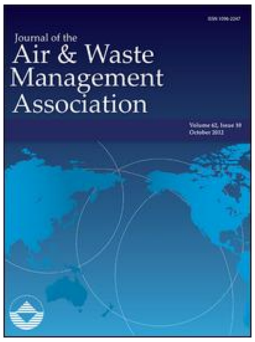

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Fine Particulate Matter Emissions Inventories: Comparisons of Emissions Estimates with Observations from Recent Field Programs

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ABSTRACT

Emissions inventories of fine particulate matter ($PM_{2.5}$) were compared with estimates of emissions based on data emerging from U.S. Environment Protection Agency Particulate Matter Supersites and other field programs. Six source categories for $PM_{2.5}$ emissions were reviewed: on-road mobile sources, nonroad mobile sources, cooking, biomass combustion, fugitive dust, and stationary sources. Ammonia emissions from all of the source categories were also examined. Regional emissions inventories of PM in the exhaust from on-road and nonroad sources were generally consistent with ambient observations, though uncertainties in some emission factors were twice as large as the emission factors. In contrast, emissions inventories of road dust were up to an order of magnitude larger than ambient observations, and estimated brake wear and tire dust emissions were half as large as ambient observations in urban areas. Although comprehensive nationwide emissions inventories of $PM_{2.5}$ from cooking sources and biomass burning are not yet available, observational data in urban areas suggest that cooking sources account for approximately 5–20% of total primary emissions (excluding dust), and biomass burning sources are highly dependent on region. Finally, relatively few observational data were available to assess the accuracy of emission estimates for stationary sources. Overall, the uncertainties in primary emissions for $PM_{2.5}$ are substantial. Similar uncertainties exist for ammonia emissions. Because of these uncertainties, the design of $PM_{2.5}$ control strategies should be based on inventories that have been refined by a combination of bottom-up and top-down methods.

IMPLICATIONS

Characterizing emissions is a critical step in understanding the processes that lead to the accumulation of $PM_{2.5}$ in the atmosphere; however, in many cases, the uncertainty in emission estimates for $PM_{2.5}$ are a factor of two or more. Greater confidence in emissions inventories can emerge from using a combination of emission estimates (bottom-up methods) and comparisons with ambient observational data (top-down methods).

INTRODUCTION

Emissions inventories can be used to establish statewide and nationwide trends in air quality or to prioritize emission sources in specific geographical areas. They can also be used as inputs to models used to predict ambient air quality on specific days. The temporal resolution of the emissions inventory depends on the purpose of the inventory. Emissions inventories that are used to establish air quality trends at regional or national scales need only have information about average emission rates. However, emissions inventories that will be used in models that predict air quality on specific days or that are used to predict the likelihood of extremes in air pollutant concentrations must consider both average emission rates and daily variability in emissions.

The goal of this review is to assess the accuracy of emissions inventories of fine particulate matter (PM; PM <2.5 μm in aerodynamic diameter [$PM_{2.5}$]) and one of its precursors, ammonia. Precursors of secondary $PM_{2.5}$ include ammonia, nitrogen oxides, sulfur dioxide, and hydrocarbons (HCs). Of these $PM_{2.5}$ precursors, this review addresses only ammonia, because the emissions inventories for nitrogen oxides and sulfur dioxide are reasonably accurate, and the uncertainties in the HC emissions inventory merit a separate review. Ambient observations of $PM_{2.5}$ mass and composition are compared with $PM_{2.5}$ mass and composition anticipated based on emissions inventories. Annual and seasonal emissions inventories are the primary focus of this review, because this comparison uses annually and seasonally averaged observational data. Comparisons were performed for a variety of geographical regions. The review of $PM_{2.5}$ emissions inventories is grouped into sections based on major emission source categories, specifically, stationary sources, area sources, mobile sources (on-road and nonroad), and fugitive dust.

Stationary sources include industrial and nonindustrial stationary equipment or processes that can be identified by name and location and are considered significant sources of air pollution emissions. Examples of stationary sources include industrial and commercial boilers, electric utility boilers, turbine engines, wood and pulp processors, paper mills, industrial surface coating facilities, and refinery and chemical processing operations.

Area sources principally associated with primary emissions of $PM_{2.5}$ include cooking, residential combustion, and outdoor fires. Area sources can also include stationary sources that are not inventoried as major stationary sources because of their low emission rates but when present in large numbers can collectively emit significant quantities of pollutants to an area.

Mobile sources are generally categorized as on-road or nonroad. Emissions from on-road mobile sources result from the use of cars, trucks, and buses traveling on public roadways. Nonroad sources include a wide variety of internal combustion engines that are mobile but that are not used on public roadways (e.g., construction and industrial equipment, residential and commercial lawn mowers, locomotives, airplanes, and boats). Anthropogenic fugitive dust sources are often included as part of the on-road and nonroad inventories and include material suspended in the atmosphere because of driving on unpaved roadways, agricultural operations, construction activities, and other sources.

Two previous reviews of $PM_{2.5}$ emission estimation methods conducted by North American Research Strategy for Tropospheric Ozone (NARSTO) serves as the starting point for this assessment. These previous reviews broadly characterized the spatial and temporal distribution of emissions and qualitatively assessed levels of uncertainty in emission estimates for PM and PM precursors[1] and described emission estimation tools and tools for evaluating the accuracy of emissions inventories.[2]

The current work assesses the accuracy of primary PM and ammonia emission estimates using the wealth of measurements of source and ambient air quality collected during recent field campaigns such as the U.S. Environmental Protection Agency (EPA) PM Supersites Program.[3] Because emissions inventories are generally made more accurate through gradually refined emission estimates (bottom-up methods) and comparisons with ambient observational data (top-down methods), the review presents both bottom-up and top-down methods for each source type. The review is organized by major emission source categories of $PM_{2.5}$. For each source category, the review examines the following: (1) source types; (2) emission estimation methods and magnitudes of estimated emissions; (3) estimated spatial and temporal variability; (4) PM composition and size distributions and comparisons; and (5) comparisons of PM characteristics based on emission estimates with those based on observations.

The review of ammonia emissions, the only PM precursor examined, will be presented as a single section that groups together emission source categories. Summary tables are provided for each source category.

Bottom-Up Methods: Emission Estimates Based on Emission Rates

Emissions inventories can be developed and aggregated in a variety of ways, but a common approach is to assemble emissions for a particular time period in a particular region. Emission estimates used in this assessment are based on activity, emission factors, and control factors, as shown in eq 1.

$$\text{Emission rate} = \text{Activity factor} \times \text{Emission factor} \times \text{Control factor} \quad (1)$$

An activity factor characterizes source use, an emission factor is the amount of pollutant released per unit activity, and a control factor is the fractional decrease of pollutant release by an air pollution control device (APCD).

Examples of activity factors include the amount of coal burned by a stationary combustion source each year, the distance traveled by an automobile in a day, or the barrels of oil produced each month at a refinery. Examples of emission factors are the emissions per kilometer traveled by an automobile or the emissions per mass of fuel combusted. Emission factors are generally assumed to be representative of long-term averages for all sources in the emission category represented by the emission factor. The emission factors may require adjustment to convert the activity into a temporal and spatial unit suitable for the inventory, and it is desirable to have emission factors for PM that specify PM composition and size. Emission factors for PM are compiled in the Factor Information Retrieval Data System,[4] in the EPA document series *AP-42*,[5] in *Locating and Estimating Air Toxic Emissions*,[6] in state-reported test data, and in the scientific literature.

Control factors account for the fractional reduction of the uncontrolled emission by an APCD used before the point of release. Control factors vary by source type and by region, because pollution control is often mandated by state or local agencies.

Speciation of PM emissions and allocation of PM emissions into size ranges is generally performed by multiplying the total PM emission rate by source composition profiles and size profiles. Composition profiles describe the distribution of major PM components for the source; many composition factors are summarized EPA's SPECIATE database[7]; size distributions are more variable and can range from a simple distribution of PM emissions into fine and coarse fractions to nearly continuous distributions of the relative number of concentrations as a function of size.

Top-Down Methods: Emissions Estimates Based on Ambient Observations

A variety of analytical and computational tools are used to compare ambient observations of PM concentrations and compositions to information in PM emissions inventories. These methods for comparing ambient observational data to emissions inventories can be used to infer whether sources are missing from the inventory or whether the relative emission rates in the inventory are accurate. Among these tools are trajectory analyses, source-receptor models, and chemical-transport models. The primary tools that are discussed in this review are source-receptor methods. Descriptions of the remaining tools can be found in the reviews of $PM_{2.5}$ source resolution methods.[2]

Receptor models describe ambient air pollutant concentrations as a sum of contributions from individual or composite sources. Factor analysis is used to identify one or more distinct factors, or groupings of pollutants, that influence the variability of air quality at the chosen receptor in a consistent manner. These factors, together with a measure of the strength of each factor, account for the total variability in pollutant composition and magnitude observed at the receptor site.[8,9] Because the factors are determined from ambient observations and without any knowledge of the contributing emission sources, they can provide an independent evaluation of the emissions inventory. However, they require relatively large datasets and do not identify specific emission sources. In recent years, this technique has been used successfully to identify PM source contributions in Pittsburgh, PA; Atlanta, GA; Seattle, WA; and other cities.[10–12] Another source-receptor modeling approach, referred to as chemical mass balance (CMB), seeks to explain measurements of ambient air pollutants at a downwind receptor location as a linear sum of specific upwind source types. Instead of determining factors, CMB estimates the strength of each specific contributing source type. As such, this approach requires information about source composition.[9] CMB has been used recently to evaluate source contributions in Los Angeles, CA; the San Joaquin Valley, CA; the Southeastern United States; and other locations.[13–15] Both factor analysis and mass balance approaches provide information limited to the period represented by the dataset, and both approaches will meet with limited success when the study area is influenced by collinear, reactive, or variable pollutant emissions.

EMISSION SOURCE CATEGORIES FOR PRIMARY PM EMISSIONS

On-Road Mobile Sources

Source Types. Approximately 235 million on-road vehicles were registered in the United States in 2002.[16] Passenger cars accounted for 136 million of these vehicles; 93 million were trucks, and 5 million were motorcycles. In addition, there were 1.8 million truck tractors (heavy-duty trucks) and 800,000 buses registered.[16]

Passenger cars, as well as light- and heavy-duty pickups, vans, and sport utility vehicles are dominated by spark-ignition gasoline-powered engines. In contrast, the heaviest classes of trucks are powered predominantly by compression-ignition (diesel) engines. Although heavy diesel trucks comprise a small fraction of the total fleet, they contribute proportionally more to total vehicle miles traveled (VMTs) and consume more fuel per mile. Trucks with more than 26,000-lb gross vehicle weight make up approximately 3% of the truck fleet but contribute more than 10% of all truck VMTs and more than 23% of all truck fuel used.[17]

Emission Estimation Methods and Magnitude of Emissions. On-road mobile sources emit two major categories of primary PM: exhaust emissions and fugitive dust (road dust, brake wear, and tire wear). During the period from 1990 to 2000, on-road mobile sources were estimated to have emitted 34% of the national $PM_{2.5}$ emissions,[18] as reported in the National Emissions Inventory (NEI). Road dust is estimated to make up approximately 89% of these on-road $PM_{2.5}$ emissions. Because of the large fraction of total primary $PM_{2.5}$ emissions that are attributed to road dust in the NEI, and because of the uncertainties in these emission estimates, the analyses in this review focus on road dust as a separate emission category. Whenever percentages are reported, they are given as a fraction of the primary emissions, excluding fugitive dust. Primary PM exhaust emissions depend on the vehicle class, vehicle age, fuel type and sulfur content, vehicle mileage, vehicle maintenance, and the emission controls used in the vehicle.

There are two distinct approaches to estimating primary on-road mobile-source PM emissions: estimation of emissions based on fuel consumption and estimation of emissions based on VMTs. Emission estimates based on fuel consumption require knowledge of the total amount of fuel sold for a geographic region and an experimentally determined average emission rate per unit mass of fuel burned. Emission factors have been determined for typical mixes of on-road diesel and gasoline engines.[19–23] There are several limitations to emissions estimated using this approach. An in-depth description of the on-road fleet characteristics and operating conditions is not considered, and special efforts must be made to incorporate cold-start and other classes of emissions. Another serious concern with this method is the accuracy of the emission factors. The algorithms used to determine emission factors are based mostly on engine and vehicle certification tests, and it is not clear how well they relate to actual driving conditions.[24] Several studies have shown that emissions can increase significantly under driving conditions that are outside the range of certification tests.[24–26] Few studies have specifically evaluated this variability for PM emissions, but the work on CO and nitrogen oxide (NO_x) emissions indicates that engine performance is highly variable and that similar variability in PM emissions is likely.

Emissions estimates can also be based on VMTs and emission factors. This approach requires detailed characterizations of the on-road fleet, such as the vehicle models, mileage, and age, as well as the total VMTs by each vehicle class. Estimates of emission rates (in mass/distance traveled) and the VMTs for each vehicle class in the geographic area of interest are typically obtained using software tools developed by EPA (MOBILE 6.2, previously PART5)[27] and the California Air Resource Board EMFAC (Emission Factor Model).[28] VMT data are usually available at much greater spatial and temporal resolution than fuel consumption data.

The fuel-based and VMT-based approaches are complementary, however, and when used in concert can be used to characterize uncertainties in emissions inventories. For example, Pokharel et al.[29] compared fuel-based estimates (from remote sensing data) and MOBILE 6 estimates of CO, HC, and NO emissions in the Denver, CO, area, and found that MOBILE 6 estimates were 30–70% higher for CO, 40% lower for HC, and 40–80% higher for NO than fuel-based estimates. Dreher and Harley[30] made a similar comparison for fine elemental carbon (EC) emissions from diesel trucks in the San Francisco Bay area of California. They determined the fuel-based emission factors from measurements taken in the Caldecott tunnel, east of San Francisco. Dreher and Harley[30] compared the fuel-based estimates to the California Air Resources Board (CARB) model estimates in 1996 and found that the upper bound from the fuel-based method was 4.5 times greater than the EMFAC model prediction on a typical weekday. When the CARB estimates were corrected so that total activity (fuel use or VMT) corresponded with the total activity predicted by Dreher and Harley,[30] the fuel-based EC estimate was still a factor of 3.6 higher than the VMT estimate. It is important to note the limitations of making such comparisons. These estimation methods often take measurements made under specific driving conditions and apply them to all on-road traffic. Each method may use different assumptions about driving conditions. Therefore, these comparisons cannot be used to quantify the exact differences in estimation methods. However, they do suggest that currently there is a relatively wide uncertainty band (a factor of $\geq$2) in on-road tailpipe emissions as characterized by fuel-based and VMT-based estimates.

Although estimates of tailpipe emissions have been developed and refined for decades, there is little information on and considerable uncertainty in the magnitude of noncombustion PM emissions, such as tire wear and brake wear. Comparisons between ambient measurements of tire wear source signatures and emission estimates based on activity indicate that tire wear may be underestimated in activity-based emissions inventories. Schauer et al.[13] performed source apportionment for four locations in the Los Angeles area over a 2-day period in the summer of 1993. At one site they were not able to measure any tire wear particulate. At the other sites they found that tire wear made up between 1% and 3.4% of total $PM_{2.5}$. An evaluation of CARB's emissions inventory showed that, in Los Angeles County, tire wear constituted approximately 0.65% of $PM_{2.5}$ emissions in the summer of 1990 and approximately 0.71% of $PM_{2.5}$ emissions in the summer of 1995. Although these are not direct comparisons because the exact locations and times are not the same, the results can be evaluated as a crude comparison and suggest that activity-based emissions inventories underestimate tire wear.

Finally, there is also uncertainty in brake wear emissions. The EPA emission factor that is used for brakes comes from a 1983 study by Cha et al.[31] and is approximately 8 mg/km. This study was performed on brakes composed of asbestos and may no longer be accurate for today's nonasbestos brakes. Two more recent studies examining emissions from brakes used chamber data and roadside measurements to estimate emissions and found emission rates that varied by up to an order of magnitude.[32,33] Bhagwan et al.[32] estimated that 3–9 mg/km of PM are emitted from brakes, whereas Abu-Allaban et al.[33] estimated that 0–80 mg/km of PM are emitted from brakes. The discrepancies in these studies and the changes in brake composition suggest that brake emissions estimates need further examination.

Spatial and Temporal Distribution. At spatial scales above 1 km, the distribution of on-road emissions is performed by distributing the emissions along highway segments based on data on the total miles traveled for major roadways by vehicle class.[34]

At spatial scales less than 1 km, the situation is more complex. Several studies have shown that fine[35] and ultrafine[36] particle concentrations drop off rapidly near roadways. Kuhn et al.[35] found that concentrations of $PM_{2.5}$ nitrate and $PM_{2.5}$ EC were greater near a highway in Southern California, but concentrations of crustal elements (Na, Mg, Al, Si, S, Cl, K, Ca, Ni, Zn, and Ba) and coarse PM were similar both near and far from the highway. Zhu et al.[36] found that number and surface area of ultrafine PM dropped off rapidly in the first few hundred meters from the roadway. These results suggest that atmospheric processing of on-road PM emissions occurs rapidly near roadways and that exposure to some types of on-road emissions will be strongly dependent on distance from the roadway source.

Based on these results and the fact that fuel-based emission estimates are generally based on statewide fuel sales data, the spatial and temporal allocation of on-road emissions should be based on VMTs or similar information derived from travel demand modeling. Even if more spatially and temporally resolved data were available for fuel sales, the data would be difficult to apply to spatial and temporal emission allocation, because vehicles can refuel in one place and then proceed to travel long distances.

Composition. Primary particles from on-road sources are typically characterized as sulfates, carbonaceous PM (made up of organic carbon [OC] and EC), lead, brake wear PM, or tire wear PM. On a mass basis, a majority of fine PM emissions are carbonaceous. Schauer et al.[37] observed that $PM_{2.5}$ emitted from catalyst-equipped, gasoline-powered vehicles on dynamometers was 54% carbon or two thirds or more carbonaceous if typical ratios of carbon to OC of 1.2–1.4 are used. Higher carbonaceous fractions were reported for noncatalyst-equipped vehicles.

The ratio of OC to total carbon (TC) in on-road vehicle emissions varies by vehicle class and driving conditions, as well as EC measurement methods. Schauer et al.[37] observed a ratio of ~0.81 for the dynamometer study described above. During the Northern Front Range Study, ratios of 0.7 and 0.4 were observed for gasoline vehicles and diesel vehicles, respectively.[38,39] The OC/TC fraction of $PM_{2.5}$ released by mixed traffic in the Sepulveda Tunnel in Los Angeles was 0.47.[40] This fraction was 0.7 for $PM_{2.5}$ released from mixed traffic in a tunnel in Milan, Italy,[41] and 0.28 and 0.6, respectively, for heavy-duty diesel vehicles and light-duty vehicles in the Caldecott Tunnel east of Oakland, CA.[22]

Shah et al.[42] used a mobile emissions laboratory attached to the tailpipe of heavy-duty diesel trucks to measure OC and EC emissions under varying driving conditions. The OC/TC ratio for $PM_{2.5}$ was 0.80 during initial cold start and idling, 0.62 in stop-and-go traffic, 0.30 for light-to-medium traffic arterial road driving, and 0.35 for highway driving. These results are summarized in Table 1. The large variations in PM composition with changing driving conditions are likely to introduce uncertainties when OC and EC in ambient samples are used to assess the magnitude of diesel and gasoline traffic emissions. The speciation of $PM_{2.5}$ is also estimated by MOBILE 6.2. Sulfate emission rates are estimated based on catalyst type, air injection type, average speed, and fuel economy. Brake and tire wear

Table 1. Summary of OC/TC ratios in PM emitted from on-road vehicles.

Reference	Vehicle Type	Measurement Approach	PM_{10} or $PM_{2.5}$	OC/TC (g/g)
37	Light-duty gasoline	From tailpipe on dynamometer	$PM_{2.5}$	0.81
38, 39	Light-duty gasoline	From tailpipe on dynamometer	PM_{10}	0.65–0.74
38, 39	Light-duty diesel	From tailpipe on dynamometer	PM_{10}	0.35–0.45
40	Mixed heavy-duty (3%) and light-duty (97%) gasoline and diesel	From ambient air in tunnel	$PM_{2.5}$	0.47
41	Light-duty gasoline and diesel	From ambient air in tunnel	$PM_{2.5}$	0.7
22	Heavy-duty diesel	From ambient in air tunnel	$PM_{2.5}$	0.28
22	Gasoline and diesel	From ambient in air tunnel	$PM_{2.5}$	0.6
42	Heavy-duty diesel	From tailpipe during cold-start and idling conditions	$PM_{2.5}$	0.8
42	Heavy-duty diesel	From tailpipe during stop-and-go traffic driving	$PM_{2.5}$	0.62
42	Heavy-duty diesel	From tailpipe during light-to-medium traffic driving	$PM_{2.5}$	0.3
42	Heavy-duty diesel	From tailpipe during highway driving	$PM_{2.5}$	0.35

$PM_{2.5}$ are estimated using the same factors for all of the vehicle classes. OC and EC are estimated for diesel vehicles (these categories are combined for gasoline powered vehicles because of lack of data) by subtracting all of the other classes of PM from total PM, then assigning the remaining $PM_{2.5}$ to OC and EC based on typical ratios of organic and EC in diesel exhaust.

These attempts at speciation again may not capture much of the variability present with different operating conditions. Sulfate emission rates, which are dependant on speed, are one example. MOBILE 6.2[43] uses two data points for emission rates, one for emissions at 19.6 miles per hour (mph) and one for emissions at 34.8 mph. The program uses a linear interpolation for emission rates from vehicles traveling between these speeds. All of the travel at speeds below 19.6 mph uses the value for 19.6 mph, and all of the travel at speeds above 34.8 mph uses the value for 34.8 mph. In addition, sulfate emission data are based on tests performed on vehicles using fuel containing 340 ppm of sulfur. To determine emission rates for vehicles using new, lower sulfur fuel, the emission factors are linearly scaled based on the sulfur fuel content. The accuracy of this method is untested.

Size Distributions. Mobile sources generally emit particles in the ultrafine (<0.1 μm diameter) range[43–45]; number concentrations are typically greatest for particles with diameters between 10 and 20 nm.[35,36] These initial size distributions change rapidly, as a function of distance from roadways, because of coagulation, evaporation, or condensation of semivolatile organics; deposition; and other processes. These processes and source characteristics vary with temperature and other atmospheric conditions, creating seasonal variations in the particle size distributions near roadways. Zhu et al.[36] report that for similar fleets and wind fields, the near-roadway concentrations of particles in the 6–12 nm range were greater in winter but these concentrations decreased faster than those during the summer as the distance from the roadway increased. Because diesel vehicles contribute higher levels of low-volatility particulate than gasoline vehicles,[35] seasonal variations in particle sizes from mobile sources will depend on fleet composition.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. On-road mobile source emissions of $PM_{2.5}$, reported for 1990–1999 through the NEI for the regions of the United States in which EPA Supersites were located, indicate that on-road mobile sources contributed 12% of nondust $PM_{2.5}$ in California; 8% in Texas and Missouri; 10% in New York, Pennsylvania, and Maryland; and 5% in Georgia (national average = 4%).[46] In addition, on-road mobile sources contributed 6% of nondust $PM_{2.5}$ in Arizona from 1996 to 2001. These source strengths and the emissions inventory information described above are summarized in Table 2. The source strengths based on emission inventories can be compared with source apportionment studies done as part of the Supersite and other field measurement programs; however, in these source apportionment studies, it is not possible to separate the contributions of emissions from

Table 2. Summary of on-road emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	4% of nondust emissions
Major source types	Gasoline and diesel vehicles (cars, trucks, motorcycles)
Bottom-up approach	Data on fuel consumption and emission factors from tunnel studies; data on emissions per distance traveled and estimates of distance traveled by vehicle type
Top-down approach	Molecular markers of diesel/gasoline engine combustion products
Techniques used to spatially allocate emissions	Emissions scaled to total vehicle miles traveled on each road segment
Techniques used to temporally resolve emissions	Based on surveys of driving behavior and data on roadway use
Composition	Varies with driving conditions and vehicle: typical composition is 30–40% OC and 50% EC (diesel) and 60% OC and 20–30% EC (gasoline)
Size fraction	Ultrafine: particle peaks at ~10 and 20 nm

on-road and nonroad sources; therefore, the comparisons are presented after estimates of nonroad mobile source emissions are described.

Nonroad Mobile Sources

Source Types. Nonroad sources are diverse in both their engine design and fuel use. They can include diesel engines in off-road vehicles, military equipment, trains, and ships, as well as gasoline engines in recreational vehicles, lawn care equipment, and some aviation vehicles. Approximately 66% of the 140 million estimated nonroad engines are associated with residential lawn and garden equipment, which are dominated by small spark-ignition engines.[47] A total of 95% of all equipment uses gasoline, 4% uses diesel, and less than 1% uses compressed natural gas (CNG) and liquefied petroleum gas (LPG).

Although lawn and garden equipment and gasoline-powered engines dominate the number of nonroad engines, emissions of $PM_{2.5}$ are largely attributed to diesel equipment. According to the 1990–2000 NEI data for $PM_{2.5}$ emissions, diesel engines account for at least 60% of nonroad emissions, whereas gasoline and CNG or LPG engines account for 15% and 1%, respectively, of total nonroad emissions. Recreation and lawn and garden equipment represent 7% of all nonroad $PM_{2.5}$ emissions, construction equipment represents 31%, industrial equipment represents 8%, farm equipment represents 15%, aircraft represent 4%, marine vessels represent 11%, and rails represent 10%.[47]

Estimation Methods and Magnitude. During 1990–2000, nonroad mobile sources contributed 8% of national $PM_{2.5}$ emissions on average, excluding fugitive dust.[18] Most nonroad mobile equipment emissions are estimated using EPA's NONROAD model,[47] with the exception of aircraft, ship, and locomotive emissions. The NONROAD model considers 80 engine categories of equipment. Emission factors within each category are scaled by power level and adjusted for in-use operation, engine deterioration, and fuel sulfur content. Differences in operation are accounted for using transient adjustment factors, which were determined by study of high-load and low-load conditions for tractors, crawler dozers, backhoes, rubber tire loaders, skid steer loaders, arc welders, and excavators. Each engine category is assigned a high-load factor, a low-load factor, or no factor, and pre-1998 vehicles are given default high- and low-load factors of 1.23 and 1.97, respectively. Adjustments for engine deterioration are accounted for using deterioration factors taken from the CARB OFFROAD model. The final emission factor adjustment accounts for decreases in fuel sulfur content from the pre-1998 value of 3300 ppm.

The limited number of verifications of nonroad emission factors that has been conducted illustrates the variability in nonroad emissions based on fuel choice, controls used, and other factors not currently incorporated into nonroad emissions inventories. Alander et al.[48] found that PM emissions from a single two-stroke chain saw could vary by 4% to 16% even when tested under a single set of operating conditions. When conditions such as presence of a catalyst and fuel type and lubricating oil type were varied, the emissions differed by almost a factor of two. The presence of a catalyst had the largest effect on PM emissions and reduced PM mass emissions by 19–50% but resulted in a larger number of total particles being formed. The study also found that emissions from an engine without a catalyst increased when reformulated fuel and mineral lube oil were used together or when alkyl fuel and biodegradable lube oil were used together but decreased when the fuel and lube oil pairs were exchanged. Christensen and Westerholm[49] evaluated PM emission factors for lawnmowers (primarily four-stroke engines) and found emission factors that also varied when tested under a single set of operating conditions and reductions in emissions with a catalyst. In contrast to the previous findings, however, they found that emissions did not vary significantly with two different fuels that were tested as long as no catalyst was present.

Nonroad emissions from aircraft, ships, and locomotives are not estimated using the NONROAD model. Instead, aircraft emissions are estimated from engine types and landing/takeoff cycles; locomotive emissions are estimated based on actual fuel usage and track mileage,[50] and ship emissions are estimated by multiplying NO_x emission estimates by a PM/NO_x ratio based on large uncontrolled diesel engines.[51] Emissions from these sources are even less documented than the other nonroad sources. In the case of ship emissions, this can be explained by a lack of data regarding the fuels used by ships. Corbett and Koehler[52] estimated that total PM emissions in 1999 from ships (on rivers and in coastal areas) for the states of Oregon and Washington were approximately 443 t/yr, which is approximately 0.3% of nondust, primary $PM_{2.5}$ emissions. Current data are not sufficient to form a national ship emissions inventory for PM; however, several data collection efforts have been initiated.

Temporal and Spatial Resolution. The NONROAD model estimates emissions on national, state, or county levels based on proprietary data on equipment populations from Power Systems Research (PSR), a market research company.[47] PSR provides national population estimates and the NONROAD model estimates county populations for each equipment type by using socioeconomic spatial allocation surrogates. The county equipment population estimates have much uncertainty, and EPA encourages state and local agencies to perform surveys or to use other measures to derive local estimates. However, such surveys are costly, and only a few have been performed.[53] Temporal emission allocations are based on data from the Nonroad Engine and Vehicle Emission Study, CARB's OFFROAD model, and EPA assessments of typical use.[54,55] Allocations are performed on seasonal, monthly, and daily time scales. The lowest allocations are assigned to winter months: 15% of the emissions are allocated to winter months, 36% are allocated to summer months, and 24% of emissions are allocated to fall and spring months. Allocations also vary by equipment use: equipment that is used daily, such as for recreational, construction, or agricultural purposes, is allocated consistent emissions for every day of the week, whereas lawn and garden equipment is allocated 50% higher emissions on weekdays than on weekends. On a daily basis, most nonroad emissions are allocated using a single diurnal profile with peak emissions occurring between 9:00 a.m. and 9:00 p.m.[47]

Composition. Limited information regarding the composition of PM emissions from nonroad engines is available. Liang et al.[56] found that 82–86% of total PM emissions from a diesel generator were OC, although only 3.5% of the emissions could be speciated to greater detail. In their study of lawnmower emissions, Christensen and Westerholm[49] found that fuel type and catalyst affected the polyaromatic HC (PAH) content of the PM. Emissions from engines using a catalyst with the alkylate fuel had the lowest PAH content. PAH emissions were also observed in chain saw emissions.[57] Particle-phase PAH concentrations between 6 and 28 μg/m^3 were measured in chain saw exhaust when aliphatic petroleum fuel with ester-based synthetic lubricating oil was used. These concentrations were much higher (48–130 μg/m^3) when conventional fuel with mineral lubricating oil was used.

Size Distributions. The NONROAD model assumes that 100% of particulate is less than 10 μm and that 92% of particulate from spark ignition and 97% of particulate from compression ignition (diesel) engines is less than 2.5 μm. Very little work has been done to further quantify these size distributions. More research will need to be performed to understand how the size fraction of nonroad PM emissions varies with fuel type, operating conditions, weather conditions, and other factors.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. Nonroad mobile source emissions of $PM_{2.5}$, reported through the NEI for the period from 1990 to 1999, for the regions of the United States in which EPA Supersites were located, indicate that nonroad mobile sources contributed 14% of nondust $PM_{2.5}$ emissions in California; 13% in Texas and Missouri; 11% in New York, Pennsylvania, and Maryland; 6% in Georgia; and 7% in Arizona.[46] Combined on-road and nonroad mobile source emissions contributed 26% of total $PM_{2.5}$ emissions in California; 21% in Texas and Missouri; 21% in New York, Pennsylvania, and Maryland; 11% in Georgia; and 13% in Arizona.[46] These source strengths and the emissions inventory information described above are summarized in Table 3.

Table 3. Summary of nonroad emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	8% of nondust emissions
Major source types	Construction equipment, agricultural equipment, aircraft, trains, ships, and lawn care equipment
Bottom-up approach	Data on equipment populations and emissions factors developed for some types of equipment
Top-down approach	Molecular markers of diesel/gasoline engine combustion products (not easily distinguishable from on-road diesel emissions)
Techniques used to spatially allocate emissions	Emissions scaled to county equipment population data
Techniques used to temporally resolve emissions	Seasonal, weekly, and diurnal patterns determined for typical use
Composition	Mostly carbonaceous but not well characterized
Size fraction	Mostly <2.5 μm but not well characterized

Comparisons of these bottom-up inventory estimates with the results of source apportionment studies performed during field programs, such as the PM Supersites Program, indicate inaccuracies in the nonroad and mobile emissions inventories. A summary of these comparisons is provided in Table 4. Source apportionment studies for the San Joaquin Valley are generally consistent with statewide California estimates (26% of $PM_{2.5}$ because of mobile sources), although studies for Los Angeles indicate higher percentages of PM because of mobile sources that are higher than statewide averages.[58–61] For example, 26% of primary emissions, excluding fugitive dust, are attributed to vehicle exhaust in the San Joaquin Valley during winter. For an episode on September 24–25, 1996, in the Los Angeles area, 28% of nondust emissions were attributed to diesel engines, 21% were attributed to gasoline engines with catalytic converters, 15% were attributed to gasoline engines without catalytic converters, and 4% were attributed to high-sulfur fuel combustion.[58–62] Schauer et al.[13] performed source apportionment at four sites in the South Coast region of California. The samples were collected over 48 hr during a severe smog event and showed that mobile sources contributed between 39% and 57% of the primary, nondust $PM_{2.5}$ depending on the site.

Fraser et al.[63] report that, at two Houston-area sites, mobile sources contribute approximately 25–33% of total PM. These predictions are close to the level of on- and off-road emissions predicted by the NEI for Texas and Missouri (21% because of mobile sources).

A source apportionment study for Philadelphia, PA,[58] reports that 76–88% of primary nondust PM comes from mobile sources. This is much greater than is predicted by the NEI (approximately 21% for on-road and nonroad vehicles). This source apportionment study, however, found that most PM in the area was secondary sulfate, and it only evaluated two primary nondust sources (mobile sources and residual oil burning). Therefore, the lack of other primary sources in the study may have caused an overestimation of the mobile source contribution to PM in the area.

A 2-yr study from 1998 to 2000 in Atlanta[11] estimated that 56% of primary $PM_{2.5}$ (excluding fugitive dust) came from on-road sources, with an additional 8% coming from railroad traffic. These estimates are significantly higher than the emissions inventory prediction for Georgia (11% because of mobile sources). One possible explanation for the difference in source contribution estimations is that PM observed in downtown Atlanta is not representative of regional PM. Zheng et al.[15] evaluated 5 months of data from the same Atlanta monitoring site and found that 42% of the primary $PM_{2.5}$, excluding dust, could be attributed to diesel exhaust, and 6% could be attributed to gasoline exhaust. These estimates are smaller than those reported by the previous work but are still larger than the fraction estimated in the NEI for Georgia.

Table 4. Comparison of top-down and bottom-up mobile source emissions inventories.

Location	Inventory Estimates National Emissions Inventory Average for 1990–2000 (% of nondust emissions)[46]			Source Apportionment Estimates (% of nondust emissions) On-Road and Nonroad Are Usually Not Separated	Reference (source apportionment)
	On-Road	Nonroad	On-Road + Nonroad		
California	12	14	26		
San Joaquin Valley				~26 (winter)	58–61
Los Angeles			18	39–68	13, 58–62
Texas/Missouri	8	13	21		
Houston				25–33	63
New York/Pennsylvania/Maryland	10	11	21		
Philadelphia				76–88	54
Georgia	5	6	11		
Atlanta				48–64	12, 16
Arizona			13		
Phoenix				~80	64
National average	4 (34 when road dust is included)	8	12	31–74	12, 16, 115–119

Although there was no supersite in Arizona, other field studies have been performed there. For Phoenix, AZ, Lewis et al.[64] found that a third of weekday, nondust, primary contributions were because of diesel engine emissions and that roughly half was because of gasoline engine emissions. The diesel contribution decreased on weekends, whereas gasoline increased. This is significantly greater than the NEI prediction of 13%.

The inconsistencies addressed here highlight the fact that receptor models often provide profiles that are only relevant to a limited area and time frame. Because there is a great deal of spatial and temporal variation in mobile source emissions, ambient measurements may not be broadly applicable on a statewide scale. However, it will be important to determine whether the discrepancies between the various source apportionment techniques and between top-down and bottom-up emissions inventory methods are because of inconsistent time and spatial scales or more fundamental differences in emissions estimates. If the latter is the main factor, these discrepancies will need to be resolved.

Summary for Mobile Source Emissions. There are significant uncertainties in the magnitude of on-road mobile source emissions estimates. The magnitude of these uncertainties is exemplified by the fact that the limited studies comparing fuel-based inventories and VMT-based inventories differ by up to a factor of two to four. Other areas of uncertainty include the prediction of tire wear and brake wear emissions, which may have uncertainties of up to an order of magnitude. Temporal allocations of on-road mobile source emissions are believed to be accurate in locations where in-depth traffic studies have been performed. However, many areas of the country lack the travel demand information needed to temporally allocate these emissions at a fine enough scale to use in photochemical models. Spatial allocations of these emissions are limited by availability of traffic information on a large scale and by the uncertainty in the change of particle size distribution with distance from roadways. Although it is well established that the fine PM in mobile emissions is mostly carbonaceous, the fraction of the carbon that is EC versus the fraction that is OC can vary significantly by source type, engine age, driving conditions, and measurement method. This variability must be characterized before OC/EC split fractions can be accurately used to attribute sources of gas and diesel mobile sources using EC as a tracer.

Nonroad emissions estimates are even more uncertain than on-road emissions, because the emissions from many nonroad sources are still uncharacterized or not characterized well enough to account for differences in engine size and type, fuel type, oil type, and typical engine loads for a single source. In addition to uncertainty in the emission factors, there is also a great deal of uncertainty in the activity factors for nonroad sources. Although EPA has population estimates of construction equipment in many counties, a study in Houston showed that these default estimates can have significant uncertainties.[53] In many cases, temporal allocations are general and not based on studies of specific types of equipment. Finally, there is little information on size and composition of PM emissions from nonroad equipment.

Cooking

Source Types. Cooking emissions are released by the commercial, institutional, and residential sectors; however, the majority of information documenting these emissions pertains to the commercial sector. Commercial cooking sources include underfired char broilers, automated char broilers, clamshell griddles, flat griddles, and deep-fat fryers.[65,66]

Estimation Method and Magnitude. Although the California state emissions inventory has inventoried $PM_{2.5}$ emissions from cooking sources for some time, this emission source has only been included in the NEI since 2002. CARB[67] estimates that in 2004, 4.4% of $PM_{2.5}$ emissions in California, 8.8% of $PM_{2.5}$ emission in Los Angeles County, and 2.1% of $PM_{2.5}$ emissions in Fresno County, CA, came from cooking. Regional emissions estimates from the 2002 NEI are not yet available.

Similar cooking emissions estimations methods have been developed by CARB[66] and EPA.[68] Both entities estimate the activity rate from the number of each type of cooking equipment used at food establishments and the average rate of food cooking (pounds of food per year) for each type of equipment. These activity data are multiplied by emission factors (pounds of PM per 1000 lbs of food cooked) for each type of cooking equipment. CARB and EPA emission factors are very similar. For example, for underfired char broiling, EPA uses an emission factor of 31.6 lb $PM_{2.5}$/t of food cooked, whereas the Sacramento Air Quality Management District uses a factor of 32 lb/t. For flat griddle frying, EPA factor is 4.5 lb/t and the CARB factor is 5.6 lb/t, and for clamshell griddle cooking, EPA factor is 0.85 lb/t and the CARB factor is 2 lb/t.[66,68]

Temporal and Spatial Resolution. Cooking emissions scale with the population and the prevalence of restaurants and are therefore highest in residential areas and lowest in rural and industrial areas. Temporal patterns will vary by location, but in the Bay Area of California, restaurant sales are usually higher in the summer months.[65] For spatial allocations, the Sacramento Metropolitan Air Quality Management District account scales the number of eating establishments to the population; the Bay Area Air Quality Management District uses countywide taxable restaurant sales data to determine the distribution of restaurants at the county level.

Composition. A series of studies have examined the composition of PM emitted from cooking and found that the emissions consist mostly of organic matter. Kleeman et al.[69] found that the $PM_{2.5}$ emissions from char broiling meats were almost entirely organic, with trace quantities of sodium ions, potassium ions, sulfate ions, nitrate ions, chloride ions, aluminum, strontium, and barium. Hildemann et al.[70] found the $PM_{2.5}$ from frying and char broiling to contain 68–73% organic matter, with only trace quantities of potassium. Potassium in cooking emissions is important, because potassium is also used as a tracer for wood smoke.[71] Studies that have speciated the organic portion of the $PM_{2.5}$ emissions have identified unique organic tracers that can be used in source apportionment modeling.[71,72] Schauer et al.[71] identified 71 organic compounds in the low-volatility vapor and particle phases resulting from meat char broiling and frying operations. *N*-Alkanoic acids (e.g., *n*-hexadecanoic acid or palmitic acid), *n*-alkenoic acids (e.g., 9-octadecenoic acid or oleic acid), and carbonyls (e.g., *n*-aldehydes and alkan-2-ones) were present in the highest concentration, although carbonyl concentrations are highest in char broiling operations, and oleic acid concentrations are highest in frying operations. The difference in relative emission rates of these two acids can be used to distinguish between meat char broiling and frying emissions in source apportionment studies.[72]

Size Distributions. The size distribution of $PM_{2.5}$ is expected to vary by cooking operation. Kleeman et al.[69] report that size fractions or particulate from char broiling meat peak between 0.1 and 0.2 μm, although some larger particles 3 μm or less were observed. CARB found that char broiling results in a larger fraction of fine particles, whereas deep-frying results in a higher fraction of large particles. As a result of this difference, 62% of the PM emissions from cooking operations in California in 2004 were estimated to be $PM_{2.5}$.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. Cooking emissions estimates by CARB (approximately 4% of nondust emissions in the state) are approximately equal to source apportionment estimates developed for Los Angeles (estimates were unavailable for the San Joaquin Valley), which ranged from 2% of $PM_{2.5}$[59–61] to 5–11% of $PM_{2.5}$.[13] In contrast, Fraser et al.[63] report that, for Southeast Texas, cooking emissions account for 10–20% of total $PM_{2.5}$ emissions. Finally, Zheng et al.[15] predicted that meat cooking contributed an average of 6% to primary, nondust $PM_{2.5}$ in Atlanta. Summaries of these comparisons and the emissions inventory information described above are provided in Tables 5 and 6. Again, Texas and Georgia measurements cannot be compared with bottom-up estimates, because regional data from the 2002 NEI are not yet available.

Summary for Cooking Emissions. Estimation methods for cooking emissions are still being developed, and the exact magnitude of this source is still unknown. However, for the few cities mentioned in this work, ambient and bottom-up emissions estimates are in reasonable agreement, indicating that PM from commercial cooking establishments contributes 2–20% of the mass of primary $PM_{2.5}$ emissions. Refining these estimates will be challenging because of the myriad differences in cooking activity and emission factors and the lack of truly unique tracer species for this source. Although potassium makes up approximately 0.2–0.4% of total $PM_{2.5}$ mass from cooking emissions, it also contributes 0.2–0.8% of total $PM_{2.5}$ mass in wood smoke.[70,73] Thus, estimates of cooking emissions based on potassium measurements must correct for wood

Table 5. Summary of commercial cooking emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	No data available from 1990–2000; cooking was included in the 2002 NEI
Major source types	Char broilers and deep fat fryers
Bottom-up approach	Inventories done for Los Angeles scaled by population
Top-down approach	Molecular tracers of meat cooking
Techniques used to spatially allocate emissions	Scaled to population or restaurant sales data
Techniques used to temporally resolve emissions	Temporal profiles determined based on monthly restaurant sales
Composition	68–73% carbonaceous (palmitic and oleic acids are potential tracers), trace amounts of potassium and other ions
Size fraction	62% <2.5 μm with number concentration peaks at 0.1 and 0.2 μm

Table 6. Comparison of top-down and bottom-up cooking emissions inventories.

Location	Inventory Estimates CARB Emissions Inventory Average for 2004 (% of nondust emissions)[67]	Source Apportionment Estimates (% of nondust emissions)	Reference (source apportionment)
California	4		
Los Angeles	8	2–11	13, 59–61
Houston		10–20	63
Atlanta		6	15

smoke emissions, which can vary significantly, as discussed in the next section. The use of other tracers (EC and levoglucosan for wood smoke and palmitic and oleic acid for cooking) in combination with potassium may help distinguish cooking from wood smoke emissions.

Residential Fuel Combustion

Source Types. Residential fuel combustion emission sources include wood combustion (wood stoves and fireplaces), natural gas space heaters, distillate fuel oil space heaters, natural gas water heaters, natural gas cooking, other natural gas combustion, and liquefied petroleum fuel combustion.

Estimation Method and Magnitude. During 1990–2000, residential fuel combustion sources accounted for 11% of the national $PM_{2.5}$ emissions, on average, excluding fugitive dust,[19] and 18% of the primary $PM_{2.5}$ emissions in California; 14% in Texas and Missouri; 17% in New York, Pennsylvania, and Maryland; and 7% in Georgia.[46] The CARB inventories were consistent with the NEI, with a statewide estimate of 14.7% and estimates in Los Angeles and Fresno of 5.7% and 9.8%, respectively. Most of the $PM_{2.5}$ emissions from this source are associated with wood combustion (e.g., 95% in California), with smaller contributions from natural gas heaters.

Estimates of $PM_{2.5}$ emissions from wood stoves are calculated from activity rates and emission factors. Activity rates are based on the wood fuel consumption rate and the number of homes that use wood-burning stoves for heating. Emission factors are taken from EPA's AP-42. Estimation methods used by CARB and EPA only vary in the estimation of activity rate. CARB estimates activity based on average daily temperature, surveys of fuel use as a function of temperature, and the number of wood-burning fireplaces obtained from the U.S. census. EPA estimates activity based on climate zone, type of wood-burning appliance (e.g., fireplaces: without inserts; fireplaces: inserts—catalytic, non-EPA-certified; fireplaces: inserts—noncatalytic, EPA-certified; fireplaces: inserts—catalytic, EPA-certified; wood stoves: conventional; wood stoves: catalytic; and wood stoves: noncatalytic), and whether the county is urban or rural.

Temporal and Spatial Resolution. EPA distributes residential fuel combustion emissions spatially and temporally by designating seasonal scaling factors for each of five climate zones. In the temperate state of California, residential fuel combustion is spatially allocated at the county level using census data for the number of wood-burning households and temporally allocated based on daily, weekly, and monthly activity patterns. Burning is assumed to be uniform throughout the week but variable over the course of a day (i.e., mostly in the morning and early evening). In addition, burning activity is restricted to the months of October through April with a peak in December, January, and February.

Composition. Several studies have investigated the composition of wood smoke emitted from fireplaces. These studies found the particle mass is predominantly composed of OC (43–59%) with smaller amounts of EC (1.4–5%).[70,73] Trace elements are found in varying amounts in different types of wood, which compromise their use in source apportionment studies. Sulfate, nitrate, chloride, ammonium, potassium, bromine, titanium, iron, rubidium, aluminum, zinc, barium, and strontium have been measured in varying quantities in hardwood and softwood smoke.[69] Potassium has been a potential tracer for wood smoke, and various studies have found that it makes up 0.28–0.88% of emitted particle mass.[70,73] However, because it is also found in other sources (meat cooking), its use as a tracer for wood smoke is problematic. Speciation of the organic fraction of wood smoke PM has been able to identify between 30% and 50% of the $PM_{2.5}$ mass emitted.[73,74] These studies found that levoglucosan, a pyrolysis product of cellulose, is commonly found in all wood smoke and has the potential to be a useful tracer.[73] As with potassium, however, the amount of levoglucosan emitted varies with wood type and combustion conditions. Finally, several tracers can be used to distinguish between hardwood and softwood smoke. Schauer et al.[14] assert that some of the most useful tracers for this task would be propionylsyringol and butyrylsyringol from hardwood and resin acids from softwood.

Size Distributions. CARB estimates emissions of PM, coarse PM (PM_{10}), and $PM_{2.5}$ from residential fuel combustion. More than 90% of wood combustion PM is assumed to be less than 2.5 μm, and 100% of natural gas and liquid petroleum gas is assumed to be less than 2.5 μm. EPA assumes that 100% of PM emitted from wood combustion is less than 2.5 μm. Kleeman et al.[69] reported that overall PM size distribution peaked between 0.1 and 0.2 μm.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. Comparison of the estimates of $PM_{2.5}$ emissions to ambient observations is difficult because of the variable contribution of wildfires, prescribed burns, and other outdoor fire sources. Because of a lack of unique tracer species, source apportionment studies cannot differentiate between wood smoke from residential fuel combustion and outdoor fires. Therefore, observational data on the contribution of biomass burning should be viewed as an upper bound of the potential contribution of residential burning. A summary of top-down and bottom-up comparisons for residential fuel combustion emissions estimates and the emissions inventory information described above are provided in Tables 7 and 8.

Observational data collected in California suggest that in excess of the 18% of $PM_{2.5}$ can be attributed to this source type. In the San Joaquin Valley in autumn, observational data suggest that 32% of the primary, nondust $PM_{2.5}$ in the San Joaquin Valley is from residential fuel combustion.[1] In Los Angeles, observational data suggest that PM from wood burning is less than 30% of the PM_{10} and less than 29% of the observed $PM_{2.5}$.[1] Schauer et al.[14] predicted a much lower contribution from vegetative detritus burning (1%) in Los Angeles. Observational data collected in Houston[75] indicate that biomass burning contributes 1–2% of total $PM_{2.5}$, whereas the NEI allocates 14% of the $PM_{2.5}$ mass to this source type. No estimation of residential fuel combustion or wood smoke contributions was made for the Northeastern United States in the NARSTO assessment.[58] A factor analysis-based source allocation study in Atlanta[11] estimated that wood smoke contributed 22% of $PM_{2.5}$, and a CMB-based source allocation study[15] estimated that wood combustion contributed 23% of $PM_{2.5}$. Both of these values are higher than the NEI estimate of 7% for Georgia.

Summary for Emissions from Residential Fuel Consumption. Estimation methods for residential wood combustion have received significant attention, but estimates from this source are still uncertain, because top-down methods of validation have difficulty distinguishing controlled burning in fireplaces from wildfire emissions and prescribed burning. The choice of tracer species on which to base observational estimates will also affect the accuracy of the results.

Wildfires and Prescribed Burns

Source Types. Outdoor fires include wildfires, prescribed burns, slash burns, and agricultural field burns. Dennis et al.[75] define wildfires as "unwanted and accidentally, maliciously, or naturally ignited fires that occur in wildland" and prescribed burns as "non-agricultural controlled forest/understory, grassland, and rangeland management fires, excluding slash burning." Slash burning is defined as "planned non-agricultural fires of biomass residues resulting from timber harvesting practices and land clearing operations." Agricultural field burning is defined as "agricultural land clearing burning, planting preparation burning, stubble burning, crop residue/waste burning, and burning of standing fields." Estimates of $PM_{2.5}$ in emissions from fires in the United States in 2002 range from 1.48 Tg yr^{-1} to 2.4 Tg yr^{-1}.[76]

Estimation Methods and Magnitude. From 1990 to 2000, wildfires and prescribed burns were estimated to have emitted 23% of total national primary $PM_{2.5}$ emissions, excluding fugitive dust.[18] Emissions are generally estimated from the burned surface area, the fuel consumption per unit area, and the emission factor per unit mass of fuel burned. A variety of methods can be used to evaluate each of these inputs.

Table 7. Summary of residential fuel combustion emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	11% of nondust emissions
Major source types	Wood stoves, fireplaces, and fuel oil space heaters
Bottom-up approach	Census data on wood burning in homes
Top-down approach	Molecular tracers of wood burning
Techniques used to spatially allocate emissions	Allocated using census data
Techniques used to temporally resolve emissions	Allocated based on typical use or using a correlation to monthly temperatures
Composition	43–59% OC and 1–5% EC, 0.3–0.9 potassium (often used as a tracer)
Size fraction	Mostly <2.5 μm with a number concentration peaks between 0.1 and 0.2 μm

Burned area has traditionally been estimated based on reports from fire management agencies,[75,77] but recently a variety of investigators have used satellite observations to estimate the size of fires and estimate emissions from the fires. Both methods are subject to errors and uncertainties. Satellite images can be compromised by clouds and thermal hot spots because of sources other than fires (e.g., flaring in oil and gas production). In addition, fires that occur between satellite passes may be undetected. Conventional fire incident reports may overreport or underreport the size of a fire or multiply report a single fire in multiple reporting systems. Wiedinmyer et al.[76] used the Moderate Resolution Imaging Spectroradiometer (MODIS) fire and thermal anomalies product to detect fires and estimate their size on a 1-km resolution[76,78,79] and found that MODIS predicted sizes varied by 2%, 64%, and 82% for three fires for which there were also incident reports. They conclude that fire scars from satellites may provide more accurate data on fire sizes. Mendoza et al.[80] used Advanced Very High Resolution Radiometer images to identify the location of and to estimate the size of fires in Northeastern Mexico on a resolution of several hundred square meters and found many more fires detected by satellites than reported through the incident database. In contrast, Mullet[81] reported good agreement between MODIS detected fires and fire incidents at more than 300 acres for the summer of 2000 in Texas.

Fuel loadings and emission factors used for fires are highly dependent on land cover information, which provides data on species type and biomass density. A variety of different information sources are available, as summarized by Wiedinmyer et al.,[76] and the choice of land cover dataset can lead to an uncertainty of a factor of two or more in the emission estimation.

Temporal and Spatial Resolution. Interannual variability in fire emissions, especially for wildfires, can be large. Wiedinmyer et al.[76] estimated a threefold variation in emissions

Table 8. Comparison of top-down and bottom-up residential fuel combustion emissions inventories.

Location	Inventory Estimates: National Emissions Inventory Average for 1990–2000 (% of nondust emissions)[46]	Inventory Estimates: CARB[67]	Source Apportionment Estimates (% of nondust emissions)	Reference (source apportionment)
California	18	15	18	1
San Joaquin Valley			32	1
Fresno		10		
Los Angeles		6	1–29	1, 14
Texas/Missouri	14			
Houston			1–2	75
New York/Pennsylvania/Maryland	17			
Georgia	7			
Atlanta			22–23	11, 15
National average	11			

among the years 2002, 2003, and 2004, and Dennis et al.[75] found a factor of two difference between 1996 and 1997 for large wildfire emissions in Texas; however, total outdoor fire emissions from year to year varied by less than 10%, because there was little difference in the extensive emissions associated with rangeland and crop burning. These fires, which individually may be small and undetected by satellite, can still collectively dominate emissions.

Seasonal variability in emissions from outdoor fires is also large. Wiedinmyer et al.[76] report frequent large fires in the summer in the United States and Canada and in the spring in Mexico, whereas Dennis et al.[75] report large fire emissions during winter, early summer, and fall because of agricultural burning in Texas, depending on the crop type.

Fires are generally assumed to have no day-of-week dependence. Multiday fires may vary significantly in intensity. If emissions are based on fire incident reports, a temporal distribution of emissions must be assumed. For example, Junquera et al.[77] assigned emissions uniformly over the length of the fire. Satellite data, in contrast, have the potential to provide some measure of variations in daily intensity of multiday fires.

Composition and Size Distributions. A number of investigators have reported on the composition and size distributions of wood smoke, although many of these tests have been conducted in fireplaces rather than in open-air settings. Most ambient studies report unimodal size distributions in the 10-nm to 0.5-μm size range and particles that are more than 70% carbonaceous.[82] Hays et al.[82] suggest molecular markers to distinguish conifer wood smoke from other sources. Other markers may be possible. Particle number distributions vary with burning conditions (ignition, flaming, and smoldering) and with the fuel type.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. Relatively few comprehensive annual or seasonal inventories of fires have been compiled, limiting the ability to compare observational measurements of molecular markers for fires with emissions inventories. Junquera et al.[77] compared CO concentrations in individual fire plumes, measured by aircraft, to predicted fire emissions and found that the measurements were within the error bounds (plus or minus a factor of two) of the emission estimates. Hudson et al.[83] used aircraft measurements to assess the contributions of biomass burning to total PM concentrations during a North American field campaign and estimated that approximately a third of the PM transported from Asia to North America was attributable to biomass burning. This will be important to note future comparisons of bottom-up and top-down fire emissions estimates, because intercontinental transport is a source that significantly affects ambient concentrations but is not captured in current emissions inventories.

Summary for Emissions from Outdoor Fires. Both activity-based emissions inventories and source resolution studies suggest that biomass combustion is a significant source of $PM_{2.5}$ in North America. The emissions inventory data are summarized in Table 9. Assessments of the accuracy of inventories of fire emissions are limited, because $PM_{2.5}$ emissions from residential combustion, wildfires, prescribed burns, agricultural burning, and other fire types possess few unique tracer species. Uncertainties in emissions inventories of this source type are compounded by variability in fire frequency, spatial location and area, and intensity. Satellite data provide much potential for providing the continual updates to fire activity data that will be necessary for improving emission estimates from large outdoor fires, but the effect of small, frequent fires that are not detectable in current satellite fire products should not be ignored.

Fugitive Dust

Source Types. Fugitive dust comes from many sources, including paved roads, unpaved roads, construction sites, aggregate handling and storage piles, industrial wind erosion, abrasive blasting, and agricultural sites. According to the NEI, for the years 1990–2000, 21% of fugitive dust originated from paved roads, 52% originated from unpaved roads, 23% originated from construction sites, and 4% originated from other sources.[18]

Table 9. Summary of emissions inventories for wildfires and prescribed burns.

Magnitude as predicted by the NEI (1990–2000)	23% of nondust emissions
Major source types	Wildfires, prescribed burns, slash burns, and agricultural field burns
Bottom-up approach	Estimates of acreage burned, fuel loading per acre, and emissions per t of fuel burned
Top-down approach	Molecular markers of cellulose combustion
Techniques used to spatially allocate emissions	Wildfire locations recorded as point sources; agricultural burning allocated based on land use
Techniques used to temporally resolve emissions	Wildfire dates recorded; agricultural burning allocated based on typical agricultural practices
Composition	>70% carbonaceous
Size fraction	Unimodal size distributions in the 10-nm to 0.5-μm size range

Estimation Method and Magnitude. The NEI predicts that 41% of all $PM_{2.5}$ emissions came from fugitive dust sources between 1990 and 2000.[18] Road dust emission factors are based on an EPA estimation procedure that takes into account particle size fractions, the magnitude of silt loading on roadways, the average vehicle speed, and the average vehicle weight traveling on a roadway. The emission factor formulas that take these variables into account are different for paved and unpaved roads. The paved-road formula was developed from a linear regression of 65 road tests.[5] Default silt-loading levels are based solely on road type, not location. Silt loading and road dust levels vary geographically, so these default values may not represent actual conditions in many areas. For example, Etyemezian et al.[84] found that, for Idaho roads, EPA's default silt loading values were low by a factor of 1.5–3.8. In addition, Carvacho et al.[85] found that the clay content of a soil affects its likelihood of entrainment. Gillies et al.[86] found that unpaved road dust emissions vary linearly ($R^2 = 0.95$) with vehicle speed and vehicle weight, contrary to EPA emission factor, which assumes that emissions vary with vehicle weight to a power of 0.45 or 0.7, depending on the silt loading of the road. In addition, EPA equations predict that emissions from roads dominated by light-duty vehicles are proportional to the square root of vehicle speed. Again, measurements by Gillies et al.[86] predict a linear relationship ($R^2 = 0.77$) between speed and emissions.

Dust emissions from construction activities vary by the types of activities taking place. However, EPA uses a single emission factor of 1.2 t/acre/month to predict average construction site PM_{10} emissions. This emission factor was developed from a single study of apartment and shopping area construction sites. For more accurate estimates of particular activities, AP-42 recommends using emission factors developed for surface coal mining activities, including drilling and blasting, loading and unloading aggregate material and debris into trucks, construction equipment transport, bulldozing, scraping, land clearing, and material handling.[5] One study, which developed a $PM_{2.5}$ emission factor for dirt carryout from a construction site in Kansas City, MO (0.2 g/vehicle), has not yet been incorporated into the AP-42 emission factors.[87] The current emission factors come from a limited number of studies, and there are not yet enough data to create factors that vary based on climate, intensity of construction activity, type of construction activity, or silt content of the site.

Temporal and Spatial Resolution. $PM_{2.5}$ emissions are typically spatially and temporally resolved using the allocations for on-road emissions (for road dust) and nonroad emissions (for construction dust). However, this approach does allow for differentiation between traffic on paved and unpaved roads.

Morey et al.[88] attempted to spatially allocate unpaved road dust emissions in the San Joaquin Valley by overlaying road classification data obtained from the California Department of Transportation onto a land-use geographic information system map of the area. By assuming that all roads in urban areas were paved, they were able to estimate the fraction of roads that were unpaved by land-use category. This approach to spatially distributing unpaved and paved road emissions is rigorous but still involves multiple assumptions, is labor intensive, and is applicable only to specific areas.

Temporal allocations can also be challenging to perform. In urban areas, where unpaved roads are less common, it may be reasonable to allocate road dust emissions using the same temporal allocations given to tailpipe emissions. This assertion is supported by a study performed by Kidwell and Ondov,[89] who used factor analysis to show that road dust emissions correlated with times of heavy traffic in College Park, MD. The temporal allocation of traffic and road dust emissions on unpaved roads is more complicated. Morey at al[88] documented harvest activity by crop and nonharvest activity (i.e., road traffic) by road type in the San Joaquin Valley to allocate fugitive dust emissions on a monthly basis. Because nonharvest activity was relatively constant throughout the year, activity factors for harvest activities could be determined for those months during which the particular crops were harvested. Overall, in San Joaquin County and Fresno County, VMTs on unpaved roads peaked during July, August, and September and were lowest during January, February, and March. This type of analysis is useful but very labor intensive and again may not be practical for developing an NEI.

Composition. The chemical composition of fugitive dust sources is expected to be similar in regions with consistent exposed soil composition. Samples of cement, gravel, and dust collected from paved and unpaved roads, agricultural fields, a dry lake bed, a landfill, an asphalt plant, and a parking garage in Mexico City, Mexico, revealed similar high concentrations of silica, iron, and calcium.[90] Aluminum concentrations were high in all of the samples except the cement and gravel, and high OC concentrations were observed in the asphalt, cement, and gravel samples. A study conducted in Hong Kong using samples of parkland soil, urban soil, paved road dust, cement, and aggregate[91] revealed similar results, leading the authors to conclude that Si, Al, K, Ca, Ti, and Fe are good markers for fugitive emissions from soil and road dust. Rogge et al.[92] investigated the chemical speciation of dust from cattle feedlots and found that unique organic compounds released by the cow's digestive system, for example, 5β-stigmastanol, epi-5β-stigmastanol, and C_{18} fatty acids, were also present in the soil and could be used as unique tracers for source apportionment studies.

Size Distributions. Size distributions vary depending on the source of fugitive dust. A study of fugitive dust in Mexico City reports that $PM_{2.5}$ makes up between 20% and 26% of total PM from paved road, agricultural, landfill, and asphalt fugitive dust sources. For unpaved road dust, this percentage dropped to 13%, and for dust from dried lake bed sediments and cement, the percentages were 31% and 34%, respectively.[90] Because of the differences in size distributions, the emissions from these various fugitive dust sources may have different residence times in the atmosphere. Etyemezian et al.[93] report that, for dust emitted from a rural road, there was no measurable deposition of PM_{10} 100 m downwind of the road, but 25% of larger particles (19.7 μm) were removed over the same distance.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. Fugitive dust emissions of $PM_{2.5}$, reported through the NEI for the period 1990–1999, for the regions of the United States in which EPA Supersites were located, indicate that dust sources contributed 46% of total $PM_{2.5}$ emissions in California; 63% in Texas and Missouri; 46% in New York, Pennsylvania, and Maryland; and 39% in Georgia.[46] These source strengths and the emissions inventory information described above are summarized in Table 10.

Malm et al.[94] used ambient observations of $PM_{2.5}$ composition measured at 143 sites in the Interagency Monitoring of Protected Visual Environments program to estimate soil contributions at each site. The highest contributions (1–3.1 μg/m^3) were observed at sites in the Southwestern United States; sites in the Northeastern and Northwestern United States had contributions of less than 0.5 μg/m^3, whereas most of the rest of the United States had intermediate values. Urban sites contained higher proportions of fine soil than surrounding rural sites, suggesting resuspension from heavy road traffic in those areas. Contributions of fugitive dust were low during the winter, with sudden increases in April. For most locations, this springtime peak was attributed to long-range transport from Africa and Asia, not to changes in local emissions patterns.

Comparisons of observational data with the NEI reveal some discrepancies in the inventory. For example, a receptor model study of the Denver area revealed source contributions of fugitive dust that were half of that reported in the NEI.[1] Fraser et al.[63] used CMB methods to determine that road dust in the Houston area contributed less than half the strength of on-road sources. The NEI for Texas predicts dust contributions of up to an order of magnitude greater than the other on-road emission sources. A summary of these comparisons is provided in Table 11.

Table 10. Summary of fugitive dust emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	41% of total $PM_{2.5}$ emissions
Major source types	Paved roads, unpaved roads, construction sites, aggregate handling and storage piles, industrial wind erosion, abrasive blasting, and agricultural sites
Bottom-up approach	Emission factors based on road conditions and vehicle speed
Top-down approach	Use of silica and alumina tracers
Techniques used to spatially allocate emissions	Spatially resolved using the allocations for on-road emissions (for road dust) and nonroad emissions (for construction dust)
Techniques used to temporally resolve emissions	Temporally resolved using the allocations for on-road emissions (for road dust) and nonroad emissions (for construction dust)
Composition	Similar to consistent exposed soil composition (varies by region)
Size fraction	Mostly coarse PM

Countess et al.[95] explain these discrepancies by the fact that the emissions have a low release height and no upward momentum and, consequently, redeposit very close to the emission point. Research to investigate this discrepancy and its justification has yielded inconsistent results. Etyemezian et al.[93] measured particle flux in West Texas next to a road in the middle of rangeland where the surface roughness was very small. Veranth et al.[96] placed cargo containers in the path of the fugitive dust plume to simulate the surface roughness in a residential setting. Etyemezian et al.[93] found that less than 9.5% (with 95% confidence) of the emitted PM_{10} was deposited within 100 m, and they corroborated their results with a Gaussian model. Veranth et al.,[96] on the other hand, found that within 95 m of the road, between 86% and 89% of the PM_{10} emitted was deposited. They found that a Gaussian model significantly underpredicted the percentage of mass deposited (30%). This underprediction is likely because of the fact that the surface roughness height is similar to the plume height. In addition, the large roughness height means that there will be steep wind velocity gradients at the plume height. The Gaussian model does not account for either of these two conditions. These two studies show that the potential for deposition near the roadway is highly dependent on the geometry of the surroundings. In addition, it should be noted that factors other than surface roughness will affect this near-source deposition. Ground cover, wind turbulence, and particle size distributions also must be properly characterized before proper predictions of deposition can be made.

Although near-source deposition may be a significant process, it is important to note that other factors may also be influencing the discrepancies noted here. Factors that may not be properly accounted for when making bottom-up predictions of emissions include wind speed, soil type, soil moisture, ground cover, and the type of activity causing the emissions. In addition, ambient concentrations will likely be affected by wind direction, sampling

Table 11. Comparison of top-down and bottom-up fugitive dust emissions inventories.

Location	Inventory Estimates: National Emissions Inventory Average for 1990–2000 (% of total $PM_{2.5}$ emissions)[46]	Bottom-Up Inventory Overestimation as Compared with Source Apportionment Estimates	Reference (comparison of source apportionment to bottom-up inventory)
California	46		
Texas/Missouri	63		
Houston		More than an order of magnitude	63
New York/Pennsylvania/Maryland	46		
Georgia	39		
Denver		Factor of two	1
National average	41		

location in relation to the source, wind speed, and turbulence. A full review of all of these factors will likely be necessary to pinpoint the cause of the discrepancies between ambient and predicted dust concentrations.

Summary for Emissions from Fugitive Dust Sources. Bottom-up emissions inventory techniques generally do a poor job of predicting ambient dust concentrations. Several areas of uncertainty have been identified here. First, emission factors fail to properly account for variations in road dust emissions because of vehicle weight and speed. In addition, silt loading content is not well characterized in many locations, and default values are likely to be inaccurate for large areas of the country. In addition, dust emission factors for construction sites do not properly account for silt loading, soil moisture, or activity type and intensity. Finally, other processes may affect overall ambient concentrations after emissions take place. One theory proposes that near-source deposition is significant and has not been accounted for. Research of this hypothesis has had mixed results. Fugitive dust makes up approximately half of predicted $PM_{2.5}$ emissions in the NEI and, thus, it is paramount that this source be accurately characterized. A significant amount work will be required to fully determine the source of the large discrepancies between top-down and bottom-up predictions of $PM_{2.5}$ fugitive dust emissions.

Point Sources

Source Types. According to EPA's NEI, between 1990 and 2000 the source types that contributed most to overall point source primary PM emissions were electric utility fuel combustion (20% of point source PM_{10} and 17% of point source $PM_{2.5}$), industrial fuel combustion (19% of PM_{10} and 22% of $PM_{2.5}$), metals processing (14% of PM_{10} and 17% of $PM_{2.5}$), and the mineral products industry (19% of PM_{10} and 15% of $PM_{2.5}$). Coal combustion contributed 80–90% of electric utility fuel combustion emissions. Industrial fuel combustion was split among coal combustion, wood waste combustion, and oil and gas combustion. Other sources that contributed less than 10% of national point source PM emissions included chemical manufacturing plants and petroleum refineries (not including fuel combustion), agricultural sources, wood pulp and paper manufacturing plants, and storage and transport facilities.

Estimation Methods and Magnitude. During 1990–2000, point sources were estimated to have emitted 18% of total national primary $PM_{2.5}$ emissions, excluding fugitive dust. Emissions from point sources are typically calculated using AP-42 factors. AP-42 has separate sections for coal, oil, gas, and wood combustion. EPA uses a lettered grading system from "A" (excellent) to "E" (poor) to identify the reliability of emission factors. An "A" rating typically is applied to emission factors based on high-quality source tests taken from a statistical sampling of facilities. In contrast, an "E" rating is typically based on a small number of source tests that may have significant uncertainties. Coal emission factors range from "A" to "E" with substantially more "E" ratings than any other rating. Oil combustion emissions and metals processing facilities received ratings of "A" or "B." The remaining industries received lower ratings: wood processing industries rated "D," cement facilities rated "C" through "E," and foundries rated "E." This heterogeneity in the reliability of point source emission factors suggests that there will be significant uncertainty in estimates of point source emissions.

Temporal and Spatial Resolution. Spatial resolution for point sources is fairly straightforward, because NEI entries include latitude and longitude locations for each point source. Temporal resolution of emissions can be more difficult. The NEI is reported as an annual average. Default temporal allocations often assume constant operation. More precise temporal allocations require knowledge of the operating schedule and throughput of specific point sources and, thus, are not often performed for every point source.

Composition. EPA publishes split factors that indicate average compositional breakdown of $PM_{2.5}$ for various sources.[97] The $PM_{2.5}$ is speciated into OC, EC, sulfates, nitrates, and other species emitted directly from the point source. Because of relatively limited data emerging from field studies on the relative strengths of specific industrial sources, as described below, it is beyond the scope of this review to evaluate PM composition data for all of the industrial sectors in point source PM emissions inventories. However, the data for coal combustion are briefly reviewed because of the significance of this source nationally.

Coal combustion $PM_{2.5}$ emissions can vary in chemical composition depending on the composition of the coal. EPA assumes a composition that is 1% OC, 1% EC, 12% sulfates, and 85% other material.[98] Recent findings[98] suggest that this source may be more accurately described as 20% OC, 1% EC, 16% sulfate, 0.5% nitrate, and 62.5% other. The "other" portion of the emission is determined based on the ash content and trace element composition (antimony, arsenic, beryllium, cadmium, chromium, cobalt, lead, manganese, and nickel) of the coal. Accurate description of the "other" composition requires knowledge of the ash content and the trace element content of the coal, even if emission factors receive a rating of "A" by EPA. Some of the changes in the split factors may be because of recent work that more completely accounts for the condensable emissions from coal combustion. AP-42 also contains emission factors that do not require inputs of ash fraction and trace metal concentration in the coal, but these factors have received an "E" rating. In the event that the emission is controlled, AP-42 emission factors for trace metals are available that do not require knowledge of the coal characteristics. Factors for emissions from facilities that use venturi scrubbers, spray dry absorbers, or wet limestone scrubbers with fabric filters or electrostatic precipitators all received ratings of "A," except for the factor for chromium VI, which received a rating of "D."

Size Distributions. AP-42 also provides information that can be used to estimate the fractional particle size distribution of combustion PM emissions from a number of different fuel types with a number of different controls. All of the size-resolved emission factors have ratings of "C" or lower. In general, uncontrolled coal combustion creates particles, more than 50% of which (mass basis) are more than 10 μm (in aerodynamic diameter) and less than 10% of which are less than 1 μm. A summary of other size distributions is presented in Table 12.

Control devices shift the distribution toward smaller particles, although the specific amount of this shift varies by type of control device. Electrostatic precipitators are predicted to increase the percentage of large particles, whereas scrubbers and cyclones are predicted to decrease the percentage of large particles.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations. From 1990 to 1999, $PM_{2.5}$ point source emissions averaged 10% of $PM_{2.5}$ emissions in California; 19% in Texas and Missouri; 37% in New York, Pennsylvania, and Maryland; and 17% in Georgia.[46] This regional variability in total point source emissions is even more pronounced when individual industrial categories are considered. For instance, the Gulf Coast from Houston to Louisiana houses much of the petroleum refining capacity in the United States; the emissions from industrial fuel combustion, as well as other petroleum and synthetic organic chemical manufacturing processes, are concentrated in this area of the country. Analogously, the Ohio River Valley experiences concentrated emissions from coal-fired power plants. These source strengths and the emissions inventory information described above are summarized in Table 13.

Table 12. Summary of PM size distribution of uncontrolled PM emitted during fuel combustion.

Fuel Type	Fraction <1 μm (g/g) (%)	Fraction >10 μm (g/g) (%)	AP-42 Rating
Coal	≤10	>50	C or lower
Fuel oil	14–39	14–38	C or lower
Natural gas	~100	0	C or lower
Wood/bark	67	10	E

It is difficult to evaluate the contribution of source apportionment studies to refine point source emissions inventories, because very few of the studies specifically address contributions of point sources. Rather, point source contributions are often embedded in the "other" or "unknown" fraction, because source profiles specific to the point sources in that region are not available. The source apportionment study for the San Joaquin Valley[58] attributed 43% of $PM_{2.5}$ to "unknown" sources. Various studies of the Los Angeles area have attributed 17%[58] or 37–57%[13] of $PM_{2.5}$ to "other sources" and 12–13% of $PM_{2.5}$ to "background" sources.[58] All of these estimates are consistent with the NEI, because they are greater than the NEI of point sources. A CMB study of Atlanta[15] $PM_{2.5}$ attributed 1% of the ambient $PM_{2.5}$ to point and area sources that use natural gas combustion, 12% to "other organic" sources, and 8% to "other" sources. A factor analysis study[11] of the same area attributed 10% of the $PM_{2.5}$ to metal processing facilities and another 6% to a cement kiln and another carbon-rich source. These estimates are close to the fraction of $PM_{2.5}$ emissions attributed to point sources in the NEI for Georgia (17%, excluding fugitive dust). A summary of these top-down and bottom-up comparisons is provided in Table 14.

PM PRECURSOR EMISSIONS

Source Types

A large portion of atmospheric PM is formed by atmospheric processing of organic and inorganic precursor species. Organic precursors include HCs emitted from anthropogenic sources (e.g., motor vehicles and power plants) and biogenic sources (e.g., trees and other vegetation) that can be oxidized into low-volatility products that partition into PM. Inorganic precursors include NO_x,

Table 13. Summary of point source emissions inventories.

Magnitude as predicted by the NEI (1990–2000)	18% of nondust sources
Major source types	Electric utility fuel combustion, industrial fuel combustion, metals processing, and the mineral products industry
Bottom-up approach	Data on individual point sources for PM_{10}
Top-down approach	Spatial distributions of PM mass
Techniques used to spatially allocate emissions	Not applicable: major point sources are at known locations
Techniques used to temporally resolve emissions	Some major point sources have continuous emissions monitoring
Composition	Varies greatly between source types
Size fraction	Varies greatly between source types

Table 14. Comparison of top-down and bottom-up point source emissions inventories.

Location	Inventory Estimates National Emissions Inventory Average for 1990–2000 (% of nondust emissions)[46]	Upper Bound of Source Apportionment Estimates (includes "other" and "unknown" sources) (% of nondust emissions)	Reference (source apportionment)
California	10		
San Joaquin Valley		43	58
Los Angeles		17–57	13, 58
Texas/Missouri	19		
New York/Pennsylvania/Maryland	37		
Georgia	17		
Atlanta		16–21	11, 15
National average	18		

SO_2, and NH_3 and can form particle-phase ammonium sulfate and ammonium nitrate. Although relatively little uncertainty exists in the NO_x and SO_2 emissions inventories, significant uncertainties still exist in the HC and ammonia emissions inventories. Because the HC inventory uncertainties are varied enough to merit a separate review, this review focuses on the current state of the ammonia inventory.

Major sources of ammonia include livestock, forests, and other biogenic sources; agricultural production; motor vehicles; and wastewater treatment plants. The NEI reports that from 1990 to 2001, livestock accounted for 74% of total national ammonia emissions. Other major contributors included agricultural crops (13%), on-road vehicles (5%), agricultural chemical manufacturing (3%), and waste disposal and recycling (2%).[18] A comprehensive national inventory has also been prepared by Carnegie Mellon University (CMU),[99] which attributes the bulk of ammonia emissions to animal husbandry and fertilizer application (85%).[100]

The relative importance of animal husbandry and fertilization emissions will vary by state based on the intensity of these activities. However, few state-specific emissions inventories of ammonia have been compiled. A recent ammonia inventory for the state of Texas reports 80% of emissions from livestock, 8% of emissions from fertilizer and other agricultural production, 5% from scrubland and grassland, 2% from on-road vehicles, 2% from other consumer and commercial sources, 1% from forests, and 1% from wastewater treatment facilities.[101,102] Because this inventory used some different emission factors than those used in the NEI, comparisons between the two inventories can be used to characterize uncertainties in emission estimates.

Emission Estimation Methods and Magnitude of Emissions

Ammonia emissions from livestock are usually determined using animal-specific emission factors. These emission factors have units of kilograms of NH_3 per animal per year. Values reported for the state of Texas[101,102] and in the CMU model[99] are generally comparable, although some of the factors are identical, and others vary by as much as 100%. Studies have shown that ammonia emission rates from animal feeding operations are also affected by climate, manure storage practices, land application method of manure, and animal age,[103] although the quality of the studies that inform these dependencies are variable. Some of the studies obtained values from data collected over several months, whereas others used data that were collected over the period of a single day. Emission factors are used to estimate emissions from animal feeding operations in most ammonia emissions inventories. An alternative approach performs a nitrogen mass balance on the entire animal feeding operation,[103,104] thereby accounting for interactions between different systems in the animal feeding operation that cannot be accounted for using single emission factors. This approach has yet to be implemented on a large scale, however, and not all of the data needed for this approach are commonly available.[103,104]

Fertilizer emissions are estimated by multiplying fertilizer application rates by the mass fraction of the fertilizer that is volatile. Data on fertilizer application rates in the CMU inventory are based on data from the U.S. Geological Survey and the Association of American Plant Food Control Officials,[105] whereas the data on fertilizer application rates in the Texas inventory are from the Association of American Plant Food Control Officials and the University of Kentucky.[106] Most of the data are within 20% in the two compilations.

Ammonia emissions from on- and off-road vehicles are typically calculated using EPA's MOBILE and NONROAD models described in previous sections. Finally, ammonia emissions from trees, shrubs, and grasses are determined using land cover maps, emission factors for different plant species, foliage density, temperature, and solar radiation. Emission factors for vegetation types reported by Corsi et al.[101,102] and the CMU model in kilograms of NH_3 per meter squared per year are not consistent: values used for scrub and grassland vary by a factor of four, and values used for oak and pine forests differ by two orders of magnitude. This has the potential to cause large discrepancies in the emissions inventory. Corsi et al.[101,102] explain the uncertainty in ammonia emissions in forests by the lack of measurements at the forest canopy and the significant variability in biochemical processes and environmental conditions that effect the factors.[101,102] As an example, the emission factor for ammonia emissions from a forest vary from $1.36–1.77 \times 10^{-2}$

kg/m^2/yr[107] for forests in South Korea to 3.65×10^{-4} extrapolated from measurements of bare soil[108] to 0.1–7 $\times 10^{-3}$ for fertilized forests,[109–111] to 3.5×10^{-6} for unfertilized forests[112] in the United States.

Spatial and Temporal Distribution

The CMU model estimates ammonia emissions on a county basis. More resolved emissions are possible if land use is allocated at a finer resolution, where livestock, fertilizer, and vegetation emissions are based on the land cover type, and mobile source emissions are allocated to urbanized areas. Because the majority of ammonia emissions come from livestock and fertilizer applications, ammonia emissions will be greatest in agricultural areas.

Ammonia emissions vary temporally[113] and, on average, are greatest in the spring months (March to June) and lowest during the winter. There is also a smaller peak in ammonia emissions in the early fall. Emissions from nonroad engines (agricultural equipment, lawn and garden equipment, etc.) are highest in the summer months and lowest during the winter, with temporal allocations factors that are more than twice as high in June, July, and August compared with January.[99] Emissions from farm animals are also highest in the summer and lowest in the winter with the summer temporal allocation factor being approximately 2.5 times the winter temporal allocation factor. Finally, fertilizer emissions have a dramatic peak in the spring with a smaller peak in the early fall months. On a diurnal basis, biogenic and agricultural emissions are highest during the hours with the greatest sunlight, nonroad equipment emissions are greatest during working hours (9:00 a.m. to 5:00 p.m.), and on-road vehicle emissions peak during morning and evening rush hours.

Regional Comparisons of Emission Estimates with Observed Ambient Concentrations

Comparisons between ammonia emissions inventories and observational data are complicated by the atmospheric partitioning of ammonia between the gas phase (as ammonia) and the particulate phase (as ammonium). To reconcile this issue, ammonia emissions can be compared with observations of total ammonia (ammonia plus ammonium); however, measurements of gas-phase ammonia are relatively sparse. The most comprehensive comparison of ammonia emissions with ambient observations has been reported by Gilliland et al.,[100] who compared observational data with predicted total ammonia concentrations obtained by inputting refined NEI emission estimates into a regional photochemical model. The analysis indicated that the NEI was reasonably accurate; however, uncertainty in the emissions inventory could not be quantitatively estimated because of issues with the observational data.

Only limited studies of ammonia and ammonium concentrations in individual regions are available. Pavlovic et al.[114] compared observed ammonia and ammonium concentrations at an urban/industrial Houston site to predictions from a photochemical model and found agreement that was within a factor of two.

SUMMARY AND RECOMMENDATIONS FOR POLICY-MAKERS

As documented in the previous sections, substantial uncertainty (more than a factor of two) exists in most segments of $PM_{2.5}$ emissions inventories and in the emissions inventories of certain PM precursors; these uncertainties can pose challenges in assessing the effectiveness of proposed PM control strategies. Greater confidence in emissions inventories can emerge from iterative emission estimates (bottom-up methods) and comparisons with ambient observational data (top-down methods). EPA Fine PM Supersites provide examples of these coordinated assessments, as described in the case studies below.

Houston, TX

Using data from the Houston Supersite, Allen and Fraser[115] performed both bottom-up and top-down assessments of emission source strengths. The key findings emerging from these analyses are described below.

- Fires are a sporadic but significant source of $PM_{2.5}$ emissions in Texas. Estimates based on measurements of levoglucosan (a molecular marker of wood smoke) and on estimates of fire acreage and land cover burned are consistent in indicating that wildfires and controlled burns of biomass contribute of order 1–2% of the annual average total $PM_{2.5}$ mass in the Houston-Galveston area.[75,77,116]
- Geological sources of $PM_{2.5}$ (dust) are a relatively minor contributor to total $PM_{2.5}$ mass in the urban Houston-Galveston, TX, area. Buzcu et al.[116] and Yue and Fraser[117] have estimated that road dust accounted for an average of 0.3 $\mu g/m^3$ of $PM_{2.5}$ mass (2% of total $PM_{2.5}$ mass) in August and September 2000, at a ship channel area site (Houston Regional Monitoring Site No. 3). Other sites had much lower contributions from dust.
- Primary mobile source emissions are significant; these emissions account for approximately a quarter to a third of $PM_{2.5}$ mass. Mobile source emissions contribute 3–6 $\mu g/m^3$ to seasonal average PM concentrations, depending on the site. Of these emissions, approximately half are because of diesel emissions and half are because of emissions from gasoline engines with catalytic converters, although there is some variation from site to site.
- Primary emissions from cooking are significant; these emissions account for ~10–15% of $PM_{2.5}$ mass. Concentrations of molecular markers in $PM_{2.5}$ in Southeast Texas indicate that 1.4–2.6 $\mu g/m^3$ of $PM_{2.5}$ observed in Southeast Texas can be attributed to emissions from cooking. The concentrations are highest in residential areas and lowest in industrialized areas. The $PM_{2.5}$ concentrations attributed to cooking represent 10–20% of the observed, annual average $PM_{2.5}$ concentrations, making cooking a significant source.

Atlanta, GA

Source apportionment studies performed in Atlanta have used both CMB and positive matrix factorization (PMF) techniques. The comparison of the results obtained through these two techniques with bottom-up emissions inventory data gives some indication of the magnitude of uncertainties in quantifying emissions in Atlanta. The 1996–2001 NEI data for the 28-county Atlanta metro area was queried for comparison.[46] Some key findings are outlined below.

- Although the NEI predicts that fugitive dust makes up a large potion of $PM_{2.5}$ emissions (30% for Fulton County and 45% for the 28 county Atlanta Metro area),[46] ambient measurements show that fugitive dust makes up only between 3% and 15% of total primary $PM_{2.5}$ in Atlanta.[11,15,118–122]
- On-road and nonroad mobile source emissions are major contributors to $PM_{2.5}$ mass. Bottom-up emissions inventories predict that they account for between 7% and 16% of total $PM_{2.5}$ emissions including dust (two thirds to three quarters of which come from diesel engines),[46,119] whereas source apportionment studies show mobile sources contributing 31% to 74% of total primary $PM_{2.5}$ emissions, with diesel emissions making up between 50% and 97% of mobile source emissions.[11,15,118–122]
- Emissions inventories predict that residential fuel combustion contributes 3–6% of total primary $PM_{2.5}$ emissions, including dust, depending on the location.[46,119] Ambient measurements show that 8–46% of primary $PM_{2.5}$ mass can be apportioned to wood smoke sources.[11,15,118–122] Wood smoke measurements vary dramatically with time of year. Zheng et al.[15] took measurements throughout the year and found that wood smoke made up only 15% of measured fine primary aerosol during the summer (July and August) and 46% of fine primary aerosol during the winter (January), with fall and spring values being intermediate.
- Both bottom-up emissions inventories[119] and ambient measurement studies[15,119] show that cooking contributes to approximately 5% of total primary $PM_{2.5}$ emissions during most of the year. Zheng et al.,[15] however, do report a higher contribution (15%) during the summer.
- Other local sources, including cement kilns, asphalt manufacturing, and metal processing facilities generally contribute approximately 10% or less of total primary fine aerosol emissions in Atlanta.[11,15,118–122]
- All of the top-down source apportionment assessments used data from a single monitoring site (the Atlanta Supersite). These studies are fairly consistent in their assessments of which sources are most significant. However, there is a large amount of variability in the predicted magnitudes of specific sources.

Washington, DC, and Baltimore, MD, Metro Area

NEI data from 1996–2001 were compiled for the Baltimore and Washington metro area to determine source contributions.[46] Most of the source apportionment studies that have been performed for the Baltimore and Washington metro area used the PMF technique.[123–127] One study did perform a CMB analysis for the Baltimore area,[128] but this study only apportioned metal PM and, thus, cannot be compared with the emissions inventories or PMF studies that characterize source contributions to total PM. The PMF studies that were performed used a variety of techniques and data. Early studies only used two OC fractions.[123,124] Later studies used additional data in the PMF analysis, including eight temperature-resolved OC fractions,[125,126] as well as wind speed and daily and seasonal variations.[126] A summary of the results of these PMF studies and emissions inventory data from the NEI[46] follows.

- The NEI predicts that 33% of total primary $PM_{2.5}$ emissions for the Baltimore and Washington metro area come from fugitive dust.[46] PMF studies show that between 6% and 20% of primary $PM_{2.5}$ consists of fugitive dust.[124–126] One PMF study was able to differentiate between road dust and soil dust.[126] One technique predicted that 15% of fugitive dust is soil dust, whereas the other technique predicted that 56% of fugitive dust is soil dust. Several studies have used back trajectories to suggest that the soil dust portion of fugitive dust does not come from local sources but instead originates from Asian and Saharan dust storms.[125,126]
- The NEI for the Baltimore/Washington metro area predicts that 16% of primary $PM_{2.5}$ originates from industrial and electric utility fuel combustion.[46] Most of the PMF studies identified a heavy oil combustion factor, which is believed to be from industrial and electric utility facilities.[124–127] This factor was calculated to contribute between 3% and 14% of total primary $PM_{2.5}$ emissions. One study, which performed seasonal analysis, showed a 78% increase in these emissions in the winter versus the summer.[124]
- The NEI predicts that 14% of primary $PM_{2.5}$ originates from incineration and open burning of waste for the Baltimore/Washington metro area.[46] Four PMF studies identified an incinerator source that contributed between 10% and 23% of total primary fine aerosol emissions to the region.[124–127] One study, which included data from an extended period (1988–1997), noted that incinerator emissions decreased with time corresponding with incinerator closings in the area and also noted that incinerator emissions were higher in the winter.[125]
- The NEI predicts that metal processing facility emissions should account for just 2% of the primary $PM_{2.5}$ emissions in the Baltimore/Washington metro area but 11% of those same emissions in the city of Baltimore.[46] One source apportionment study using data from downtown Baltimore

shows that a steel plant emits 26% of total primary $PM_{2.5}$.[127]

- Residential fuel combustion should contribute 13% of total primary fine aerosol emissions in the Baltimore/Washington area according to bottom-up techniques.[46] Two PMF studies found vegetative burning or wood smoke to be a major source of $PM_{2.5}$. They predicted that this source should account for 5% and 36% of total emissions.[123,126]
- Mobile sources are a major source of PM emissions, but the bottom-up and top-down techniques do not show agreement on the magnitude of this source. Bottom-up methods indicate that mobile sources account for 12% of total primary $PM_{2.5}$ emissions (31% gasoline and 69% diesel).[46] PMF studies estimate mobile source contributions to be between 38% and 71%.[123–127] The source apportionment studies, which were able to differentiate between gasoline and diesel vehicles, showed that between 79% and 98% of mobile source emissions came from gasoline combustion.[125–127]
- The NEI does not include sea salt. However, all but one of the source apportionment studies reported sea salt as a measurable source of emissions in this area. They predict that sea salt should account for between 6% and 15% of total primary $PM_{2.5}$ emissions.[124–127]

Fresno, CA

NEI data from 1996 to 2001 were compiled for Fresno County[46] and compared with source apportionment studies performed in Fresno. In contrast to the studies done in the Baltimore/Washington area, the Fresno studies have primarily relied on CMB analyses. One study uses data from as far back as 1988–1989.[129] The other studies use a dataset that was collected in 1995 and 1996.[14,130,131] Studies determining overall source contributions to primary $PM_{2.5}$ in Fresno using the more recent supersite data have not yet been published. All of the data reported in this section are taken from monitoring sites within the city of Fresno. A summary of the currently published findings follows.

- Three NEI source categories that are major contributors to primary $PM_{2.5}$ in Fresno County would be measured as wood smoke and vegetative burning. These three categories are residential fuel combustion, agriculture and forestry (including prescribed burns), and other combustion. The NEI reports that these categories contribute 5%, 23%, and 20% (48% combined) to primary $PM_{2.5}$ emissions.[46] CMB studies predict that wood smoke and vegetative burning contribute between 36% and 65% of these emissions.[14,129–131] The studies reporting the highest contributions for these sources took place during the winter months (December and January) when residential fuel combustion is likely to be highest.[130] During the rest of the year, vegetative burning emissions may be dominated by agricultural burning instead of residential fuel combustion.
- The NEI reports that mobile source emissions accounted for 8% of total primary $PM_{2.5}$ emissions in Fresno County between 1996 and 2001.[46] The CMB studies show the mobile source contribution ranging from 12% to 50%.[129–131] Two studies differentiated gasoline and diesel emissions. One study predicts that gasoline combustion made up 42% of total mobile source emissions,[131] whereas the other predicts that gasoline combustion made up 18% (January 1996) to 23% (December 1995) of total mobile source $PM_{2.5}$ emissions.[14] The relatively high contribution from diesel engines may be because of the frequent use of farm equipment and trucks in this agricultural area.
- The 1996–2001 NEI did not include meat cooking. One CMB study included meat cooking in its source profiles and predicted that this source contributed 13% of total primary $PM_{2.5}$ emissions.[131]
- The NEI reports that 32% of $PM_{2.5}$ emissions in Fresno County were from fugitive dust.[46] One study showed very low dust contributions, below 1%.[130] The other studies that included this profile predict that dust makes up between 3% and 13% of total primary $PM_{2.5}$ emission.[14,129,131]

Los Angeles, CA

There have been several source apportionment studies in Los Angeles. Most of the data used was collected before the supersites program. Three CMB studies use data from 1982,[132] 1993,[13] and 1996.[131] These studies all took measurements at four locations, one coastal, one downtown Los Angeles, and two downwind of downtown. These measurements allow some evaluation of how sources change in different portions of the metro area. There has been one PMF study that used data collected between 1988 and 2003.[133] This study took measurements in the San Gorgonio wilderness, which is downwind of Los Angeles. This measurement location allows for some analysis of Los Angeles sources without the data being weighted by sources close to the monitoring site. These four source apportionment studies can be compared with the NEI source contributions from 1996 to 2001 for the five-county metro area.[46]

- The study using data from 1982 was the only one of the CMB studies that included the wood smoke source in its analysis (the other two used data from the summer months when there is not likely to be much wood smoke, unless there are wildfires). This CMB study showed that at sites within the city, wood smoke contributed between 9% and 19% of total primary $PM_{2.5}$ emissions.[132] Data from the site downwind of the city only showed wood smoke accounting for 4% of emissions. The PMF study, also downwind of the city, did not identify wood smoke as a significant source of $PM_{2.5}$.[133] Finally, the NEI shows that residential fuel combustion (the main source of wood smoke) made up 10% of total primary $PM_{2.5}$ emissions.[46] The NEI also apportions an additional 9% of fine aerosol emissions to "other combustion" sources. Some of these may have profiles similar to wood smoke.

- Mobile sources are a significant contributor to $PM_{2.5}$ in Los Angeles. There was a large variability in the estimated source magnitude between the source apportionment studies. If the source contributions from the four Los Angeles area sites are averaged, the three CMB studies predict that mobile sources account for 28%,[131] 48%,[132] and 68%[13] of total primary $PM_{2.5}$ emissions. The PMF study showed mobile sources contributing to 43% of total primary $PM_{2.5}$ emissions downwind of the city.[133] There was no noticeable geographic trend shown in the studies that used multiple sites. All of the source apportionment studies separated diesel and gasoline emissions. These studies predict that diesel combustion accounts for between 55% and 94% of mobile source $PM_{2.5}$ emission.[13,131–133] The NEI predicts that mobile sources should only account for 18% of fine aerosol emissions in the Los Angeles metro area, 62% of which would be from diesel engines.[46]
- In the Los Angeles area, bottom-up and top-down estimates of fugitive dust contributions are in fairly good agreement. The NEI predicts that 34% of fine aerosols come from fugitive dust.[46] The source apportionment studies predict that fugitive dust accounts for between 14% and 40% of primary $PM_{2.5}$.[13,131–133] Again, there was no apparent geographic trend within the metro area. Several of the studies using ambient measurements were able to differentiate between different sources of fugitive dust. One study found that 14–33% of the fugitive dust came from crustal material sources rather than road dust.[131] Another study found that 46% of the fugitive dust was from local soil, whereas the rest originated as Asian dust.[133]
- The 1996–2001 NEI did not account for cooking emissions. CMB studies identified between 3% and 21% (most sites showed between 7% and 15%) of primary $PM_{2.5}$ coming from cooking emission.[13,131,132] The PMF study did not identify cooking as a significant source.[133]
- Two of the source apportionment studies were able to identify sea salt emissions. One study found that it only contributed 1–2% of primary fine aerosol emissions,[131] whereas the other found that it contributed 18%.[133]
- The NEI included some source categories that were not identified by ambient measurements. It predicted that three of these source categories would contribute a total of approximately 15% of primary fine aerosol emissions. These source categories included agriculture/food industry emissions (does not include cooking), wood/paper industry emissions, and solid waste incineration and open burning of trash.[46]

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