source 2; M3 = miscellaneous source 3; M4 = miscellaneous source 4; PM10 = the concentration of PM10 actually measured, as opposed to the concentration predicted by the CMB model. The predicted concentration is equal to the sum of the predicted source concentrations shown here.

^aSmelter background aerosol.

^bCement plant sources: gypsum and lime handling (Ryan et al., 1988); kiln stacks, gypsum pile, and kiln area (Chow et al., 1993).

^cCopper ore and ore crusher.

dCopper ore tailings.

^eCopper smelter building.

fHeavy-duty diesel exhaust emission.

gBackground aerosol.

^hMarine aerosol (Kao and Friedlander, 1995) or marine aerosol, road salt, and sea salt plus sodium nitrate (all others).

iMotor vehicle exhaust from diesel and leaded gasoline.

jResidual oil combustion.

kSecondary organic carbon.

^lBiomass burning.

^mPrimary crude oil.

 $n_{NaCl} + NaNO_3$.

OLime (Chow et al., 1992b) or lime/gypsum mining operations (Kao and Friedlander, 1995)

PRoad sanding material.

9Asphalt industry.

^r Phosphorus/phosphate industry.
^S Regional sulfate.
^t Steel mills.
^u Refuse incinerator.
^V Local road dust, coal yard road dust, steel-haul road dust.
WIncineration.
^X Unexplained mass.
YMunicipal incinerator.
^Z Antimony roaster.
aaOther sources.

Table 16-10. Source contributions to ambient $PM_{2.5}$, as estimated by chemical mass-balance studies

		Source contribution to PM _{2.5} : concentration C (μ g/m ³),									, and	l fi					
Site (reference)	F	PG		PC		MV		PV		AS		AN		Л1		Л2	
	С	F	С	F	С	F	С	F	С	F	С	F	С	F	С	F	C
Arizona																	
Phoenix (central) (Chow et al., 1991)	3.1	0.09	0.0	0.00	20.0	0.80	2.3	1.00	0.2	1.00	2.6	0.93	0.0	0.00	0.0	0.00	0.0
Phoenix (west) (Chow et al., 1991)	2.0	0.07	0.0	0.00	20.0	0.80	10.0	1.00	0.4	1.00	2.9	0.94	0.0	0.00	0.0	0.00	0.0
Phoenix (Estrella Park) (Chow et al., 1991)	4.3	0.12	0.0	0.00	9.7	0.97	0.9	1.00	1.2	0.75	0.0	0.00	0.0	0.00	0.0	0.00	0.0
Phoenix (Gunnery Rg.) (Chow et al., 1991)	2.4	0.12	0.0	0.00	4.5	0.82	0.0	0.00	1.0	1.00	0.0	0.00	0.0	0.00	0.0	0.00	0.0
Phoenix (Pinnacle Pk.) (Chow et al., 1991)	0.7	0.10	0.0	0.00	2.6	0.90	1.0	1.00	0.8	0.89	0.0	0.00	0.0	0.00	0.0	0.00	0.0
South Scottsdale (Chow et al., 1991)	2.1	0.08	0.0	0.00	14.0	0.74	7.4	1.00	0.6	1.00	3.6	1.00	0.0	0.00	0.0	0.00	0.0
California ^a		ŀ		ŀ		ļ		ļ		ŀ		1		1			
Bakersfield (Chow et al., 1992a)	2.5	0.06	0.2	0.10	9.2	1.21	4.5	0.70	4.8	0.86	14.4	1.13	0.3	0.18	0.0	0.00	1.
Crows Landing (Chow et al., 1992a)	2.4	0.08	0.0	0.00	2.2	1.00	2.1	0.62	2.6	0.96	7.6	1.10	0.8	0.55	1.3	1.03	0.
Fellows (Chow et al., 1992a)	3.4	0.12	0.2	0.17	1.7	0.80	1.9	0.57	4.9	0.97	9.9	1.32	0.6	0.40	1.4	0.94	7.3
Fresno (Chow et al., 1992a)	2.3	0.07	0.0	0.00	9.2	1.36	5.9	1.16	3.5	0.97	12.4	1.19	0.5	0.49	0.4	5.00	0.3
Kern Refuge (Chow et al., 1992a)	2.0	0.13	0.0	0.00	2.3	1.03	2.0	0.50	3.2	0.98	11.1	1.34	0.5	0.32	0.3	0.38	0.7
Los Angeles (Watson et al., 1989)	3.2	0.13	0.0	0.00	5.5 ^b	1.02	0.0	0.00	7.3	0.96	7.0	0.63	0.2 ^c	0.15	11.1	1.41	0.0
Stockton (Chow et al., 1992a)	4.6	0.13	0.0	0.00	7.2	1.39	3.7	0.78	2.8	0.91	8.4	1.21	0.7	0.42	0.1	n.a.	0.5
Nevada ^a		ŀ		!		!		!		!		I		I			
Las Vegas (Watson et al, 1989)	0.0	n.a.	0.0	n.a.	8.9	n.a.	0.1	n.a.	0.0	n.a.	0.0	n.a.	0.0	n.a.	0.0	n.a.	0.6
Pennsylvania		I		!		!		!		!		I		I			
Philadelphia (Dzubay et al., 1988)	0.9	0.09	0.0	0.00	2.6	1.00	0.0	0.00	19.4	0.97	0.0	0.00	0.7 ^d	0.94	0.1 ^e	0.29	4.

PG = primary geological; PC = primary construction; PMV = primary motor-vehicle; PV = primary vegetative burning; SAS = secondary ammonium sulfate; SAN = secondary ammonium nitrate; M1 = NaCl + NaNO3, except as noted; M2 = secondary organic carbon, except as noted; M3 = primary crude oil, except as noted; M4 = miscellaneous source 4; PM₁₀ = the concentration of PM₁₀ actually measured, as opposed to the concentration predicted by the CMB model (the predicted concentration is equal to the sum of the predicted source concentrations shown here); C = concentration of PM_{2.5} due to source, in μ g/m³; F = PM_{2.5} fraction of PM₁₀ from source (equal to PM_{2.5} concentration shown in this table divided by PM₁₀ concentration for same site and source in Table 16-9).

^aAll (published) measured categories are included for California and Nevada.

bIncludes motor vehicle exhaust from diesel and leaded gasoline.

^CMarine aerosol.

dMunicipal incinerator.

^eAntimony roaster.

fOther sources.

TABLE 16-11. THE RATIO OF ROAD-DUST PM TO MOTOR-VEHICLE EXHAUST PM: CMB SOURCE APPORTIONING VERSUS THE EMISSIONS INVENTORY

	PM ₁₀	Coarse PM ₁₀	PM2.5
CMB: geologic/motor vehicles ^a	1.7	19.6	0.30
Urban OEI: paved roads/motor vehicles ^b	17.0	52.0 (68.5)	8.9 (5.3)
OEI/CMB	9.9	2.7 (3.5)	29.7 (17.7)

^aCalculated from the data of Tables 16-9 and 16-10. In each PM size category, the ratio is equal to the sum of all $\mu g/m^3$ concentrations in the "geologic" category divided by the sum of all $\mu g/m^3$ concentrations in the "motor-vehicle" category. That is, we add up the concentrations first, then take the ratio.

^bCalculated from the data of Table 16-8. The values in parentheses correspond to the newly revised emission inventory estimates of Table 16-8. As we mention above, the emission inventory that we use does not reflect these recent revisions.

TABLE 16-12. ATMOSPHERIC RESIDENCE TIME AS A FUNCTION OF PARTICLE SIZE

Particle size	Particles be	elow 1.5 km		l troposphere up oopause		
μm	days residence ^a	ratio with 0.1 μm	days residence ^a	ratio with 0.1 μm		
0.1	7.61	1.00	18.47	1.00		
0.3	7.90	0.96	20.26	0.91		
0.5	7.85	0.97	19.95	0.93		
1	7.53	1.01	18.00	1.03		
1.5	7.04	1.08	15.41	1.20		
2	6.44	1.18	12.82	1.44		
2.5	5.81	1.31	10.54	1.75		
3.00	5.19	1.47	8.66	2.13		
3.50	4.61	1.65	7.15	2.58		
4.00	4.08	1.87	5.95	3.10		
4.50	3.61	2.11	5.00	3.69		
5.00	3.20	2.38	4.25	4.35		
6.00	2.53	3.01	3.14	5.88		
7.00	2.03	3.75	2.41	7.66		
8.00	1.65	4.61	1.89	9.77		
9.00	1.36	5.60	1.53	12.07		
10.00	1.14	6.68	1.25	14.78		

^aResidence time is based on the following empirical equation from Wiman et al. (1990: Figure 2):

$$\tau = \left[\left(\frac{1}{K} \right) \cdot \left(\frac{r}{R} \right)^2 + \left(\frac{1}{K} \right) \cdot \left(\frac{R}{r} \right)^2 + \frac{1}{\tau_{wet}} \right]^{-1}$$

where:

 τ = residence time (sec onds, converted to days in the Table)

$$K = constan \ t = 1.28 \cdot 10^8$$

 $r = particle \ radius \ (\mu m)$

 $R = 0.3 \mu m$

 $\tau_{wet} = 6.9 \cdot 10^5 \ (below \ 1.5 \ km)$

 $\tau_{wet} = 1.8 \cdot 10^6$ (middle troposphere up to tropopause).

TABLE 16-13. COMPARISON OF MOTOR VEHICLE PM EMISSIONS BACK-CALCULATED FROM FIELD STUDIES AND EMISSIONS CALCULATED BY PART5/AP-42-- STUDIES OUTSIDE OF THE U.S. MIDWEST (GRAMS/MILE)

				grams/mile		Ratio
note	Location and type of road	Year of study	PM size	Field study	AP-42	AP-42: study
a	Pennsylvania, expressway tunnels	1970-79	PM10	0.331	1.26	3.8
b	Seattle WA, 2-lane road (low bus)	1991	PM2.5	0.070	0.61	8.7
b	Seattle WA, 2-lane road (high bus)	1991	PM _{2.5}	0.107	0.81	7.6
С	Baltimore MD, expressway tunnel	1993	PM ₁₀	0.212	2.03	9.6
d	Los Angeles CA, freeway	1974	PM5	0.100	0.28	2.8
d	Los Angeles CA, freeway	1974	>PM5	0.071	0.34	4.8
e	Davis CA, freeway (low AP-42)	1994	PM ₁₀	0.029	0.09	3.1
e	Davis CA, freeway (high-AP-42)	1994	PM ₁₀	0.029	0.40	13.9
f	Sacramento CA, suburban intersection (low study, low AP-42)	1994	PM ₁₀	0.209	1.56	7.5
f	Sacramento CA, suburban intersection (low study, high AP-42)	1994	PM ₁₀	0.209	8.53	40.8
f	Sacramento CA, suburban intersection (high study, low AP-42)	1994	PM ₁₀	2.092	1.56	0.7
f	Sacramento CA, suburban intersection (high study, high AP-42)	1994	PM ₁₀	2.092	8.53	4.1
g	Phoenix AZ (low study, low AP-42)	1994	PM10	0.004	0.37	91.5
g	Phoenix AZ (low study, high AP-42)	1994	PM10	0.004	2.56	641.1
g	Phoenix AZ (high study, low AP-42)	1994	PM10	0.008	0.37	44.3
g	Phoenix AZ (high study, high AP-42)	1994	PM ₁₀	0.008	2.56	310.5

^aFrom Table 16-6 (Pierson and Brachaczek, 1983).

bFrom Table 16-6 (Balogh et al., 1993).

^cFrom Table 16-6 (Whittorf et al., 1994)

dFrom Cahill et al. (1994), who report the results of studies they did 20 years earlier. ">PM5" means "greater than PM5". The AP-42 g/mi estimates are theirs, not ours, and apparently are from the 1974 emission-factor equation, not from the current emission-factor equation.

^eFrom Cahill et al. (1994), who measured PM₁₀ levels across Interstate 80 in the Central Valley of California. In the "low AP-42" comparison, Cahill et al. (1994) specify the current AP-42 emission factor equation (EPA, 1995a; equation D2 here) with weight (W) equal to 1.5 tons, and silt loading (sL) equal to 0.011 g/m². In the "high AP-42" comparison, they specify the current AP-42 emission factor equation with weight W equal to 2.5 tons, and silt loading sL equal to 0.034 g/m².

fFrom Cahill et al. (1994), who measured PM₁₀ levels across a busy intersection in South Sacramento. The "high study" value of 2.1 g/mi is based on the total measured mass of PM. The "low study" value of 0.21 g/mi is based on the mass of the individually identified compounds. We believe that the "high study" estimates, of total mass, are the most accurate.

In the "low AP-42" comparisons, Cahill et al. (1994) specify the current AP-42 emission factor equation (EPA, 1995a; equation D2 here) with weight (W) equal to 1.5 tons, and silt loading (sL) equal to 0.90 g/m². In the "high AP-42" comparisons, they specify the current AP-42 emission factor equation with weight W equal to 2.5 tons, and silt loading sL equal to 3.8 g/m^2 .

gFrom Barnard (1996), who reports the results of experiments in Phoenix, Arizona, in which roadside PM was measured and a range of emission rates was back-calculated with a dispersion model. The lowest emission rate was 0.004 g/mi (we refer to this as "low study"), and the highest was 0.00826 g/mi (we refer to this as "high study).

In the "low AP-42" comparisons, Barnard (1996) specifies the current AP-42 emission factor equation (EPA, 1995a; equation D2 here) with weight (W) equal to 3.0 tons, and silt loading (sL) equal to $0.02~\rm g/m^2$. In the "high AP-42" comparisons, he specifies the current AP-42 emission factor equation with weight W equal to 3.0 tons, and silt loading sL equal to $0.40~\rm g/m^2$.

TABLE 16-14. SIZE DISTRIBUTION OF PARTICLES FROM VARIOUS FUGITIVE DUST SOURCES (PERCENT OF PARTICLES BELOW INDICATED SIZE)

	PM _{1.0}	PM2.5	PM10	PM _{2.5} /PM ₁₀
Paved roads (Fresno, California)	3.2	9.2	47.5	0.19
Unpaved roads (Bakersfield, California)	3.1	8.8	55.3	0.16
Agricultural soil (Stockton, California)	3.6	10.8	55.5	0.19
Soil/gravel (Visalia, California)	4.7	14.0	34.5	0.41
Alkaline lake bed (Owens Lake, California)	6.9	13.2	51.3	0.26

From Chow et al. (1994a).

TABLE 16-15. ESTIMATES OF CONTRIBUTION TO AIR QUALITY, RELATIVE TO CONTRIBUTION OF LDVS, PER KG OF EMISSIONS, BASED ON SIMPLE DISPERSION MODELING: ASSUMED VALUES OF INPUT PARAMETERS

	distance from source to receptor (r) km					angle	stack height (h _S) (meters)	
	agriculture monitor		urban monitor		(degrees)			
	Low	High	Low	High	Low	High	Low	High
Light-duty vehicles	20	15	8	5	0	0	0.3	0.3
Heavy-duty vehicles	16	12	9	4	0	0	3.5	2.5
Fuel combustion: electric utilities	15	20	25	30	0	0	51	122
Fuel combustion: industrial	15	20	15	20	0	0	12	25
Fuel combustion: other (mainly residential wood combustion)	15	20	8	6.5	0	0	3	7
Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes	15	20	15	20	0	0	6	18
Solvent utilization, storage and transport, and waste disposal and recycling	15	20	10	15	0	0	3	7
Non-road vehicles (trains, tractors, ships, planes, etc.)	15	20	10	15	0	0	3.0	4.5
Natural sources (e.g., wind erosion and wildfires)	5	7.5	25	30	0	0	0.1	1.0
Agriculture and forestry, and managed burning	1.5	3.5	25	30	0	0	0.1	1.0
Paved-road dust	20	15	10	6.5	0	0	0.1	0.1
Unpaved-road dust	7.5	12	30	25	0	0	0.1	0.1
Other fugitive dust (mainly construction)	15	20	8	6.5	0	0	0.1	0.5

These assumptions were made on the basis of the following general considerations:

Distance from source to receptor (r), urban monitors (analysis of health effects and visibility): We start by assuming that on average, LDVs in urban areas are several km from the urban air-quality monitors. (Air-quality monitors typically are located in relatively polluted parts of urban

areas. In such places, the density of motor vehicles usually is fairly high.) Then, to estimate the distance for HDVs, relative to the distance for LDVs, we consider two opposing factors. First, in 1990, the ratio of urban vehicle miles of travel (VMT) to total VMT was higher for LDVs than for HDVs (61% for passenger cars and 2-axle 4-tire trucks, versus 35% for "combination trucks") (FHWA, 1991). This means that HDVs themselves on average were further from urban monitors than were LDVs. However, we are interested not in the location of HDVs and LDVs per se, but rather in the location of their emissions. HDVs emit considerably more particulates (the pollutant most damaging to health and visibility) per mile of urban driving, including idling, than per mile of rural driving. LDVs also emit more in urban than in rural driving, but the difference probably is not as pronounced as the difference with HDVs. Considering both factors (VMT, and per-mile emissions), we assume that, on average, HDV *emissions*, relative to LDV emissions, could range from being are a bit closer to the monitor to a bit further from it.

We assume that power plants and to a somewhat lesser extent heavy industrial sources are located outside of urban areas and hence far from the air quality monitors. Solvent and waste sources, and non-road vehicles, are much closer to urban monitors, but probably not as close as are motor vehicles, on average. Thus, we assume that r_0 for solvents etc. and for non-road vehicles exceeds r_0 for motor vehicles. On the other hand, residential combustion sources, and construction sources, presumably are concentrated as much in urban and suburban areas as are LDV emissions. We assume that r_0 for these sources, relative to r_0 for motor vehicles, ranges from being the same to slightly greater.

Emissions from paved roads are a function of the silt loading on the road as well as the amount of traffic. If, as seems likely, silt loadings are higher in suburban and ex-urban areas than in central cities, then emissions from paved roads typically are a bit further from monitors than are emissions from vehicles themselves. Emissions from natural sources, agriculture, and unpaved roads generally occur outside or at the fringe of urban areas.

Distance from source to receptor (r), agricultural monitors (analysis of crop damages): Agricultural monitors of course are located in agricultural areas, which typically are outside of urban areas. Hence, we would expect agricultural monitors to be relatively close to natural sources, agricultural sources, and unpaved roads. We assume that heavy trucks are disproportionately close to agricultural areas.

Angle between wind vector and source-receptor vector (θ) . For the purpose of estimating the contribution to pollution of each source relative to the contribution of motor vehicles, we have assumed that, on average, the wind angle is the same for all sources. Furthermore, we find it most straightforward to analyze the case in which the receptor is directly downwind of the source. With these two assumptions, the wind angle is zero in all cases.

Stack height (h_s). For LDVs, HDVs, and non-road mobile sources, the estimates are of the distance from the ground to the top of the tailpipe. We consider that some HDVs (e.g., buses) have bottome exhausts, whereas others have top exhausts. For fuel combustion (all three categories), chemicals etc., and solvents etc., we use the 50% (low-cost) and 75% (high-cost) values from Table 16-17, except that we have substituted our own estimate of the stack height for fuel-combustion: other, in the high-cost case. We have done this because most of the emissions in this category are from residential chimneys, which are not included in the EPA (1995d) statistics, and which undoubtedly have a stack height of less than 18 m (the 75%)

value in the EPA [1995d] statistics). On the basis of our observations, we assume that residential chimneys are not more than 20-25 feet high. We assume that natural sources and agricultural sources are at or near ground level. (Flames, from wildfires or managed fires, are a few meters high typically, but inasmuch as emissions from fires account for but a minor fraction of total emissions in these categories, the weighted-average height -- that is, the average of 0 for dust sources, and a few meters for fires -- probably will not exceed a meter, which is our high case.) Dust from roads of course is emitted at ground level. Dust from construction activities presumably is emitted at or very near ground level.

TABLE 16-15 (CONTINUED)

	stack-gas velocity (v _S)			ick ter (d _S)	source velocity (vg)	
	(m/sec)		(meters)		(m/sec)	
	Low	High	Low	High	Low	High
Light-duty vehicles	0.0	0.0	0.07	0.07	15.5	15.5
Heavy-duty vehicles	2.0	1.0	0.15	0.10	11.2	11.2
Fuel combustion: electric utilities	15.5	25.3	3.40	5.30	0.0	0.0
Fuel combustion: industrial	6.5	15.2	0.80	1.70	0.0	0.0
Fuel combustion: other (mainly residential wood combustion)	4.0	5.3	0.20	1.20	0.0	0.0
Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes	4.0	4.0	0.20	0.80	0.0	0.0
Solvent utilization, storage and transport, and waste disposal and recycling	4.0	4.0	0.20	0.40	0.0	0.0
Non-road vehicles (trains, tractors, ships, planes, etc.)	1.0	2.0	0.15	0.20	7.0	11.0
Natural sources (e.g., wind erosion and wildfires)	1.0	2.0	0.50	1.00	0.0	0.0
Agriculture and forestry, and managed burning	1.0	2.0	0.50	1.00	0.0	0.0
Paved-road dust	3.0	1.0	0.30	0.30	0.0	0.0
Unpaved-road dust	3.0	1.0	0.30	0.30	0.0	0.0
Other fugitive dust (mainly construction)	1.0	2.0	0.30	0.30	0.0	0.0

Stack-gas vertical velocity (v_s): We assume a value of 0.0 for LDVs because LDV exhaust is directed downward or outward (parallel to the ground), but never upward, which means that the vertical velocity component of LDV exhaust is zero. Our estimate for HDVs considers that some HDVs (e.g., buses) have bottom exhausts pointed downward, some have top exhausts pointed sideways, some have straight-up top exhausts with flaps that deflect the upward thrust of the exhaust gases, and some have straight-up, open top exhausts. We assume that the exit velocity from non-road sources is the same as from HDVs. For fuel combustion (all three categories), chemicals etc., and solvents etc., we use the 50% (low-cost) and 75% (high-cost) values from Table 16-17. All of the other values are our estimates.

Diameter of stack (d_s). For motor vehicles and non-road mobile sources the estimates are of the size of the exhaust pipes. For fuel combustion (all three categories), chemicals etc., and solvents etc., we use the 50% (low-cost) and 75% (high-cost) values from Table 16-17. For natural sources, agriculture, roads, and fugitive dust, the estimates are of the diameter of the "footprint" of the plume at the ground.

Velocity of source (v_g). As discussed in the text, this parameter accounts for the effect on the plume of the movement of the source itself. In Report #4, we analyze the raw data from the Nationwide Personal Transportation Survey, and estimate that LDVs have an average speed of 34.7 mph (15.5 m/s) and HDVs have an average speed of 25 mph (11.2 m/s). We assume that non-road mobile sources, such as trains, are a bit slower on average than are highway vehicles.

TABLE 16-15 (CONTINUED)

	Tso		da (coarse	da	fine
	(o K)		(μ :	(µm)		m)
	Low	High	Low	High	Low	High
Light-duty vehicles	550	750	4.00	3.00	0.20	0.10
Heavy-duty vehicles	500	800	4.00	3.00	0.20	0.10
Fuel combustion: electric utilities	405	430	3.00	4.00	0.10	0.20
Fuel combustion: industrial	450	544	3.00	4.00	0.10	0.20
Fuel combustion: other (mainly residential wood combustion)	295	463	3.00	4.00	0.10	0.20
Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes	295	319	4.00	6.00	0.20	0.40
Solvent utilization, storage and transport, and waste disposal and recycling	295	298	4.00	6.00	0.20	0.40
Non-road vehicles (trains, tractors, ships, planes, etc.)	500	700	3.00	4.00	0.10	0.20
Natural sources (e.g., wind erosion and wildfires)	300	325	5.00	7.00	1.00	2.00
Agriculture and forestry, and managed burning	300	325	5.00	7.00	1.00	2.00
Paved-road dust	298	298	7.00	5.00	2.00	1.00
Unpaved-road dust	298	298	7.50	5.50	2.20	1.00
Other fugitive dust (mainly construction)	298	298	6.00	8.00	1.50	2.20

These assumptions were made on the basis of the following general considerations:

Temperature of stack gases (Tso): Data in Bosch's Automotive Handbook (1993) indicate that motor-vehicle exhaust is around 650° K. We assume that exhaust from non-road mobile sources is slightly cooler. For fuel combustion (all three categories), chemicals etc., and solvents etc., we use the 50% (low-cost) and 75% (high-cost) values from Table 16-17. We assume that dust from roads and construction is at the ambient temperature of 298 K. Emissions from natural and agricultural sources except fires also will be at the ambient temperature. On account of the higher temperature of the minor amount of emissions from fires, we have increased the

average temperature for natural sources (which include wildfires) and agricultural sources (which include managed burning) to slightly above the ambient.

Aerodynamic diameter of particles (d_a). These estimates are made on the basis of the data in section 16.2.4 above, and in Pinto et al. (1996).

TABLE 16-16: STATISTICS REGARDING AQCRS AND COUNTIES WITHIN AQCRS

	min.	max.	ave.	s. d.
For all AQCRs				
Area (mi ²) ^a	663	337,213	14,673	27,083
Number of counties ^a	1	86	13	11
Effective radius (r _r) (mi) ^b	15	328	59	35
Effective radius (r _c) of average-size county in AQCR (mi) ^C	8	134	19	12
Radius to out-of-county sources (r _o) (mi) ^d	15	268	49	27.4
<i>For small AQCRs</i> (<11,000 mi ²) (154 AQCRs)				
Area (mi ²) ^a	663	10,933	5,436	n.e.
Radius to out-of-county sources (r _o) (mi) ^d	15	51	35	8.6
<i>For large AQCRs</i> (>11,000 mi ²) (87 AQCRs)				
Area (mi ²) ^a	11,201	337,213	31,025	n.e.
Radius to out-of-county sources (r ₀) (mi) ^d	49	268	75	30.6

min. = minimum; max. = maximum; ave. = average; s. d. = standard deviation.

^aThe Bureau of the Census (1994) reports the area and number of counties of each of the 241 AQCRs in the U.S. We calculated the minimum, maximum, average, and standard deviation.

^bCalculated as $r_r = (A/\pi)^{0.5}$, where A is the area of the AQCR.. Note that the average r_r is the average of the individual calculated r_r values for each AQCR, not the r_r of the AQCR of average area A.

^cCalculated as $r_C = (A/N/\pi)^{0.5}$, where A is the area of the AQCR. and N is the number of counties in the AQCR. Note that the average r_C is the average of the individual calculated r_C values for each AQCR, not the r_C of the county of average area.

dCalculated as $r_0 = r_C + (r_r - r_C)^{0.93}$. Again, the average r_0 is the average of the individual calculated r_0 values.

TABLE 16-17. STATISTICS FOR MAJOR POINT SOURCES

	stack height (meters)				stack-gas velocity (meters/sec)			
	25%	50%	75%	90%	25%	50%	75%	90%
Fuel combustion: electric utilities	3	51	122	183	4.0	15.5	25.3	33.5
Fuel combustion: industrial	3	12	25	49	4.0	6.5	15.2	27.2
Fuel combustion: other (mainly residential wood combustion)	3	3	18	46	4.0	4.0	5.3	16.2
Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes	3	6	18	33	0.0	4.0	4.0	16.0
Solvent utilization, storage and transport, and waste disposal and recycling	3	3	7	17	0.003	4.0	4.0	9.1

	stack diameter (meters)				stack-gas temperature (degrees K)			
	25%	50%	75%	90%	25%	50%	75%	90%
Fuel combustion: electric utilities	0.20	3.40	5.30	7.30	295	405	430	586
Fuel combustion: industrial	0.20	0.80	1.70	2.70	295	450	544	672
Fuel combustion: other (mainly residential wood combustion)	0.20	0.20	1.20	2.30	295	295	463	533
Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes	0.10	0.20	0.80	1.50	294	295	319	400
Solvent utilization, storage and transport, and waste disposal and recycling	0.20	0.20	0.40	1.10	295	295	298	422

Notes: see next page.

We extracted these data from the EPA (1995d). Percentiles in column headings (25%, 50%, 75%, 90%) indicate the percent of sources that have a height, velocity, diameter, or temperature less than or equal to the value shown in the cell. The 50% values are thus the medians.

These statistics are consistent with data in the literature:

Adhikari et al. (1990) give an example of a coke even with h_S = 31 m, d_S = 0.3 m, v_S = 3.6 m/sec, and T_S = 5130 K.

Rowe et al. (1995) assume that for power plants, h_S = 40 to 150 m, v_S = 19 to 66 m/sec, d_S = 1.3 to 4.0 m, and T_S = 400 (800 for oil-fired plants).

Pasquill (1974) report field studies of diffusion in which h_S ranged from about 60 to 180 m, v_S = 1 to 30 m/sec, and d_S = 2 to 8 m.

Altshuller et al. (1996) write that there probably are less than 150 individual point sources in the U.S. with a stack height over 120 m.

See also the sample calculations in Hanna et al. (1982).

TABLE 16-18A. DEPOSITION VELOCITY OF PARTICLES AND GASES (CM/SEC)

	PM	SO ₂	SO ₄	NO ₂	NO ₃	NH3	СО
Dry depostion							
Hanna et al. (1982) ^a	f(size)	0.1 - 0.8					0.001
Dastoor and Pudykiewicz (1996) ^b	n.e.	0.1 - 0.8	n.e.	n.e.	n.e.	n.e.	n.e.
Eyre et al. (1997) ^c	0.12	0.96	0.11	0.16		n.e.	neg.
EPA (1994b) ^d	0.10	0.50	n.e.	1.00	1.00	1.00	n.e.
Langner and Rodhe (1991) ^e	n.e.	0.1 - 0.8	0.20	n.e.		n.e.	n.e.
Wet depostion							
EPA (1994b) wet deposition ^f	0.08·P (2.4 - 3.6)	0.50	n.e.	0.025·P (0).75 - 1.1)	0.014·P (0.42 - 0.63)	n.e.

n.e. = not estimated; neg. = negligible.

^aHanna et al. (1982) report an estimate that the deposition velocity of SO₂ ranges from 0.1 cm/sec over dry snow to 0.7- 0.8 cm/sec for water, countryside, medium crop, cities, and calcareous soil. They show deposition rates for PM as a function of size and other factors.

bFor their global meteorological model of sulfur transport, Dastoor and Pudykiewicz (1996) assume that the dry deposition velocity of SO₂ is 0.1 cm/sec over snow and ice, and 0.8 cm/sec elsewhere, at 1 meter height.

^cEyre et al. (1997) analyze the effect of location on the damage cost of transport emissions around London. The value under PM is for PM₁₀.

 $^{
m d}$ The EPA (1994b) develops a simple dispersion model of particulate-matter air quality. The value under NO₂ is for NO_X.

^eFor their model of the global sulfur cycle, Langner and Rodhe (1991) assume that the dry deposition velocity of SO₂ is 0.10 over snow and ice, 0.60 over land, and 0.80 over ocean, at 1.0 m height.

fP is the annual precipitation rate in inches. The value in parentheses is our calculation assuming annual precipitation of 30 to 45 inches (Bureau of the Census, *Statistical Abstract of the United States* 1992, 1992). The value under NO₂ is for NO_X.

TABLE 16-18. OUR ASSUMPTIONS AND CALCULATIONS REGARDING SETTLING AND DEPOSITION VELOCITY AND REACTION RATES OF PARTICLES AND GASES $^{\rm A}$

Deposition and settling parameters	PM	СО	NO _x , NH3	VOCs	SO _X
Settling velocity (m/sec) ^a	eq. 10a	0.0000	0.0000	0.0000	0.0000
Dry deposition velocity (m/sec)	eq. 10b-c	0.000005	0.0100	0.0050	0.0060
Ratio of wet deposition to dry deposition velocity ^b	30.0	1.0	0.9	1.0	0.5
Fraction of time with wet rather than dry deposition ^c	0.050	0.075	0.075	0.075	0.100
Calculated weighted-average deposition velocity (m/sec) ^d	see note d	0.00001	0.00993	0.00500	0.00570
Reaction rate: % reacted per hour ^e	0.0000	0.0500	0.0000	0.0000	0.0000

See the text for details.

^cThese are our estimates of the pertinent regionally weighted national average hours of precipitation per year, divided by 8760 hours per year. We assume that, on account of fugitive-dust emissions there, the Western U. S., which is relatively dry, has the bulk of PM emissions. We assume that CO, NO_X , and VOC emissions are distributed equally throughout the U.S., but that SO_X emissions are concentrated in the coal-burning Eastern U. S., which is relatively wet.

dCalculated as: $V_d = V_{d-dry} \cdot (1-F_w) + V_{d-dry} \cdot R_w/_d \cdot F_w$, where V_{d-dry} is the dry deposition velocity from this table or equation 10b or 10c, F_w is the fraction of time with wet rather than dry deposition (from this table) and $R_w/_d$ is the ratio of the wet deposition velocity to the dry (from this table).

eSee the text for details.

^aThe settling velocity for gases is zero.

^bAs shown in the text, the EPA's (1994b) assumptions about the wet deposition velocity as a function of the annual precipitation indicate that the wet deposition rate for particulates is about 30 times higher than the dry deposition rate, but that for the gases in this table, the wet deposition rate is of the same order of magnitude as the dry rate.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

A. URBAN MONITORS, EMISSION SOURCES WITHIN THE COUNTY, LOW-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)								
	fine PM ^a	coarse PM	CO	NO _x , NH3	VOCs	SO _X			
Light-duty vehicles	1.00	1.00	1.00	1.00	1.00	1.00			
Heavy-duty vehicles	0.60	0.60	0.60	0.60	0.60	0.60			
Fuel combustion: electric utilities	0.06	0.06	0.06	0.04	0.05	0.05			
Fuel combustion: industrial	0.09	0.09	0.09	0.05	0.07	0.07			
Fuel combustion: other (mainly residential wood combustion)	0.70	0.70	0.70	0.70	0.70	0.70			
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.38	0.38	0.37	0.38	0.38	0.38			
Solvent utilization, storage and transport; waste disposal; recycling	0.59	0.59	0.59	0.59	0.59	0.59			
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.57	0.57	0.57	0.57	0.57	0.57			
Natural sources (e.g., wind erosion and wildfires)	0.42	0.42	0.42	0.42	0.42	0.42			
Agriculture and forestry, and managed burning	0.42	0.42	0.42	0.42	0.42	0.42			
Paved-road dust	0.85	0.84	n.a.	n.a.	n.a.	n.a.			
Unpaved-road dust	0.37	0.37	n.a.	n.a.	n.a.	n.a.			
Other dust (mainly construction)	1.00	1.00	n.a.	n.a.	n.a.	n.a.			

See the text for details. Note that "low-cost" and "high-cost" refer to *motor-vehicle*-related costs. Thus, a high contribution by sources (such as wind erosion and construction) that are unrelated to motor-vehicle use results in a relatively small cost share for motor vehicle. n.a. = not applicable.

^aFine PM includes secondary organic aerosols.

bWe assume that the DNi for NH3 are the same as those calculated for NO_X .

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

B. Urban monitors, emission sources within the county, high-cost case

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)								
	fine PM ^a	coarse PM	СО	NO _X , NH3	VOCs	SO _X			
Light-duty vehicles	1.00	0.99	1.00	0.96	0.98	0.98			
Heavy-duty vehicles	1.00	1.00	1.00	0.96	0.98	0.98			
Fuel combustion: electric utilities	0.02	0.02	0.02	0.01	0.01	0.01			
Fuel combustion: industrial	0.08	0.08	0.08	0.06	0.07	0.07			
Fuel combustion: other (mainly residential wood combustion)	0.26	0.24	0.26	0.21	0.23	0.23			
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.06	0.05	0.06	0.04	0.05	0.05			
Solvent utilization, storage and transport; waste disposal; recycling	0.20	0.20	0.20	0.20	0.20	0.20			
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.22	0.21	0.22	0.21	0.21	0.21			
Natural sources (e.g., wind erosion and wildfires)	0.12	0.12	0.12	0.12	0.12	0.12			
Agriculture and forestry, and managed burning	0.12	0.12	0.12	0.12	0.12	0.12			
Paved-road dust	0.77	0.76	n.a.	n.a.	n.a.	n.a.			
Unpaved-road dust	0.23	0.23	n.a.	n.a.	n.a.	n.a.			
Other dust (mainly construction)	0.77	0.74	n.a.	n.a.	n.a.	n.a.			

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

C. Urban monitors, emissions outside the county, small \mathbf{AQCRs} , low-cost case

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)							
	fine PMa	coarse PM	СО	NO _X , NH3	VOCs	SO _X		
Light-duty vehicles	0.23	0.23	0.23	0.23	0.23	0.23		
Heavy-duty vehicles	0.15	0.15	0.15	0.15	0.15	0.15		
Fuel combustion: electric utilities	0.03	0.03	0.03	0.00	0.02	0.02		
Fuel combustion: industrial	0.02	0.02	0.02	0.00	0.01	0.01		
Fuel combustion: other (mainly residential wood combustion)	0.16	0.16	0.16	0.16	0.16	0.16		
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.14	0.14	0.14	0.14	0.14	0.14		
Solvent utilization, storage and transport; waste disposal; recycling	0.16	0.16	0.16	0.16	0.16	0.16		
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.16	0.16	0.16	0.16	0.16	0.16		
Natural sources (e.g., wind erosion and wildfires)	0.23	0.23	0.23	0.23	0.23	0.23		
Agriculture and forestry, and managed burning	0.23	0.23	0.23	0.23	0.23	0.23		
Paved-road dust	0.23	0.23	n.a.	n.a.	n.a.	n.a.		
Unpaved-road dust	0.23	0.23	n.a.	n.a.	n.a.	n.a.		
Other dust (mainly construction)	0.23	0.23	n.a.	n.a.	n.a.	n.a.		

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

$\mathbf{D}.$ Urban monitors, emissions outside the county, small $\mathbf{AQCRs},$ high-cost case

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)							
	fine PMa	coarse PM	СО	NO _x , NH3	VOCs	SO_X		
Light-duty vehicles	0.11	0.11	0.11	0.11	0.11	0.11		
Heavy-duty vehicles	0.08	0.08	0.08	0.08	0.08	0.08		
Fuel combustion: electric utilities	0.02	0.02	0.02	0.01	0.01	0.01		
Fuel combustion: industrial	0.03	0.02	0.03	0.01	0.02	0.02		
Fuel combustion: other (mainly residential wood combustion)	0.02	0.02	0.02	0.00	0.01	0.01		
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.02	0.01	0.02	0.00	0.01	0.01		
Solvent utilization, storage and transport; waste disposal; recycling	0.06	0.06	0.06	0.06	0.06	0.06		
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.07	0.07	0.07	0.07	0.07	0.07		
Natural sources (e.g., wind erosion and wildfires)	0.07	0.07	0.07	0.07	0.07	0.07		
Agriculture and forestry, and managed burning	0.07	0.07	0.07	0.07	0.07	0.07		
Paved-road dust	0.11	0.11	n.a.	n.a.	n.a.	n.a.		
Unpaved-road dust	0.11	0.11	n.a.	n.a.	n.a.	n.a.		
Other dust (mainly construction)	0.11	0.11	n.a.	n.a.	n.a.	n.a.		

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

E. Urban monitors, emissions outside the county, large \mathbf{AQCRs} , low-cost case

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)							
	fine PM ^a	coarse PM	СО	NO _X , NH3	VOCs	SO _X		
Light-duty vehicles	0.13	0.13	0.13	0.13	0.13	0.13		
Heavy-duty vehicles	0.09	0.09	0.08	0.09	0.09	0.09		
Fuel combustion: electric utilities	0.02	0.01	0.02	0.00	0.01	0.00		
Fuel combustion: industrial	0.01	0.01	0.01	0.00	0.00	0.00		
Fuel combustion: other (mainly residential wood combustion)	0.09	0.09	0.09	0.09	0.09	0.09		
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.08	0.08	0.08	0.08	0.08	0.08		
Solvent utilization, storage and transport; waste disposal; recycling	0.09	0.09	0.09	0.09	0.09	0.09		
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.09	0.09	0.09	0.09	0.09	0.09		
Natural sources (e.g., wind erosion and wildfires)	0.13	0.13	0.13	0.13	0.13	0.13		
Agriculture and forestry, and managed burning	0.13	0.13	0.13	0.13	0.13	0.13		
Paved-road dust	0.13	0.13	n.a.	n.a.	n.a.	n.a.		
Unpaved-road dust	0.13	0.13	n.a.	n.a.	n.a.	n.a.		
Other dust (mainly construction)	0.13	0.13	n.a.	n.a.	n.a.	n.a.		

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

F. Urban monitors, emissions outside the county, large $\mathbf{AQCRs},$ high-cost case

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)					
	fine PM ^a	coarse PM	СО	NO _X , NH3	VOCs	SO _X
Light-duty vehicles	0.05	0.05	0.05	0.05	0.05	0.05
Heavy-duty vehicles	0.04	0.04	0.04	0.04	0.04	0.04
Fuel combustion: electric utilities	0.01	0.01	0.01	0.00	0.01	0.01
Fuel combustion: industrial	0.01	0.01	0.01	0.00	0.01	0.01
Fuel combustion: other (mainly residential wood combustion)	0.01	0.01	0.01	0.00	0.00	0.00
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.01	0.00	0.01	0.00	0.00	0.00
Solvent utilization, storage and transport; waste disposal; recycling	0.03	0.03	0.03	0.03	0.03	0.03
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.03	0.03	0.03	0.03	0.03	0.03
Natural sources (e.g., wind erosion and wildfires)	0.04	0.04	0.04	0.04	0.04	0.04
Agriculture and forestry, and managed burning	0.04	0.04	0.04	0.04	0.04	0.04
Paved-road dust	0.05	0.05	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	0.05	0.05	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.05	0.05	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

G. AGRICULTURAL MONITORS, EMISSION SOURCES WITHIN THE COUNTY, LOW-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)					
	fine PMa	coarse PM	СО	NO _X , NH3	VOCs	SO _X
Light-duty vehicles	1.00	1.00	1.00	1.00	1.00	1.00
Heavy-duty vehicles	0.77	0.77	0.77	0.77	0.77	0.77
Fuel combustion: electric utilities	0.15	0.15	0.15	0.12	0.14	0.14
Fuel combustion: industrial	0.25	0.23	0.25	0.14	0.20	0.19
Fuel combustion: other (mainly residential wood combustion)	1.02	1.02	1.02	1.02	1.02	1.02
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.89	0.89	0.89	0.89	0.89	0.89
Solvent utilization, storage and transport; waste disposal; recycling	1.02	1.02	1.02	1.02	1.02	1.02
Non-road vehicles (trains, tractors, ships, planes, etc.)	1.00	1.00	1.00	1.00	1.00	1.00
Natural sources (e.g., wind erosion and wildfires)	2.84	2.84	2.84	2.84	2.84	2.84
Agriculture and forestry, and managed burning	7.40	7.35	7.41	7.27	7.34	7.33
Paved-road dust	1.18	1.18	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	2.09	2.09	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	1.47	1.47	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

H. AGRICULTURAL MONITORS, EMISSION SOURCES WITHIN THE COUNTY, HIGH-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DN $_{p',i}$)					
	fine PMa	coarse PM	СО	NO _X , NH3	VOCs	SO_X
Light-duty vehicles	1.00	1.00	1.00	0.99	1.00	0.99
Heavy-duty vehicles	0.85	0.84	0.85	0.83	0.84	0.84
Fuel combustion: electric utilities	0.02	0.02	0.02	0.02	0.02	0.02
Fuel combustion: industrial	0.30	0.28	0.30	0.23	0.27	0.26
Fuel combustion: other (mainly residential wood combustion)	0.23	0.21	0.23	0.16	0.20	0.19
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.23	0.18	0.23	0.15	0.19	0.19
Solvent utilization, storage and transport; waste disposal; recycling	0.52	0.52	0.52	0.52	0.52	0.52
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.56	0.56	0.56	0.56	0.56	0.56
Natural sources (e.g., wind erosion and wildfires)	1.21	1.19	1.21	1.18	1.20	1.20
Agriculture and forestry, and managed burning	3.40	3.30	3.41	3.28	3.35	3.34
Paved-road dust	1.22	1.22	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	1.44	1.43	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.94	0.93	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

I. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY, SMALL AQCRS, LOWCOST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DN $_{p',i}$)					
	fine PM ^a	coarse PM	CO	NO _X , NH3	VOCs	SO _X
Light-duty vehicles	0.46	0.46	0.46	0.46	0.46	0.46
Heavy-duty vehicles	0.30	0.30	0.30	0.30	0.30	0.30
Fuel combustion: electric utilities	0.07	0.06	0.07	0.01	0.05	0.04
Fuel combustion: industrial	0.04	0.04	0.04	0.00	0.02	0.02
Fuel combustion: other (mainly residential wood combustion)	0.32	0.32	0.32	0.32	0.32	0.32
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.28	0.28	0.28	0.28	0.28	0.28
Solvent utilization, storage and transport; waste disposal; recycling	0.32	0.32	0.32	0.32	0.32	0.32
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.31	0.31	0.31	0.31	0.31	0.31
Natural sources (e.g., wind erosion and wildfires)	0.46	0.46	0.46	0.46	0.46	0.46
Agriculture and forestry, and managed burning	0.46	0.46	0.46	0.46	0.46	0.46
Paved-road dust	0.46	0.46	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	0.46	0.46	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.46	0.46	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

J. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY, SMALL AQCRS, HIGH-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DNp', i)					
	fine PM ^a	coarse PM	CO	NO _x , NH3	VOCs	SO _X
Light-duty vehicles	0.31	0.31	0.30	0.30	0.30	0.30
Heavy-duty vehicles	0.21	0.21	0.21	0.21	0.21	0.21
Fuel combustion: electric utilities	0.05	0.04	0.05	0.03	0.04	0.04
Fuel combustion: industrial	0.08	0.07	0.08	0.02	0.06	0.05
Fuel combustion: other (mainly residential wood combustion)	0.06	0.05	0.06	0.00	0.04	0.03
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.06	0.03	0.06	0.00	0.04	0.03
Solvent utilization, storage and transport; waste disposal; recycling	0.17	0.17	0.17	0.17	0.17	0.17
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.18	0.18	0.18	0.18	0.18	0.18
Natural sources (e.g., wind erosion and wildfires)	0.20	0.20	0.20	0.20	0.20	0.20
Agriculture and forestry, and managed burning	0.20	0.20	0.20	0.20	0.20	0.20
Paved-road dust	0.31	0.30	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	0.31	0.30	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.31	0.30	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

K. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY, LARGE AQCRS, LOW-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DNp',i)					
	fine PM ^a	coarse PM	СО	NO _x , NH3	VOCs	SO _X
Light-duty vehicles	0.26	0.26	0.26	0.26	0.26	0.26
Heavy-duty vehicles	0.17	0.17	0.17	0.17	0.17	0.17
Fuel combustion: electric utilities	0.03	0.03	0.03	0.00	0.01	0.01
Fuel combustion: industrial	0.02	0.01	0.02	0.00	0.00	0.00
Fuel combustion: other (mainly residential wood combustion)	0.18	0.18	0.18	0.18	0.18	0.18
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.16	0.16	0.16	0.16	0.16	0.16
Solvent utilization, storage and transport; waste disposal; recycling	0.18	0.18	0.18	0.18	0.18	0.18
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.18	0.18	0.17	0.18	0.18	0.18
Natural sources (e.g., wind erosion and wildfires)	0.26	0.26	0.26	0.26	0.26	0.26
Agriculture and forestry, and managed burning	0.26	0.26	0.26	0.26	0.26	0.26
Paved-road dust	0.26	0.26	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	0.26	0.26	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.26	0.26	n.a.	n.a.	n.a.	n.a.

TABLE 16-19. MODEL RESULTS: ESTIMATED VALUES FOR $DN_{P',I,C}$, AND $DN_{P',I,OC}$, THE CONTRIBUTION TO AMBIENT POLLUTION PER U NIT OF EMISSION, FOR EACH POLLUTANT AND EMISSION-SOURCE CATEGORY, RELATIVE TO THE CONTRIBUTION OF LIGHT-DUTY MOTOR-VEHICLES

L. AGRICULTURAL MONITORS, EMISSIONS OUTSIDE THE COUNTY, LARGE AQCRS, HIGH-COST CASE

	Calculated relative contribution to ambient air quality, per kg of emission (DN $_{p',i}$)					
	fine PM ^a	coarse PM	CO	NO _X , NH3	VOCs	SO _X
Light-duty vehicles	0.15	0.15	0.15	0.15	0.15	0.15
Heavy-duty vehicles	0.10	0.10	0.10	0.10	0.10	0.10
Fuel combustion: electric utilities	0.03	0.03	0.03	0.00	0.02	0.02
Fuel combustion: industrial	0.04	0.03	0.04	0.00	0.02	0.02
Fuel combustion: other (mainly residential wood combustion)	0.03	0.02	0.03	0.00	0.01	0.01
Chemicals and allied product manufacturing; metals processing, petroleum refining; other industry	0.03	0.00	0.03	0.00	0.01	0.01
Solvent utilization, storage and transport; waste disposal; recycling	0.08	0.08	0.08	0.08	0.08	0.08
Non-road vehicles (trains, tractors, ships, planes, etc.)	0.09	0.09	0.09	0.09	0.09	0.09
Natural sources (e.g., wind erosion and wildfires)	0.10	0.10	0.10	0.10	0.10	0.10
Agriculture and forestry, and managed burning	0.10	0.10	0.10	0.10	0.10	0.10
Paved-road dust	0.15	0.15	n.a.	n.a.	n.a.	n.a.
Unpaved-road dust	0.15	0.15	n.a.	n.a.	n.a.	n.a.
Other dust (mainly construction)	0.15	0.13	n.a.	n.a.	n.a.	n.a.

TABLE 16-20. EPA-ESTIMATED EXPOSURE FACTORS FOR DIFFERENT PM EMISSION SOURCES (EPA, 1994b)

Emissions source category ^a	Effective emissions ^b	Exposure ^C	Exposure: emissions	Exposure: relative to vehice	highway
	10 ⁶ tons	10 ⁶ people- μg/m ³	people- μg/m ³ per ton	highway @ 200	highway @ 300
Fuel combustion ^e	5.21	503.1	96.6	0.38	0.25
Manufacturing ^f	1.69	328.7	194.5	0.77	0.51
Transportation total9	1.53	385.1	251.7	0.99	0.66
Highway vehicles	0.79	200/300.0	253.9/380.9	1.00	1.00
Nonhighway vehicles	0.74	185.1/85.1	249.3/114.6	0.98	0.30
Natural sources	7.52	355.1	47.2	0.19	0.12
Paved-road dust	7.49	1,639.3	218.9	0.86	0.57
Unpaved-road dust	15.52	1,788.3	115.2	0.45	0.30
Construction	9.89	5,761.1	582.5	2.29	1.53
Agriculture, other dust, managed burning	9.27	644.8	69.6	0.27	0.18
Total	58.12	11405.5	196.2	0.77	0.52

^aThese are emission categories in the EPA's official emissions inventory.

^bFrom the EPA's (1995d) estimates for the 1990 national particulate emissions inventory, except as noted. Effective emissions include the EPA's estimates of ammonium sulfate and ammonium nitrate formed from direct emissions of SO_2 , NO_X , and NH_3 . We do not use their estimates of effective emissions, because we have our own model of the formation of secondary particulate matter.

^cThe EPA's (1994b) estimates of exposure (except as noted), based on simple dispersion modeling.

dEqual to the exposure:emissions ratio for each source category divided by either: a) the exposure:emissions ratio for highway vehicles assuming that exposure to highway-vehicle exhaust PM is $200 \cdot 10^6$ people-μg/m³, or b) the exposure:emissions ratio for highway vehicles assuming that exposure to highway-vehicle exhaust PM is $300 \cdot 10^6$ people-μg/m³. We show the results for two different assumed exposures to highway-vehicle exhaust PM because, as

explained in note e, the EPA (1994b) estimates exposure to "transportation" in general but not highway vehicles specifically.

^eComprises our three fuel-combustion categories.

fThe "Transportation" source category includes non-highway vehicles as well as highway vehicles. The EPA (1994b) estimates effective emissions and exposure for the overall transportation source category, but not for the two individual components (highway and nonhighway vehicles). We need estimates for highway vehicles specifically because we wish to estimate emissions dispersion normalized to dispersion from highway vehicles.

We disaggregate total effective transportation PM emissions (1.53 million tons) into effective highway PM emissions and effective non-highway PM emissions according to the ratio of *direct* highway PM emissions to *direct* non-highway PM emissions. (Direct emissions do not include secondary ammonium sulfate or secondary ammonium nitrate.) We disaggregate total exposure to transportation PM emissions (385.1 million people $\mu g/m^3$) in two scenarios: one in which highway vehicles account for 200 out of the 385.1 million people $\mu g/m^3$ exposure, and a second in which highway vehicles account for 300 out of the 385.1. We assume that highway vehicles generally account for proportionally greater exposure than do non-highway vehicles because generally they are closer to more people than are non-highway vehicles.

gComprises our categories "Chemicals and allied product manufacturing, metals processing, petroleum refining, and other industrial processes" and "Solvent utilization, storage and transport, and waste disposal and recycling" from Table 16-15.

TABLE 16-21. DIESEL ENGINES IN THE SOUTH COAST AIR BASIN, 1982: FUEL USE, EMISSIONS, AND CONTRIBUTION TO TOTAL PARTICULATE POLLUTION

	On- road ^b	Ships	Rail- roads	Off- road	Industry	Total
Concentration (µg/m ³) ^a						
Azusa	1.41	0.01	0.24	0.21	0.00	1.87
Long Beach	3.49	0.06	0.80	0.21	0.01	4.57
Lennox	3.80	0.01	0.59	0.26	0.01	4.67
Pasadena	1.97	0.01	0.30	0.25	0.01	2.54
West L. A.	3.82	0.01	0.19	0.27	0.01	4.30
Downtown L. A.	3.53	0.01	1.72	0.31	0.01	5.58
Anaheim	2.74	0.03	0.40	0.32	0.01	3.50
Total	20.76	0.14	4.24	1.83	0.06	27.03
Fuel use and emissions						
Fuel use (10 ⁹ BTU)	191.5	6.3	22.4	39.0	7.0	266.2
PM emissions (kg/day) ^C	11,101	310	1,836	3,065	769	17,081
Shares ^d						
Fuel	0.72	0.024	0.08	0.15	0.026	1.000
Concentration	0.77	0.005	0.16	0.07	0.002	1.000
Emissions	0.65	0.018	0.11	0.18	0.045	1.000
Concentration/ emissions ^e	1.18	0.29	1.46	0.38	0.05	n.a.
Concentration/emissions, relative to on-road	1.00	0.24	1.23	0.32	0.04	n.a.

From Cass and Gray (1995), and our calculations. n.a. = not applicable.

^aCass and Gray (1995) modeled the concentration of PM pollution from each emissions source, at the seven ambient air-quality monitors, located in different parts of the South Coast Air Basin.

^bCass and Gray (1995) considered diesel autos, diesel light trucks, and diesel heavy trucks separately. We have combined them into a general "on-road vehicle" category.

^cTotal particulate mass emitted into the atmosphere in South Coast Air Basin in 1982 (Cass and Gray, 1995).

dIn each diesel-engine category, the share is equal to fuel use, concentration, or emissions in that category divided by total fuel use, concentration, or emissions.

^eEqual to the concentration share divided by the emission share, in each category. The fuel shares by source in the south coast are reasonably close to the fuel shares by source throughout California, Arizona, and Nevada (Table 6-21).

fEqual to the concentration/emission ratio in each category divided by the concentration emission ratio for on-road vehicles.

TABLE 16-22. OZONE SENSITIVITY TO VOC AND NOX EMISSIONS

Pollution ^a	VOC sensitivityb	NO _X sensitivity ^C
1.00	n.a.	n.a.
0.95	0.556	0.406
0.90	0.563	0.413
0.80	0.577	0.427
0.70	0.594	0.443
0.60	0.612	0.462
0.50	0.634	0.484
0.40	0.660	0.511
0.30	0.692	0.546
0.20	0.734	0.593
0.10	0.798	0.669
0.05	0.850	0.735
0.01	0.930	0.850
0.00	1.000	1.000

^aThe ratio of the final to the initial pollution level. As shown next, the ozone sensitivity is a function only of this ratio (equal to K, which we define below) and the value of the exponent in equation 11.

bThe percent change in ozone divided by the percent change in VOC emissions, defined as follows:

$$\gamma_{voc} = \frac{\frac{O3o - O3f}{O3o}}{\frac{VOCo - VOCf}{VOCo}}$$

where:

 γ_{voc} = the ozone sensitivity

O₃₀ = the initial ozone level (corresponding to 1.0 units of pollution)

 O_{3f} = the final ozone level, corresponding to the pollution level shown in the first column

VOC_o = the initial level of VOC pollution (1.0 units of pollution)

VOC_f = the final level of VOC pollution, shown in the first column

 NO_X = any level of NO_X pollution (this term will cancel out)

This expression can be simplified:

$$\gamma_{voc} = \frac{\frac{O3o - O3f}{O3o}}{\frac{VOCo - VOCf}{VOCo}}$$

$$O30 = VOC o^A \cdot NOx^B$$

$$O3f = VOCf^A \cdot NOx^B$$

Let VOCf / VOCo = K

$$\gamma_{voc} = \frac{\frac{VOC \circ^{A} \cdot NOx^{B} - VOC f^{A} \cdot NOx^{B}}{VOC \circ^{A} \cdot NOx^{B}}}{1 - K} = \frac{\frac{VOC \circ^{A} - VOC f^{A}}{VOC \circ^{A}}}{1 - K}$$

$$= \frac{\frac{VOC \, o^A - \left(VOC \, o \cdot K\right)^A}{VOC \, o^A}}{1 - K} = \frac{1 - K^A}{1 - K}$$

Thus, we see that, given an ozone-formation equation in which ozone is a product of NO_X and VOC emissions, the ozone sensitivity to VOCs is a function only of the ratio of final to initial VOC pollution, and the VOC exponent A.

^cThe percent change in ozone divided by the percent change in NO_X emissions, calculated analogously to the VOC sensitivity.

TABLE 16-23. EMISSIONS, POCP-WEIGHTED EMISSIONS, AND POCP- ADJUSTMENT FACTORS FOR VARIOUS VOC-EMISSION SOURCES

Source category in U. K. emissions inventory	Source category in U.S. emissions inventory	Emissions in U. K. (kt/yr.)	POCP- weighted emissions in U. K. (kt/yr.)	POCP adjustment factor ^a
Petrol exhaust	Gasoline vehicle exhaust ^b	652	506	0.78
Petroleum refining and distribution	Petroleum and related industries	134	83	0.62
Petrol evaporation	Gasoline vehicle exhaust ^b	143	87	0.61
Solvent usage	Solvents and storage	787	461	0.59
Stationary combustion	Fuel combustion by electric utilities; Fuel combustion by industry; Fuel combustion by other; Other combustion	56	27	0.49
Diesel exhaust	Diesel vehicles; Non-road engines	175	77	0.44
Industrial and residential waste	Waste disposal and recycling	3	1	0.28
Natural gas leakage	None	34	9	0.26
Chemical processes	Chemicals and allied products; Metals processing; Other industrial processes	200	43	0.21
Biogenic emissions ^C	Biogenic emissions	n.a.	n.a.	1.1

From Derwent et al. (1996), except "Source category in U. S. emissions inventory," which is our matching. POCP = photochemical ozone-creation potential. kt = kiloton

^cBased on POCP estimates of biogenic VOCs (e.g., terpene) from Derwent et al. (1996).

^aEqual to the ratio of POCP-weighted emissions to unweighted emissions. Derwent et al. (1996) refer to this as the "sector-mean POCP".

bOur runs of EMFAC7F, and the analysis by Ross et al. (1995), suggest that vehicular evaporative emissions (including refueling emissions, but not further "upstream" emissions) are about 0.4, and vehicular exhaust emissions about 0.6, of total (exhaust + evaporative) VOC emissions from motor vehicles.

Table 16-24. Adjusted sales of distillate fuel oil in Arizona, California, and Nevada in 1993, by type of end use $(10^3 \, \text{Gallons})$

	Arizona	California	Nevada	Total	Shares for all	Shares for California
Residential	224	6493	7518	14235	0.004	0.003
Commercial	7027	66780	25537	99344	0.030	0.027
Industrial	43525	39502	91183	174210	0.052	0.016
Oil company	151	10254	120	10525	0.003	0.004
Farm	11900	202780	3640	218320	0.065	0.082
Electric utility	1012	5287	1259	7558	0.002	0.002
Railroad	10212	124305	1147	135664	0.040	0.050
Ships	16	54032	0	54048	0.016	0.022
On-highway	474134	1824363	174537	2473034	0.735	0.735
Military	1415	30936	830	33181	0.010	0.012
Off highway ^a	16065	115199	14011	145275	0.043	0.046
All other	0	714	0	714	0.000	0.000
Total	565681	2480645	319782	3366108	1.000	1.000

From the EIA's *Fuel Oil and Kerosene Sales 1994* (1995). "Adjusted" means that intermediate totals, at the level of the Petroleum Administration District (PAD), have been adjusted to sum to EIA's estimate of products marketed nationally. Distillate fuel oil includes no. 1, no. 2, and no. 4 diesel fuels; it excludes kerosene and residual fuel oil.

^aIncludes construction equipment and logging.

TABLE 16-25. SOURCE-SPECIFIC FACS BY LAND COVER TYPE

		Source- specific			
Land cover	Terpenes	Olefins	Paraffins	Aromatics	FAC ^b
Oak Forest	0.149	0.023	0.135	0.121	0.047
Other Deciduous	0.262	0.028	0.167	0.149	0.082
Coniferous Forest	0.607	0.059	0.176	0.086	0.184
Scrub Land	0.374	0.021	0.216	0.189	0.116
Grass Land	0.282	0.017	0.174	0.153	0.088
Agricultural	0.261	0.055	0.354	0.299	0.084
Inland Water	0.571	0.026	0.167	0.141	0.174
Urban	0.385	0.015	0.156	0.137	0.118

From EPA (1994a: Table II-13).

FAC = fractional aerosol coefficient. The FAC multiplied by the emissions of the organic compound gives the mass of secondary organic aerosol (SOA) formed.

^aFor each source (land-cover category), the weight fraction of the particular compound (terpenes, olefins, paraffins, or aromatics) is the fraction of the particular organic compound out of total mass of organic compounds. Curiously, the weights as provided by the EPA, do not add up to one. It is unclear why this is so. However, we note that the EPA's (1994a) reported biogenic inventory apparently uses an average FAC of 12.8%, which is consistent with the figures given here.

^bThe source-specific FAC is equal to Σ FAC_i · W_i where FAC_i = the FAC for compound i (0.30 for terpene; 0.0 for olefins and paraffins; and 0.02 for aromatics), and W_i = the weight fraction of compound i FAC.

TABLE 16-26. COMPARISON OF SOURCE-APPORTIONMENTS FROM CHEMICAL MASS-BALANCE STUDIES (CMB) WITH MODELING RESULTS -- PERCENTAGES OF PM10 ATTRIBUTABLE TO FOUR SOURCES

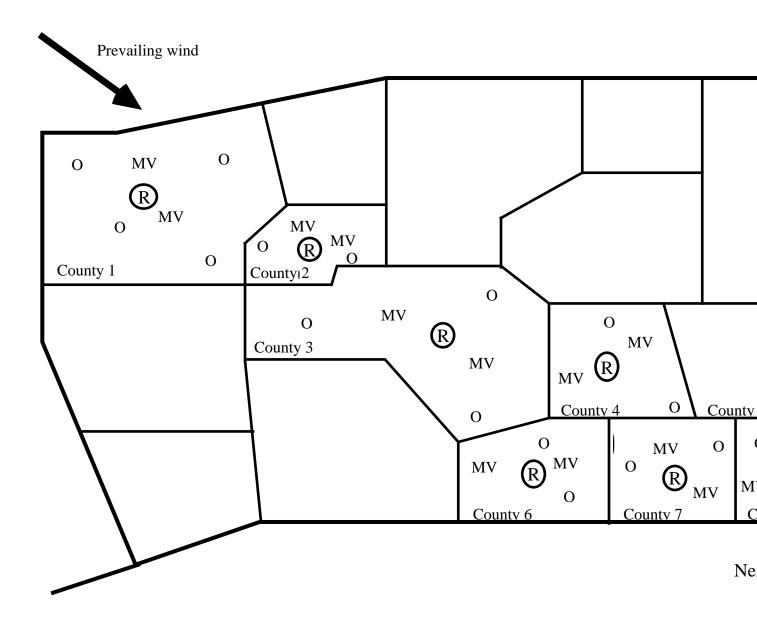
	Primary geological (CMB)		Road dust (Modeled)		Primary motor vehicle (CMB) ^a		Primary motor vehicle (Modeled) ^a		Secondary ammonium nitrate (CMB)		Secon ammo nitr (Mod
Study area	Low	High	Low	High	Low	High	Low	High	Low	High	Low
Arizona											
Phoenix	43	74	12	27	18	39	2	4	0	7	4
Corona de Tucson	54	89	16	34	2	29	2	4	0	0	5
Hayden	5	36	13	39	0	0	1	4	0	0	0
California											
Fresno	36	44	24	48	8	10	3	7	0	15	8
Bakersfield, Fellows, Kern Wildlife Refuge	32	54	20	42	4	10	3	5	0	16	9
Azusa, Burbank, Claremont, Los Angeles, Hawthorne, Lennox, Long Beach	10	43	36	57	10	45	7	9	1	28	16
Anaheim	13	53	36	57	17	36	6	8	6	37	14
Indio, Palm Springs, Riverside, Rubidoux	17	57	32	53	7	27	5	7	7	33	11
Upland	44	44	33	50	7	7	5	7	25	25	12
Chula Vista	23	33	28	45	3	5	6	8	0	0	12
Stockton	55	55	24	45	8	8	3	5	11	11	7
Santa Maria	27	27	27	44	28	28	4	6	5	5	10
Santa Barbara, Santa Ynez, Vandenberg AFB	7	28	27	45	16	43	4	6	2	5	12
San Jose	18	19	32	55	13	14	6	8	19	20	12
Crows Landing	61	61	23	43	4	4	3	5	12	12	7
Colorado											
Telluride	45	72	21	31	0	0	1	1	0	0	2
Idaho											
Pocatello	8	8	33	43	0	0	1	2	0	0	2
Illinois											
Chicago	34	37	8	18	2	3	2	3	0	0	4
Nevada											
Reno, Sparks, Verdi	37	52	13	29	27	33	2	4	1	2	5
Ohio											
Mingo	20	20	42	53	23	23	1	2	0	0	3
Stuebenville	18	18	27	42	30	30	1	4	0	0	0

Notes: see next page.

The entries in this Table give low and high estimates of the percentage of PM₁₀ attributable to each general source category in each county. With the CMB results, the "low" entry is the lowest reported CMB result in Table 16-9 for a particular county, and the "high" is the highest reported result for the county. With our modeled results, the "low" means "low motor-vehicle damage cost," and the "high" means "high motor-vehicle damage cost". That is, the modeled "low" share for secondary ammonium sulfate (SAS) is not the numerically smaller overall share, but rather the SAS share that results from the parameter values that give the low motor-vehicle damage cost. Because motor vehicles are relatively minor sources of sulfur, a higher overall SAS share in effect gives more weight to non-motor-vehicle sources of emissions, and thereby reduces the contribution of motor vehicles to ambient particulates and particulate damages.

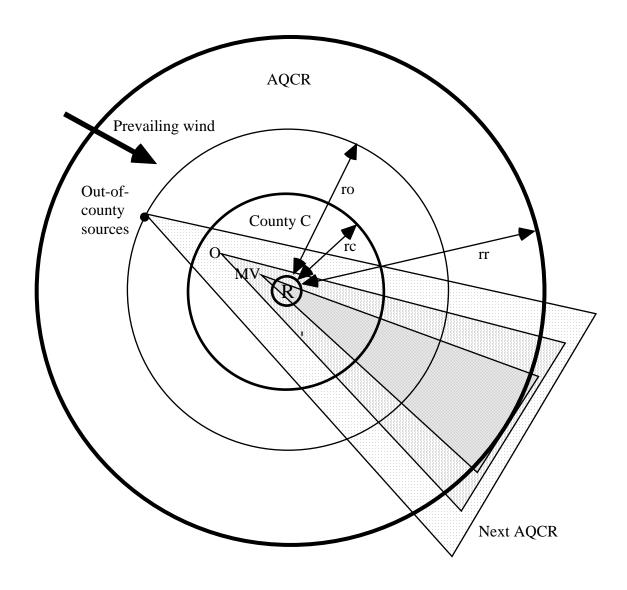
 a In the CMB studies, the category "PMV" includes only direct or primary PM emissions from motor vehicles themselves; it does not do not include secondary PM from NO_X or SO_X from motor vehicles (all such secondary PM is included under the SAN or SAS categories), or upstream motor-vehicle related emissions (which appear under "miscellaneous categories). In order to have a proper comparison, we have -- for this table only -- set up our model so that the motor-vehicle results include only direct, primary PM emissions, just as in the CMB studies. Thus, just for this comparison, we assign all secondary ammonium nitrate and secondary ammonium sulfate to the categories SAN and SAS, and leave out upstream motor-vehicle related PM altogether. Of course, the motor-vehicle-related contributions that we actually use to estimate motor-vehicle related costs include secondary and upstream PM related to motor-vehicle use.

FIGURE 16-1. MOTOR-VEHICLE EMISSION SOURCES, OTHER EMISSION SOURCES, AND RECEPTOR SITES IN COUNTIES IN AN AIR-QUALITY CONTROL REGION



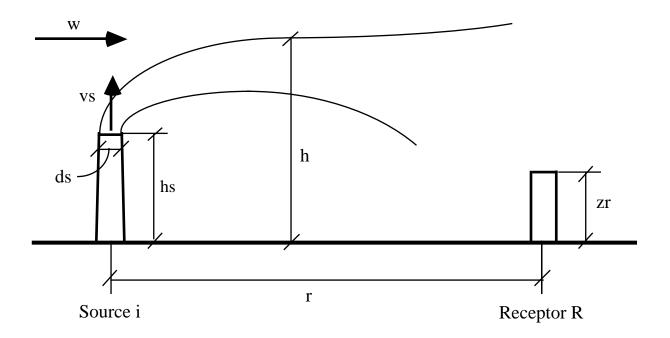
MV = motor-vehicle emission sources; O = other emission sources; R = receptor site (air-quality monitor); ACQR = Air-Quality Control Region.

FIGURE 16-2. MODELED REPRESENTATION OF MOTOR-VEHICLE EMISSION SOURCES, OTHER EMISSION SOURCES, AND RECEPTOR SITES IN COUNTIES IN AN AIR-QUALITY CONTROL REGION



AQCR = Air Quality Control Region; MV = motor-vehicle sources; O = other sources;

FIGURE 16-3. DISPERSION OF POLLUTION FROM A POINT SOURCE



w = wind velocity; v_S = stack-gas velocity; d_S = stack diameter; h_S = stack height; z_T = receptor height; h = effective height; h = distance from source to receptor