(Stanier et al., 2004). For all 24 European locations, the average upper limit percentage of particles smaller than 0.1 μm ranged from 67 to 85%.

No such large-scale summary of U.S. data is possible, because there are few long-term data on number size distributions in the U.S. Number size distribution data have been reported for an 8-year period from 2002 to 2009 in Rochester, and number concentrations averaged 4,730 cm^{-3} for 0.01 to 0.05 μm particles, 1,838 cm^{-3} for 0.05 to 0.1 μm, and 1,033 cm^{-3} for 0.1 to 0.5 μm (Wang et al., 2011). This corresponds to 90% of total particles smaller than 0.1 μm. This is a larger fraction than the European range, but the lower size limit was 0.01 μm, compared to 0.03 μm for the European network data (Wang et al., 2011). Long-term trends for this period are summarized in Section 2.5.2.1.4. These data can also be compared to earlier observations of particle number concentrations for eight size ranges for a full year from the Pittsburgh Air Quality study (Stanier et al., 2004). Using their data, it is possible to calculate that 90% of the number of particles were also smaller than 0.1 μm and that 98% were smaller than 0.2 μm.

2.5.1.1.6 $PM_{2.5}$ Components

It is useful to distinguish between bulk PM components and more finely speciated components. The term bulk component refers to a large component category like OC, sulfate, or nitrate, which is monitored in networks like CSN or IMPROVE and usually makes up a substantial portion of PM mass. It is also used to differentiate broad categories of components like OC and crustal material, which are considered bulk components, from more finely speciated components like individual organic compounds and elements, which are usually present in lower amounts.

Figure 2-19 shows contributions of sulfate, nitrate, OC, EC, crustal material, and sea salt to $PM_{2.5}$. A major change in $PM_{2.5}$ composition compared to the 2009 PM ISA (U.S. EPA, 2009) is the reduction in sulfate concentrations, resulting in a smaller sulfate contribution to $PM_{2.5}$ mass in 2013–2015 compared to what was reported for 2005–2007 in the 2009 PM ISA, especially in the Eastern U.S. As a result, at many locations sulfate has been replaced as the greatest single contributor to $PM_{2.5}$ mass by organic material or nitrate. This long-term trend demonstrating a reduction in sulfate concentrations is described in more detail in Section 2.5.2.1.4.

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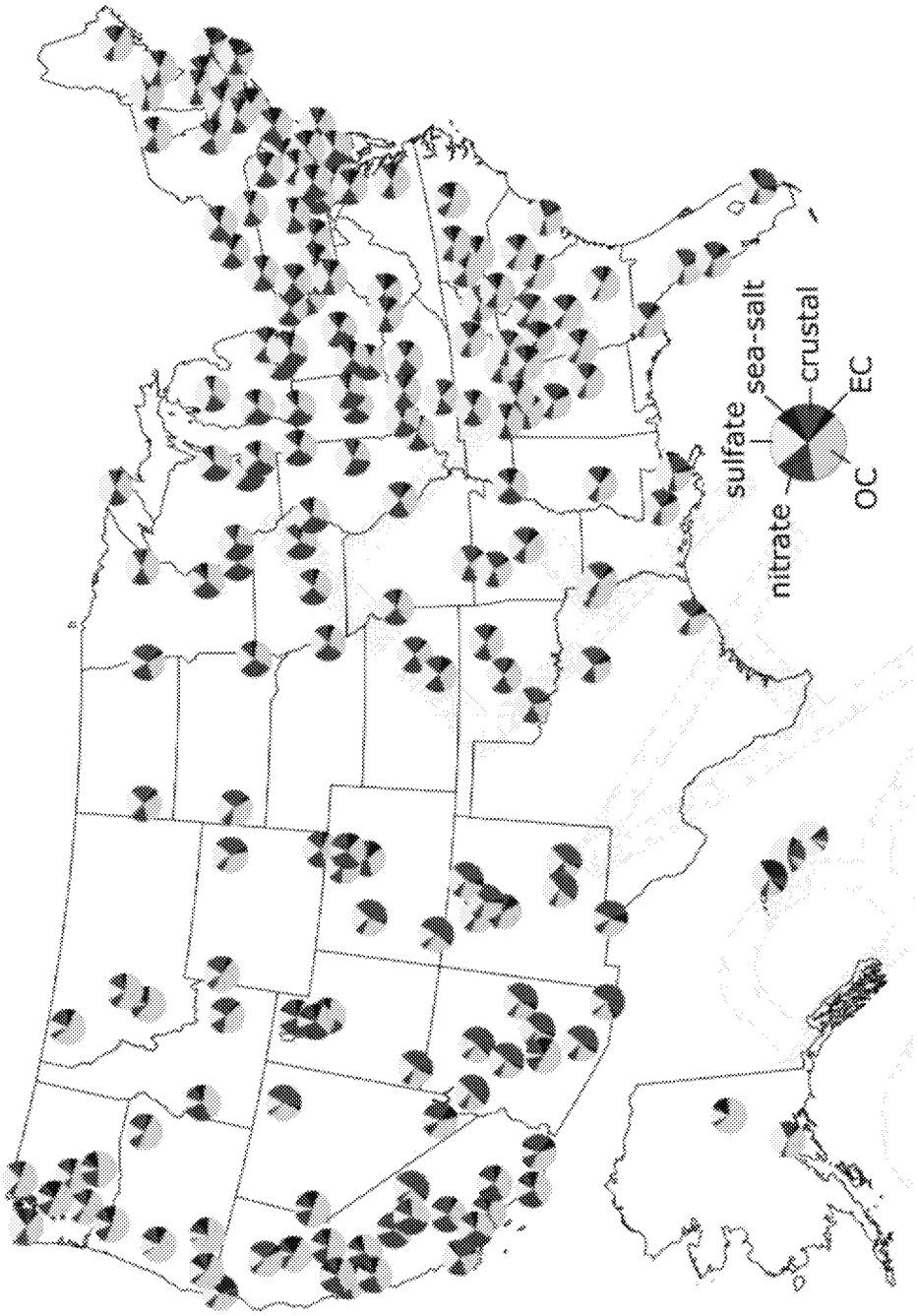

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2013–2015.

Figure 2-19 Contributions of sulfate, nitrate, organic carbon (OC), elemental carbon (EC), crustal material, and sea salt to $PM_{2.5}$.

Regional patterns of component contributions to $PM_{2.5}$ in Figure 2-19 are similar to those reported in the 2009 PM ISA (U.S. EPA, 2009). Sulfate and OC are the species with the highest

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contribution to total mass in most eastern locations and OC usually makes the greatest contribution to $PM_{2.5}$ mass in the west, although sulfate, nitrate, and crustal material can also be abundant (U.S. EPA, 2009). Urban and rural sulfate are both substantially higher in the East than in the West (Hand et al., 2012c). The highest nitrate concentrations are found in the west, particularly in California, but with some elevated concentrations in the upper Midwest. Larger contributions of OC to $PM_{2.5}$ mass observed in the Southeast and the West than in the Central and Northeastern U.S. are consistent with larger OC concentrations in those regions described in the 2009 PM ISA (U.S. EPA, 2009). The ratio of organic mass to OC mass depends on source and aerosol age, and was discussed in detail in the 2009 PM ISA (U.S. EPA, 2009). Based on speciation data from 15 cities reported in the 2009 PM ISA, EC contributed a smaller fraction of $PM_{2.5}$ mass than sulfate, nitrate, or OC, but consistently accounted for 4−11% of $PM_{2.5}$ (U.S. EPA, 2009).

Nationally, higher urban than rural OC and EC concentrations were reported by (Hand et al., 2013) and differences in urban and rural seasonal patterns for OC and EC were also observed. They also reported the highest rural OC and EC concentrations were in the Northwest and Southeast (Hand et al., 2012c). On average, OC and EC concentrations were both more uniformly distributed in the eastern U.S., but more localized in the West, with the highest urban concentrations in the West during fall and winter (Hand et al., 2013). However, EC concentrations were more consistent across cities regardless of region in the 2009 PM ISA (U.S. EPA, 2009). In the Southeastern U.S., annual average primary OC concentrations were estimated to exceed annual average secondary OC concentrations, but secondary OC could exceed primary OC at rural sites during the warmest months, and secondary OC concentration also showed little difference between urban and rural sites (Blanchard et al., 2013).

A large fraction of organic PM can be water soluble (Mwaniki et al., 2014). During the summer CalNex 2010 campaign (Kelly et al., 2014), water soluble $PM_{2.5}$ was at a maximum in the morning and in the evening in the San Joaquin Valley of California. In the same study, nitrate was present at higher concentrations than sulfate or ammonium, averaging 0.8 $\mu g/m^3$ and there were hourly average concentrations greater than 25 $\mu g/m^3$ during the winter 2013 DISCOVER-AQ campaign (Kelly et al., 2018).

Fine soil concentrations are higher in the Southwest than in other parts of the U.S., and also exhibit seasonal patterns for urban and rural sites (Hand et al., 2012c). High PM concentrations in urban desert areas were associated with a substantial contribution from crustal material from both coarse and fine PM (Wagner and Casuccio, 2014). Soil related PM also contributes substantially to $PM_{2.5}$ in populated areas of other parts of the world (Satsangi and Yadav, 2014). The pattern of higher crustal material contributions to $PM_{2.5}$ in drier areas of the Western U.S. can also be seen in Figure 2-19 with the examples of Phoenix and Denver.

There are a few new results to add to the body of literature on elemental composition, concerning both ambient observations and sources. In Southern California the most abundant elemental species was sulfur, followed by Si, Fe, Ca, and Al, with soil related elements accounting for 51% of total elemental

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mass measured. New research to investigate sources further explored the importance of brake wear, lubricating oils, gasoline and diesel combustion, secondary sulfates, sea salts, and biomass burning as sources of trace elements (Na and Cocker, 2009). New research on atmospheric iron indicate that extent of aqueous solubility of iron present in PM is related to sulfur content of the PM (Oakes et al., 2012b). In Atlanta, iron concentrations exhibit considerable fluctuation, and reach up to 300 to 400 ng/m^3 for a few hours at time, (Oakes et al., 2012b). In Atlanta Fe(II) accounted for between 5 and 35%, or an average of about 25% of total soluble iron (Oakes et al., 2012a). In rural samples copper and zinc were found to be mainly present as sulfates and also nitrates in $PM_{2.5}$ in rural samples, but copper and zinc compounds found in larger particles were similar to copper and zinc compounds found in soil (Osan et al., 2010).

2.5.1.1.7 $PM_{10-2.5}$ Components

It was noted in the 2004 AQCD (U.S. EPA, 2004) that concentrations of most elements differed between $PM_{2.5}$ and $PM_{10-2.5}$, but that concentrations of some metals were similar between the two size fractions. It was also noted that this contrasted with earlier years with less controlled combustion, when Pb and other metals were much higher in $PM_{2.5}$.

Components of $PM_{10-2.5}$ are not routinely monitored like they are for $PM_{2.5}$, and information on $PM_{10-2.5}$ composition is largely limited to specific local studies. In the Southeast, OC and nitrate made similar fractional contributions to $PM_{2.5}$ and $PM_{10-2.5}$, but there was much less sulfate and EC in $PM_{10-2.5}$ than $PM_{2.5}$, and much more major metal oxides (U.S. EPA, 2009). In Los Angeles, crustal material and trace elements accounted for 47.5% of total reconstructed coarse PM mass, with secondary ions (sulfate, nitrate, ammonium, 22.6%) and organic matter (19.7%) also making important contributions, and elemental carbon was a less significant component, accounting for less than 2% of the mass (Cheung et al., 2011). Los Angeles crustal materials had low water solubility, but Ba and Cu were modestly water soluble and activity due to reactive oxygen species was most highly associated with water soluble elements, V, Pd, Cu and Rh in Los Angeles (Cheung et al., 2012a).

In the desert southwest, crustal material is the dominant component of $PM_{10-2.5}$, sometimes accounting for more than half of the mass, followed by organic matter, accounting for 15% (Clements et al., 2014a, 2013). High correlations between $PM_{2.5}$ and PM_{10} indicated that a large component of the fine fraction was derived from dust (Clements et al., 2013). In Denver and Phoenix, $PM_{10-2.5}$ made a greater contribution to total ambient PM_{10} mass than in other cities (U.S. EPA, 2009). PM in Denver has been studied in more detail since then. Coarse PM concentrations were attributed to crustal material, road salt, vehicle abrasion and sulfate (Clements et al., 2014b).

While crustal material often makes the greatest contribution to $PM_{10-2.5}$ mass, the organic fraction also makes a substantial contribution. In the Southeast, organic and elemental carbon accounted for approximately 30% of $PM_{10-2.5}$. Primary biological aerosol particles (PBAP), which consist of microorganisms and fragments of living things, can account for a large fraction of $PM_{10-2.5}$ mass (U.S.

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EPA, 2009). These have been measured by treating collected PM with a dye which only reacts with protein containing material (Matthias-Maser et al., 2000). PBAP cannot be distinguished from other types of OC by methods used in monitoring networks. New research on sources of PBAP was summarized in Section 2.3.3. New information on the nature of bioaerosols and biological material associated with particles is well-described in the review by (Froehlich-Nowoisky et al., 2016). PBAP includes living and dead organisms, e.g., algae, archaea, bacteria and viruses, dispersal units, e.g., fungal spores and plant pollen, various fragments or excretions, e.g., plant debris and brochosomes. This class of material can range in size from 1 nm (individual proteins) to 5 millimeters (pollen grains). Summertime aerosols in Phoenix were abundant in biological compounds (e.g., sugars and fatty acids), present almost exclusively in the coarse size fraction (Cahill, 2013).

A pilot study on $PM_{10-2.5}$ species monitoring was carried out to develop target species, evaluate analytical methods and field performance, and to assess sampling and operational issues for routine measurement of $PM_{10-2.5}$ species (U.S. EPA, 2015). Samples collected in all seasons over a period of 1 year in both Phoenix and St. Louis indicated that soil oxides dominated $PM_{10-2.5}$ mass, with organic matter accounting for 10–20%. Sulfate and nitrate accounted for very little of the $PM_{10-2.5}$ mass, although they were substantial contributors to $PM_{2.5}$ mass. Soil oxides were by far the largest component in both locations throughout the year, except in St. Louis in winter, when soil and organic contributions were similar, but overall $PM_{10-2.5}$ concentrations were considerably lower (U.S. EPA, 2015).

2.5.1.1.8 Ultrafine Particle Components

There was little information on the composition of UFP presented in the 2009 PM ISA, although urban UFP was suspected to be rich in OC and EC, and sulfate was expected to be a substantial contributor in rural areas while new particle formation occurred (U.S. EPA, 2009). New research indicates that motor vehicles are a major, and frequently dominant, source of ultrafine particles in urban environments (Morawska et al., 2008). Chemical composition of these particles are determined by the composition of the used fuel and lubricating oil, driving conditions, and engine after-treatment system, as well as meteorological conditions, but generally PM from these sources consists mostly of agglomerates of solid-phase carbonaceous material, and can also contain metallic ash, adsorbed or condensed hydrocarbons and sulfur compounds, and liquid droplets consisting mainly of hydrocarbons and hydrated sulfuric acid that form very rapidly after the vehicle exhaust leaves a tailpipe (Liu et al., 2015; Saffaripour et al., 2015; Karjalainen et al., 2014; Rönkkö et al., 2014; Fushimi et al., 2011; Gidney et al., 2010; Heikkilä et al., 2009; Johnson, 2009).

The chemical composition of ultrafine particles originating from atmospheric NPF is tied heavily to their growth processes during their atmospheric aging. Direct observations during the period of atmospheric NPF show that the composition of particles originating from NPF is usually dominated by organic compounds, especially in forests (Han et al., 2014; Pennington et al., 2013; Pierce et al., 2012; Pierce et al., 2011), but also in many rural or urban environments (Bzdek et al., 2014; Setyan et al., 2014;

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Bzdek et al., 2013; Ahlm et al., 2012; Smith et al., 2008). Exceptions for this pattern are environments exposed to major sulfur emissions, in which sulfate may explain up to about half of the ultrafine particle mass (Vakkari et al., 2015; Crilley et al., 2014; Bzdek et al., 2012; Zhang et al., 2011b; Wiedensohler et al., 2009).

2.5.1.1.9 Reactive Oxygen Species

Particle acidity, oligomer formation and the production of reactive oxygen species (ROS) are interrelated, aqueous phase processes with direct consequence for aerosol concentrations, chemical composition and toxicity (Weber et al., 2016). Polymerization reactions responsible for generating oligomers in atmospheric particles require relatively high concentrations of H^+. The reactive forms of the transition metals that play a central role in production of particle phase ROS primarily exist in low pH aqueous conditions.

Sulfate is often the main acid component of $PM_{2.5}$, and largely determines its acidity. Contrary to expectations, declining SO_2 emissions along with fairly stable NH_3 emissions (see Section 2.3.2.1), have led to little long-term change in pH of $PM_{2.5}$ (see Section 2.5.1.1.6). Low pH conditions facilitate the formation of oligomers and HULIS in aqueous particles. Upwards of 90% oligomeric/high molecular weight material has been found in SOA formed in the presence of NO_X, including a substantial fraction of organic nitrogen compounds (Nguyen et al., 2011). Humic-like substances and smaller organic compounds have been implicated in the production of particle-phase ROS, along with transition metal ions, especially Cu and Mn (Verma et al., 2015).

The 2009 PM ISA described early chamber work on identifying reactive oxygen species (ROS) in secondary organic PM by Docherty et al. (2005). Under the conditions of their experiment, they produced very high yields (47 and 85%) of organic peroxides by reacting O_3 with α- and β-pinene. Reactive oxygen species include hydroxyl radical, organic peroxides and hydroperoxides. A discussion of the role of particle-phase ROS in human health effects can be found in Section 5.1.1.

Identification of individual components that act as ROS in PM is incomplete and an active area of research. The extent to which an ambient particle can engage in oxidative reactions depends on the concentration of aqueous oxidants, such as the hydroxyl radical (OH), and whether or not reactants capable of producing additional oxidants are present within the particle. Oxidants, in addition to OH, can be taken up from the atmosphere or chemically formed from processes such as photolysis of nitrate, nitrite, or hydrogen peroxide (H_2O_2), or Fenton-type reactions between H_2O_2 and Fe(II) (McNeill, 2015; Arakaki et al., 2013; Ervens et al., 2011; Herrmann et al., 2010). Organic species, such as quinones, can act as transition ion reducing agents, which allow oxidized form of an aqueous transition metal ion to produce more ROS (Shirai et al., 2012). Tuet et al. (2017) found that the identities of available reactive precursors in the particle phase, humidity and the fate the reactive intermediate were important

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determinants of particle reactivity. Atmospheric aging (oxidation) of organic aerosols has also been found to be an important indicator of ROS activity of ambient PM (Saffari et al., 2016; Verma et al., 2015).

2.5.1.2 Urban and Neighborhood Scale Variability

2.5.1.2.1 $PM_{2.5}$

Understanding spatial variation at the neighborhood and urban scale is important for interpreting data from community monitors. Because of its longer atmospheric lifetime (see Section 2.2), $PM_{2.5}$ is expected to exhibit less spatial variability on an urban scale than UFP or $PM_{10-2.5}$. In the 2004 PM AQCD (U.S. EPA, 2004) annual average $PM_{2.5}$ concentration differences between monitors within the urban area were compared for 17 urban areas. The difference in concentration between monitors with the highest and lowest concentrations ranged from less than 1 μg/m^3 (Baton Rouge, LA) to more than 8 μg/m^3 (Pittsburgh, PA). The difference exceeded 6 μg/m^3 in 6 of the 17 cities (Pittsburgh, Cleveland, Chicago, Detroit, St. Louis, Seattle), in 5 of which the highest $PM_{2.5}$ concentrations were between 20 and 22 μg/m^3. In the remaining city (Seattle) concentrations ranged from 6 to 12 μg/m^3.

The degree of spatial uniformity within urban areas also varied depending on location (U.S. EPA, 2004). Intra-urban spatial variability of $PM_{2.5}$ concentrations was discussed in considerable quantitative detail in the 2009 PM ISA, using a number of comparison statistics (U.S. EPA, 2009). In most metropolitan areas correlations between $PM_{2.5}$ monitoring sites up to a distance of 100 km from each other were greater than 0.75, with the notable exceptions of Denver, Los Angeles, and Riverside (U.S. EPA, 2009). However, while $PM_{2.5}$ concentrations at different sites within an urban area can be highly correlated, significant differences in concentration can occur on a given day (U.S. EPA, 2009).

Several recent publications have addressed urban scale spatial variability. Urban concentrations are often several μg/m^3 above regional background concentrations. For example, Indianapolis urban concentrations are on average 3.9 to 5. μg/m^3 higher than regional background (Sullivan and Pryor, 2014). Substantial spatial variation of $PM_{2.5}$ concentrations has been reported for New York City (Matte et al., 2013). Spatial variability was also demonstrated by a study indicating that $PM_{2.5}$ was present at significantly higher concentrations at urban sites than at upwind suburban sites in the greater New York area (Patel et al., 2009). Substantial differences in $PM_{2.5}$ concentrations between neighborhoods was also observed in Los Angeles (Fruin et al., 2014), but not in Boston (Patton et al., 2014). One of the contributing factors was that monitors are closer to each other in Boston, where more uniformity was observed. Sub-10 km spatial variability was identified as a contributor to poor results for satellite estimates of $PM_{2.5}$ from aerosol optical depth (AOD) using a 10 × 10 km grid (Lary et al., 2014; Chudnovsky et al., 2013b). In Indianapolis for time scales shorter than 1-day spatial variability was 2 to 3 times greater than temporal variability (Sullivan and Pryor, 2014). However, for 24-hour measurements

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of PM components temporal variability accounted for 90% of the variance in Detroit (Bereznicki et al., 2012).

Spatial variability arises from source proximity, with motor vehicles accounting for 24 to 36% and secondary sulfate for 17 to 35% of $PM_{2.5}$ among different residential monitoring areas in Detroit, MI (Duvall et al., 2012). Diesel exhaust was also identified as a major and variable source of $PM_{2.5}$ in New York City (Patel et al., 2009). Land use regression modeling based on 155 city-wide street-level locations in New York City (Clougherty et al., 2013) indicated that concentrations of $PM_{2.5}$ and other pollutants varied by more than a factor of two, with highest concentrations near midtown Manhattan. They also reported that density of oil-burning boilers along with total and truck traffic density explained more than 80% of $PM_{2.5}$ spatial variability (Clougherty et al., 2013). However, in Dallas $PM_{2.5}$ exposure was only moderately associated with motor vehicles and weakly associated with industrial sources, but strongly associated with population density (Zou et al., 2009). Overall, recent observations indicate that uniform $PM_{2.5}$ concentrations can occur, but that substantial spatial variability is also common.

2.5.1.2.2 PM_{10}

PM_{10} concentrations vary by as much as a factor of five over urban scale distances of 100 km or less and by a factor of two or more over scales as small as 30 km (U.S. EPA, 2009; Alexis et al., 2001). Differences in PM_{10} measurements across 15 cities were summarized in the 2009 PM ISA (U.S. EPA, 2009). PM_{10} concentrations were less well correlated than $PM_{2.5}$, probably because of greater spatial variability of $PM_{10-2.5}$ (see Section 2.5.1.2.3). For monitors less than 4 km apart an average correlation of 0.93 between $PM_{2.5}$ monitors and 0.70 for PM_{10} monitors was observed (U.S. EPA, 2009). Spatial and temporal differences in PM_{10} concentrations have also been predicted from models based on the geographic information system; meteorological and copollutant data for both fine and large spatial scales and distance to road; elevation; and proportion of low-intensity residential, high-intensity residential, industrial, commercial, and transportation land use within 1 km have all been reported to be statistically significant predictors of measured PM_{10} (Blanchard et al., 2014; Paciorek et al., 2009; Yanosky et al., 2009); (Yanosky et al., 2014).

2.5.1.2.3 $PM_{10-2.5}$

As indicated in the 2004 PM AQCD (U.S. EPA, 2004), the shorter lifetime of $PM_{10-2.5}$ leads to lower spatial correlations for $PM_{10-2.5}$ than for either $PM_{2.5}$ or PM_{10} concentrations (U.S. EPA, 2009, 2004). Errors in measurement (see Section 2.4.4) can also contribute to lower spatial correlations of $PM_{10-2.5}$ (U.S. EPA, 2004). Recent observations from several cities indicate that there is often, but not always, considerable spatial variability in $PM_{10-2.5}$ concentrations in urban areas, that they are often related to specific industrial sources, and that concentrations of specific chemical components can be more variable than mass. In Detroit $PM_{10-2.5}$ was 5 $\mu g/m^3$ higher in two industrial areas, and 8 $\mu g/m^3$

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higher in an area heavily impacted by traffic than average concentrations in other parts of the city, and not very consistent with central site monitor concentrations (Thornburg et al., 2009). Poor correlations between monitors were also observed in Los Angeles (Pakbin et al., 2010) and between industrial and suburban sites in Cleveland (Sawvel et al., 2015). In Rochester, NY, where major coarse particle sources were road dust and biological particles, considerable heterogeneity in both composition and concentrations were also observed between different sites (Lagudu et al., 2011).

2.5.1.2.4 Ultrafine Particles

As described in Section 2.5.1.1, UFP spatial variability increased with increasing distance between measurements, increasing source variation in the area studied, and increasing particle size within the UFP size range. (U.S. EPA, 2009). Particularly high spatial variabilities have been observed near roads with heavy traffic, where numerous observations of UFP number concentration declining sharply with distance from roadways have been reported (U.S. EPA, 2009).

More recently, spatial variability of UFP was compared between studies of two locations, Los Angeles, CA (Hudda et al., 2010; Krudysz et al., 2009; Moore et al., 2009) and Rochester, NY (Wang et al., 2012). These two studies provide an interesting comparison because the two studies were similar in domain size. The comparison is summarized in Table 2-8. It should be noted that the Los Angeles studies employed SMPS for particle size distribution measurements, while the Rochester study used a FMPS. Both Krudysz et al. (2009) and Hudda et al. (2010) indicated that regionally transported PM from upwind urban areas of Los Angeles lowered spatial variability by acting as a "homogenizing" factor during favorable meteorological conditions. This effect was not noticeable in Rochester, NY (Wang et al., 2012). Nevertheless, significant variability among sites was observed in both studies.

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Table 2-8 Comparison between two urban-scale studies of UFP seasonal and spatial variability.

	Los Angeles, CA (Krudysz et al., 2009)	Rochester, NY (Wang et al., 2012)
Area	11 × 11 km, urban	9 × 9 km, urban
Sites	13 sites	12 sites
Instrumentation	SMPS (14−793 nm), CPC (>7 nm)	FMPS (with one SMPS in a fixed site), 5.6 to 560 nm
Levels of average total number concentrations	5,300 to 27,000 particles/cm^3	9,025 (summer), 10,939 (winter), 4,955 (spring), and 14,485 (fall) particles/cm^3
Seasonal variability	Relatively higher levels observed in the fall/winter than in the summer	Relatively higher levels observed in the fall/winter than in the spring; Relatively high 100−500 mode in the summer
Coefficient of divergence (COD)	>0.2 on average for all particles measured, 0.25 to 0.6 for size-dependent average COD	No clear overall pattern
Size-dependency	Number concentrations of smaller particles (<40 nm) differ from site to site, whereas larger particles tended to have similar concentrations at various sampling locations.	No clear overall pattern

Source: Krudysz et al. (2009).

2.5.1.2.5 Chemical Components

A detailed analysis of 15 urban locations in the 2009 PM ISA (U.S. EPA, 2009) indicated a generally fair degree of spatial uniformity in bulk $PM_{2.5}$ components. Exceptions were noted in one or two cities for crustal material, nitrate, elemental carbon, organic carbon and nickel (U.S. EPA, 2009). More recent observations have focused mainly on carbonaceous components across urban areas. Black carbon (BC) concentrations were 2 to 3 times higher at urban locations than at suburban locations in the greater New York area (Patel et al., 2009). There were several reports of higher concentrations of some PM components near roads with heavy traffic than other urban locations. For example, carbonaceous aerosols exhibited substantial intra-urban variability in Detroit, MI and Cleveland, OH that was consistent with local sources, with EC higher at sites adjacent to freeways and busy surface streets (Snyder et al., 2010). Site to site variability in OC was approximately 7% at distances from 0.5 to 4 km, but between 4−27% at distances 4 to 100 km. However, more finely speciated organic components differed by as much as 60% at the 0.5 to 4 km scale and up to 200% at the 4−100 km scale (Snyder et al., 2010). PAHs and steranes

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along with OC and EC were found to be higher near roads with heavy traffic than in other urban locations (Xie et al., 2012). Differences of a factor of 2 to 3 between concentrations on major streets and at background locations in the same city in the Netherlands were also observed for chromium, copper, and iron, elements that were mainly present in the coarse fraction, as well as for black carbon and particle number count (Boogaard et al., 2011).

2.5.2 Temporal Variability

2.5.2.1 Regional Trends

Differences in national average concentrations and regional variability between data from immediately prior to this assessment and the 2009 PM ISA (U.S. EPA, 2009) were discussed in Section 2.5.1.1, which demonstrated substantial decreases in PM concentrations since publication of the 2009 PM ISA (U.S. EPA, 2009). This section expands on those observations by exploring long-term trends that extend back as far as 2000, when widespread network measurements of urban $PM_{2.5}$ began, in order to provide more complete assessment of trends.

2.5.2.1.1 $PM_{2.5}$

Figure 2-20 show how $PM_{2.5}$ concentrations have decreased substantially at almost all $PM_{2.5}$ monitoring sites between the periods 2003–2005 and 2013–2015, with especially large decreases in the Eastern U.S. Figure 2-21 also shows a decreasing trend of $PM_{2.5}$ concentrations as a time series using national data from network monitoring sites throughout the U.S. Overall, $PM_{2.5}$ concentrations have decreased substantially nationwide since the 2003–2005 period, especially in the Eastern U.S. $PM_{2.5}$ concentrations derived from satellite data also exhibit a decreasing trend, of $-0.39 + 0.10$ μg/m^3 per year averaged over a 1 by 1 degree grid (Boys et al., 2014).

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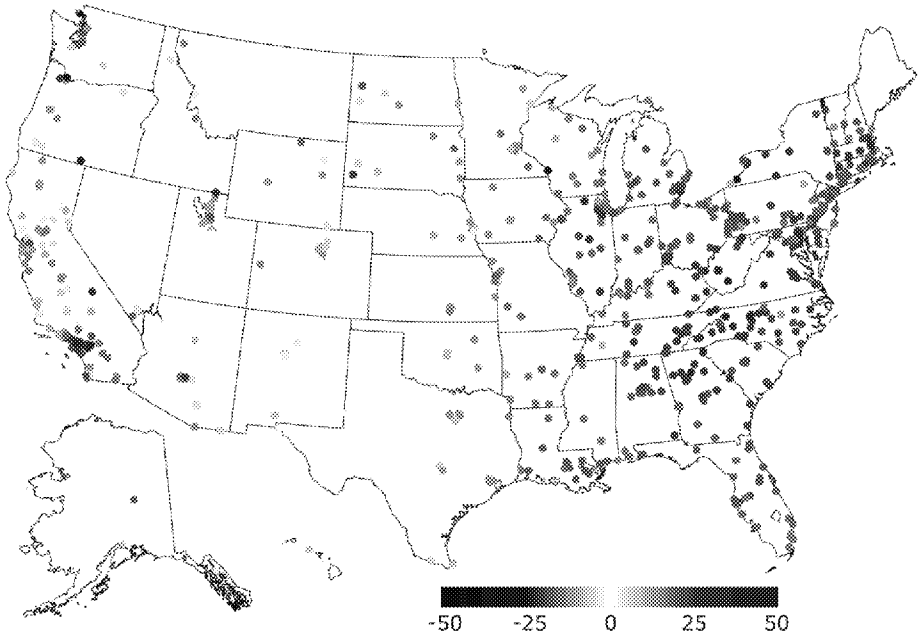

Blue indicates a decrease and red indicates an increase. Percentage increase or decrease is indicated by color intensity of the circle.

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2003–2005 and 2013–2015.

Figure 2-20 Increase or decrease in 3-year annual average $PM_{2.5}$ concentrations between 2003–2005 and 2013–2015.

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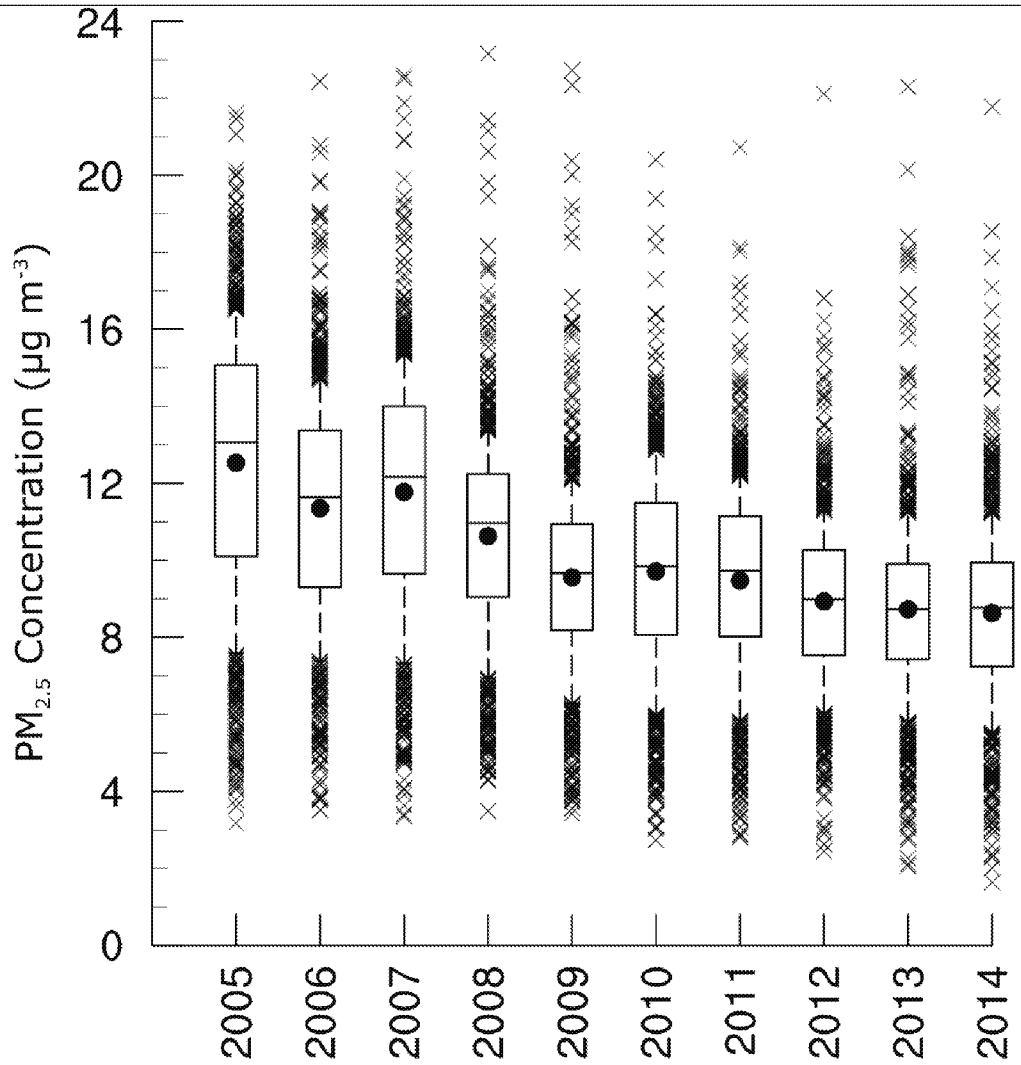

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2005–2014.

Figure 2-21 Average $PM_{2.5}$ concentration trends 2005–2014

The predominant downward trends shown in Figure 2-21 are a continuation of the decreasing trend in $PM_{2.5}$ concentration reported in the 2009 PM ISA (U.S. EPA, 2009), in which a 10% decrease in annual average $PM_{2.5}$ concentrations between the 3-year period from 1999–2001 and the 3-year period from 2005–2007 was described. Figure 2-22 shows an overall decrease in monthly and annual $PM_{2.5}$ average and 90th percentile concentrations over the 16-year period from 2000–2015, as well as a steadily shrinking summer peak, across all reporting FRM site-level monitors in the U.S. (Chan et al., 2018). Over this period $PM_{2.5}$ concentration averaged over the entire network decreased by 5 µg/m^3 and 90th percentile concentrations decreased by 9 µg/m^3 (Figure 2-22). It is evident from Figure 2-22 that the sharpest decrease occurred in 2008–2010.

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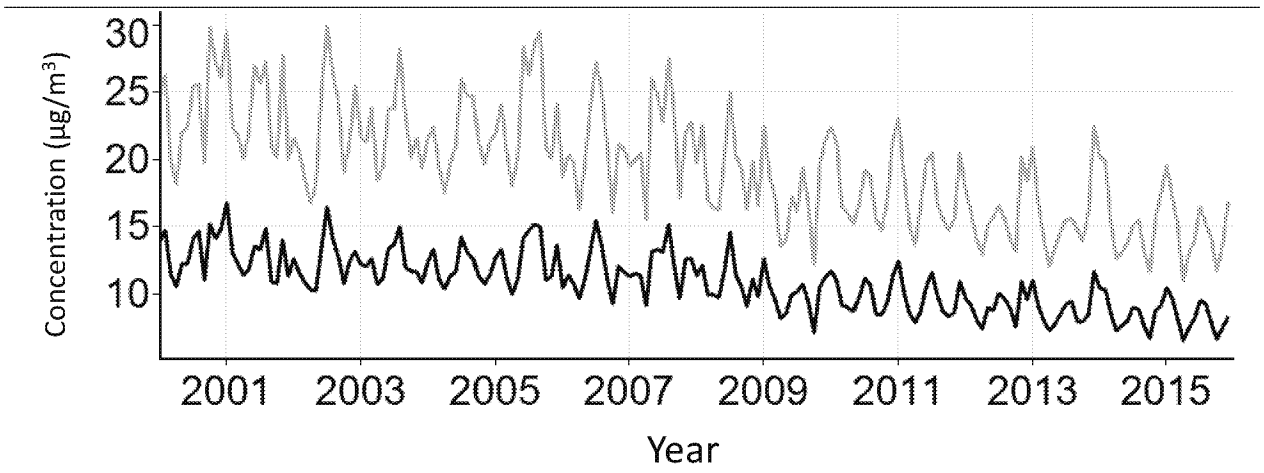

Black = mean, gray = 90th percentile.
Source Permission pending: Chan et al. (2018).

Figure 2-22 Long-term trend in national monthly and annual average $PM_{2.5}$ concentrations (µg/m³) from 2000–2015.

2.5.2.1.2 PM_{10}

Over the longer term PM_{10} has decreased steadily in several urban areas over the past several decades (U.S. EPA, 2004). Figure 2-23 shows a map of concentration trends in 98th percentile PM_{10} concentrations between 2003–2005 and 2013–2015 and Figure 2-24 shows a time series of national PM_{10} concentrations from 2005–2014. Most sites in the Eastern U.S. show decreasing concentrations over this period, consistent with the data of Table 2-5. However, there are locations in California, the Southwest, the Rocky Mountains, and the Great Plains that exhibit substantial increases in 98th percentile PM_{10} concentrations. The observed decreases in PM_{10} concentrations in many locations are consistent with similar observations for annual average $PM_{2.5}$ concentrations (see Section 2.3.4), reflecting that $PM_{2.5}$ has accounted for the majority of PM_{10} in the Eastern U.S. and a large fraction of PM_{10} throughout the U.S. over the period of decline. However, Figure 2-24 shows no evidence of a nationwide trend of decreasing PM_{10} concentrations in a time series of PM_{10} concentrations from network monitoring sites throughout the U.S.

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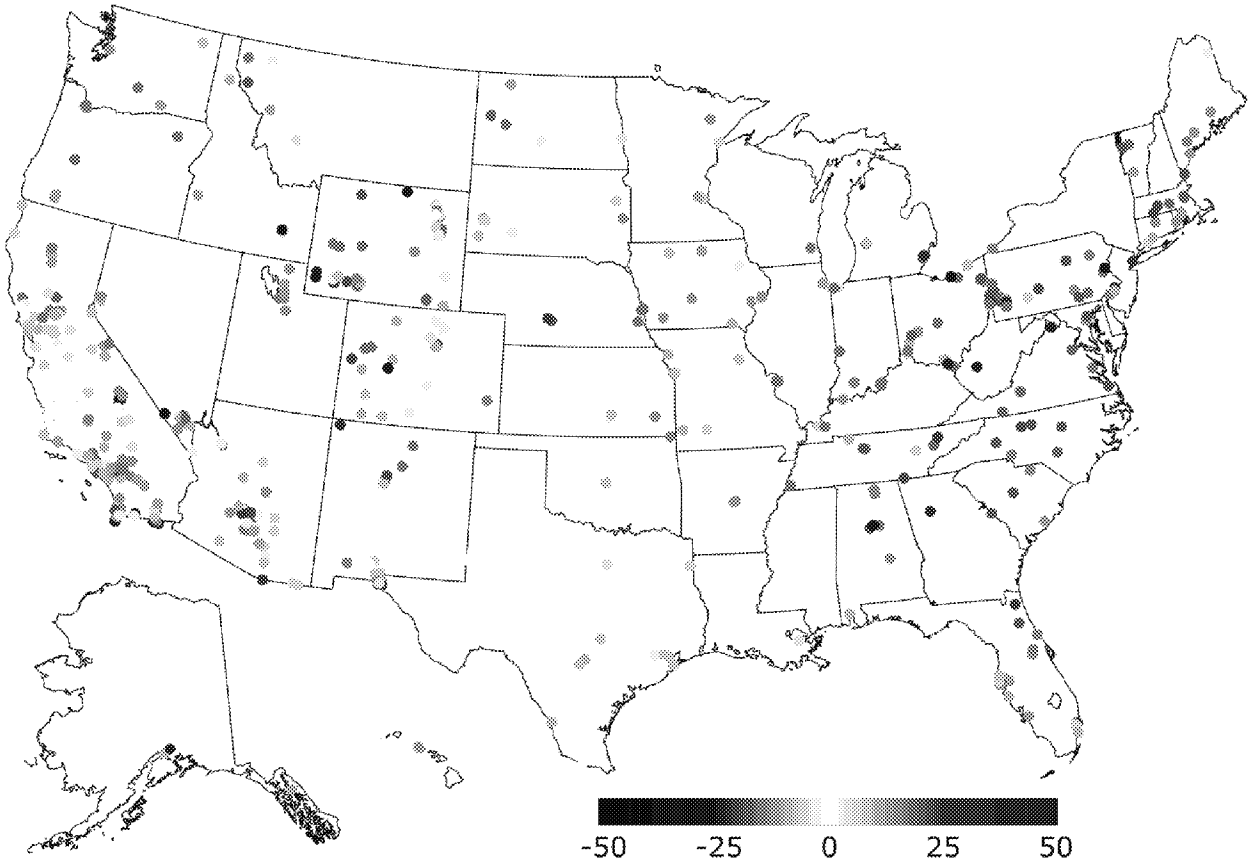

Blue indicates a decrease and red indicates an increase. Percentage increase or decrease is indicated by color intensity of the circle.

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2003−2005 and 2013−2015.

Figure 2-23 Increase or decrease in 98th percentile 24-hour PM_{10} concentrations between 2003−2005 and 2013−2015.

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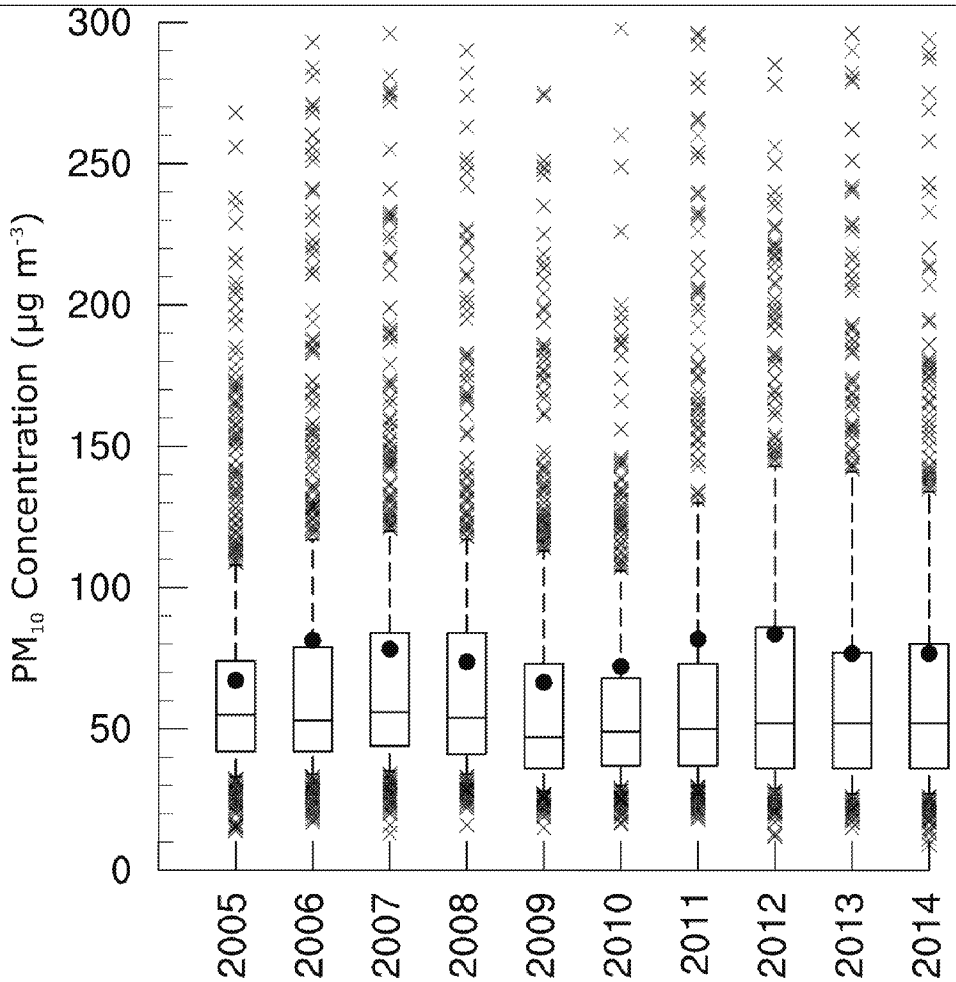

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2003−2005 and 2013−2015.

Figure 2-24 PM_{10} 2nd highest concentration trends from 2005−2014.

2.5.2.1.3 $PM_{10-2.5}$

Long-term concentration trends for urban $PM_{10-2.5}$ are difficult to determine from network data because $PM_{10-2.5}$ monitoring was too recently implemented. However, some NCore stations began $PM_{10-2.5}$ measurements in the mid-2000's and IMPROVE measurements of $PM_{10-2.5}$ have been operating even longer, and although IMPROVE sites are mostly rural, some are collocated with CSN sites. These could be analyzed for long-term trends. In a Los Angeles field study $PM_{10-2.5}$ decreased by 0.39 µg/m^3 from 19 to 15 µg/m^3 for the period 1999 to 2009 compared to 0.92 µg/m^3 for $PM_{2.5}$ over the same period (Cheung et al., 2012b).

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2.5.2.1.4 Ultrafine Particles

Information on UFP concentrations is very limited, confined to very few network monitors that only recently became operational. Data from field studies have been published periodically, but are generally insufficient to assess long-term trends of UFP in any location. One exception is 8 years of UFP data from Rochester, NY, the particle number characteristics of which were summarized in Section 2.5.1.1.5 (Wang et al., 2011). On average over the 8 years that UFP data were collected in Rochester, total particle number concentrations were greater before 2006 than after 2006. This trend was most evident for particles between 0.01 and 0.1 μm. The difference was described as probably due to several changes in local sources due to the 2007 Heavy Duty Highway Rule, a reduction in local industrial activity, and the closure of a nearby coal-fired power plant (Wang et al., 2011).

2.5.2.1.5 Chemical Components

Figure 2-25 and Figure 2-26 show changes in the distribution of bulk $PM_{2.5}$ components, between the 3-year period from 2003–2005 and the 3-year period from 2013–2015. The most noticeable difference is the change in sulfate contribution, which dominates $PM_{2.5}$ mass in the East during the period 2003–2005, but by 2013–2015 it has declined enough that it is no longer the most abundant component in many Eastern locations.

In the 2009 PM ISA (U.S. EPA, 2009), sulfate is described as the most abundant component of $PM_{2.5}$ on a national average, with nitrate, particulate organic matter and sometimes crustal material also contributing substantially to $PM_{2.5}$ mass. The relative abundance of major $PM_{2.5}$ components has changed since the 2009 PM ISA (U.S. EPA, 2009), with lower contributions from sulfate and greater contributions of nitrate and particulate organic matter as a result of the steep decline in SO_2 emissions (see Section 2.3.2.1). The resulting decrease in sulfate concentrations closely follows the recent long-term decrease in $PM_{2.5}$ concentrations described in Section 2.5.2.1.1, and is magnified for monitoring sites in the Eastern half of the U.S., where sulfate has until recently been the most abundant $PM_{2.5}$ components, and where SO_2 emissions have declined the most.

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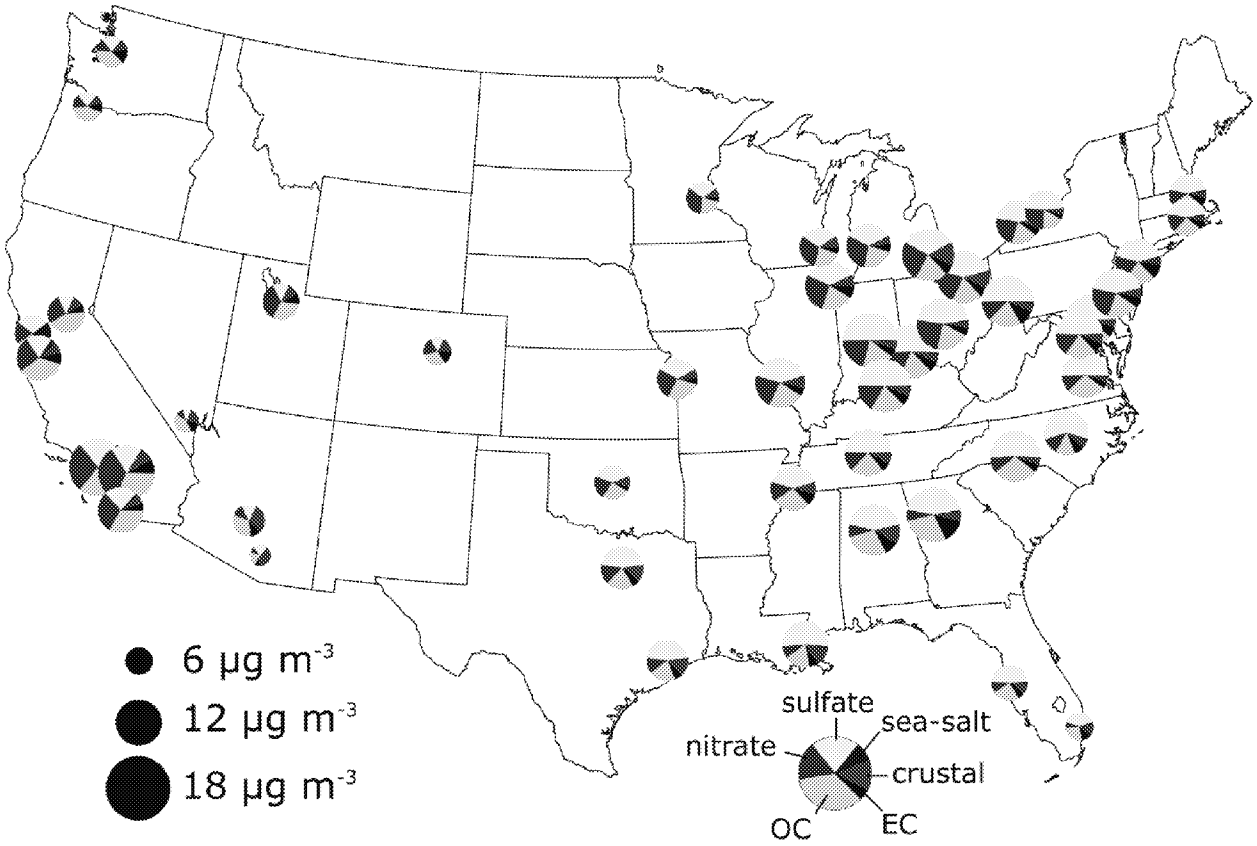

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2003−2005.

Figure 2-25 Contributions of sulfate, nitrate, organic carbon (OC), elemental carbon (EC), crustal material, and sea salt to $PM_{2.5}$ at selected sites 2003−2005.

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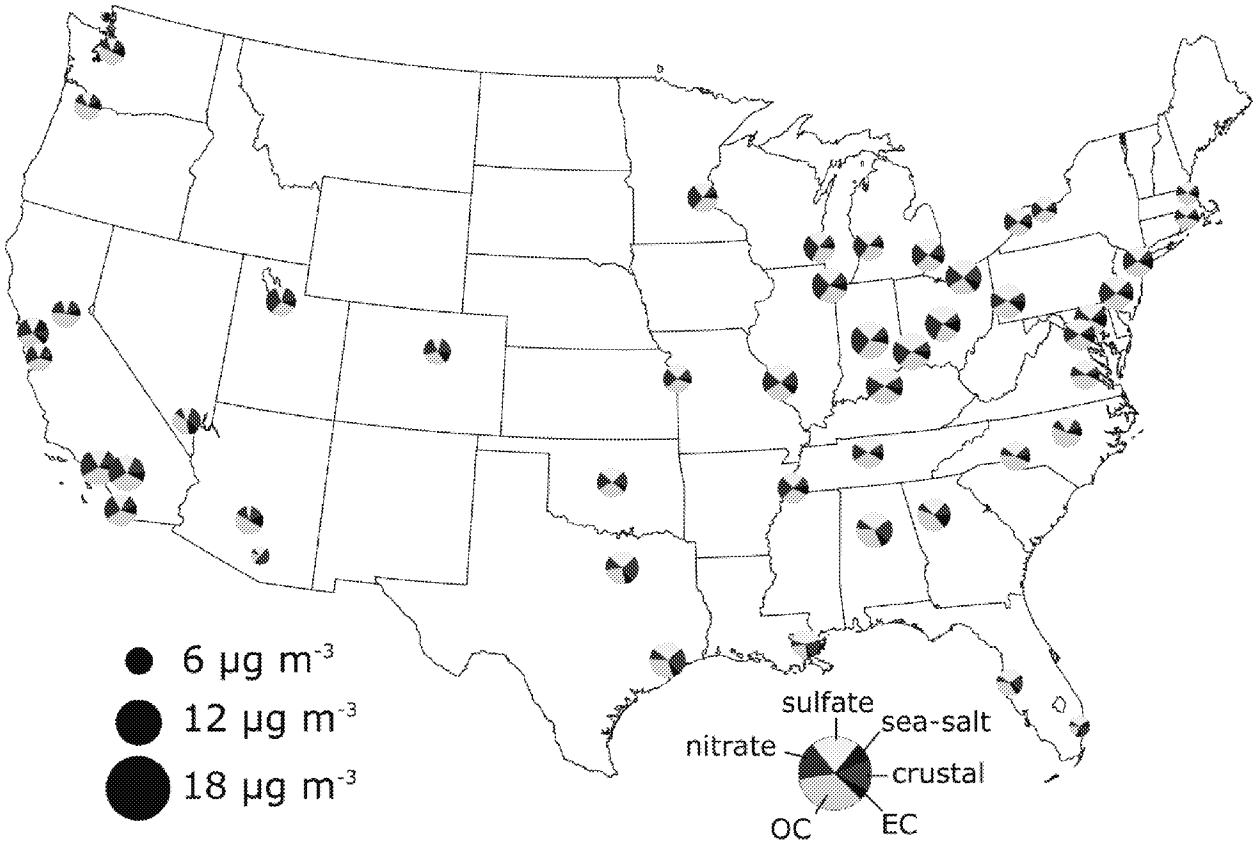

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2013–2015.

Figure 2-26 Contributions of sulfate, nitrate, organic carbon (OC), elemental carbon (EC), crustal material, and sea salt to $PM_{2.5}$ at selected sites 2013–2015.

Figure 2-27 shows $PM_{2.5}$ sulfate, nitrate and OC concentrations from 2000–2015 based on IMPROVE and CSN network data. A steep decline in sulfate concentration is observed, but less change is evident for nitrate and OC concentrations. Like the summer $PM_{2.5}$ maximum (Figure 2-22), the summer sulfate peak also declines to become almost imperceptible toward the end of the period. Based on these observations, it appears that decreases in SO_2 emissions (Section 2.3) have contributed to a substantial decrease in atmospheric sulfate concentrations. The declining sulfate concentrations are also consistent with CMAQ predictions of the sulfate response to decreasing SO_2 emissions. Because sulfate has accounted for such a large fraction of $PM_{2.5}$ mass, the decreasing trend in sulfate concentration is also manifested in lower $PM_{2.5}$ concentrations (Section 2.5.2.1.1) and smaller $PM_{2.5}/PM_{10}$ ratios (Section 2.5.1.1.4). However, sulfate is not the only $PM_{2.5}$ species that exhibited decreasing concentrations over this period, as described below.

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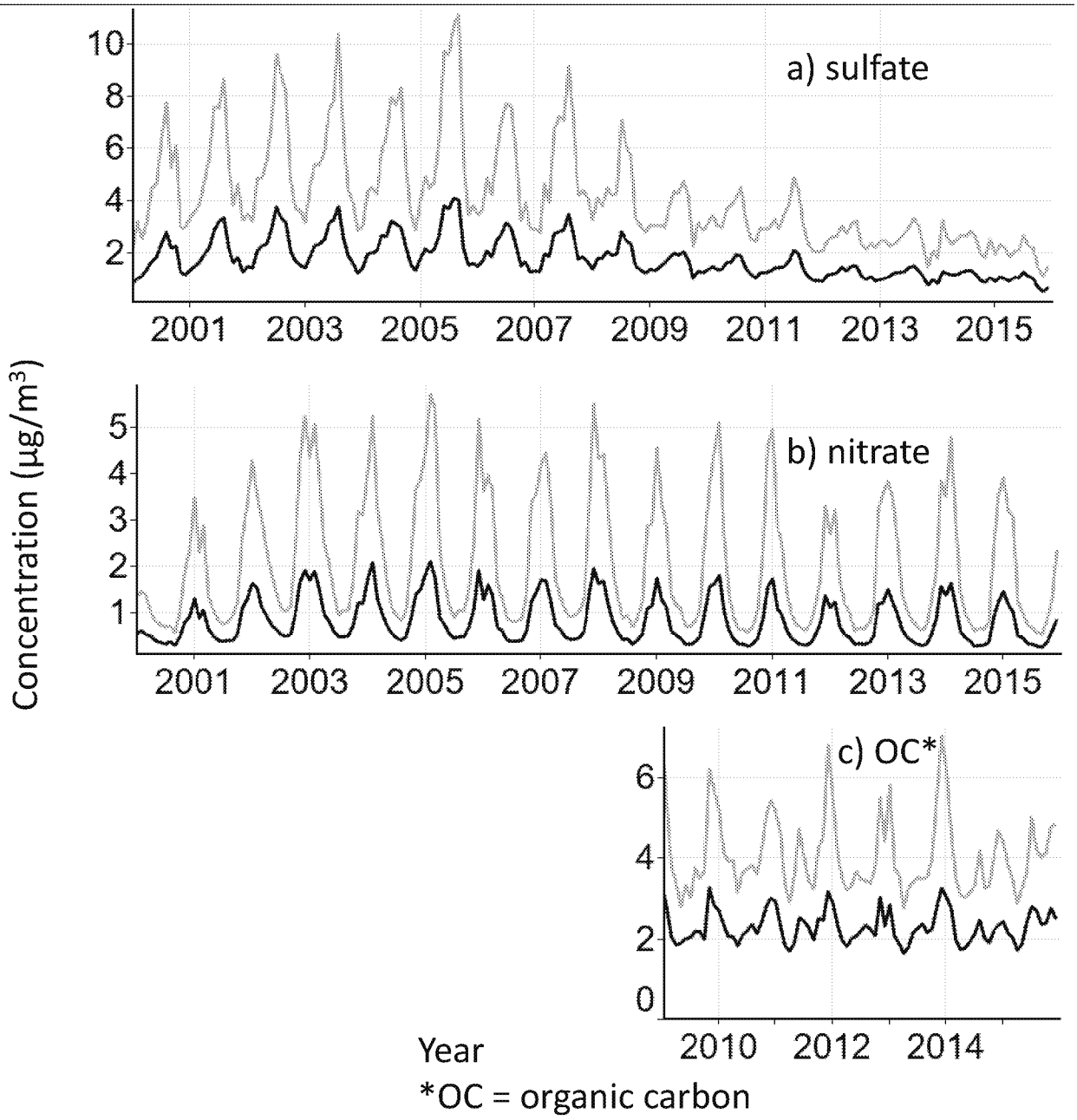

Black = mean, gray = 90th percentile.
Source Permission pending: Chan et al. (2018).

Figure 2-27 National monthly concentrations (µg/m³) of (a) sulfate, (b) nitrate, and (c) organic carbon (OC) from 2000−2015.

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Long-term trends in $PM_{2.5}$ component concentrations from the CSN and IMPROVE networks were also recently described in a series of papers (Hand et al., 2013; Hand et al., 2012a; Hand et al., 2012b). In general sulfate has decreased fairly consistently at rural sites at a rate of −2.7% per year from 1992 to 2010 (Hand et al., 2012b). An even steeper decrease in sulfate concentrations has been observed in the most recent years, of −4.6% per year at rural sites from 2001 to 2010 and −6.2% per year at urban sites from 2002−2010 (Hand et al., 2012b). This is similar to the rate of decrease of SO_2 emissions from power plants, and decreases were greater and more linear in the East, where power plant emissions had the greatest contributions to sulfate concentration (Hand et al., 2012b). However, in the winter in the northern and central Great Plains sulfate and nitrate concentrations have increased at a rate of over 5% per year over the period 2000−2010, in spite of decreased nationwide emissions (Hand et al., 2012a), and sulfate increases in spring in some parts of the West were also observed (Hand et al., 2012b). These increases could not be explained by known changes in local or regional emissions (Hand et al., 2012b). In the SEARCH network downward trends in mean annual sulfate concentrations from 1999 to 2010 ranged from -3.7 ± 1.1 to -6.2 ± 1.1% per year. The sulfate reduction was linearly related but not proportional to SO_2 decrease of -7.9 ± 1.1% per year from 1999 to 2010. Over the same period mean organic matter concentration decreased by -3.3 ± 0.8 to 6.5 ± 0.3% per year and elemental carbon by -3.2 ± 1.4 to -7.8 ± 0.7% per year (Blanchard et al., 2013). Total carbon (OC + EC) generally decreased in both urban and rural areas, with the strongest trends in the West (Hand et al., 2013).

For species that are more strongly influenced by local urban sources, trends are manifested more locally, and largely controlled by changes in local source emissions. Al, Fe, and Si decreased in Los Angeles, suggesting successful control of fugitive dust emissions, but Cu declined little, probably indicating similar contributions from brake wear (Cheung et al., 2012b).

2.5.2.2 Seasonal Variations

2.5.2.2.1 $PM_{2.5}$

Observations described in Section 2.5.2.1.1 indicated that national average $PM_{2.5}$ concentrations and 98th percentile concentrations from 2013−2015 were both higher in winter than in summer (Table 2-4), and observations described in Section 2.5.2.1.1 indicated that monthly average $PM_{2.5}$ concentrations exhibited distinct summer and winter peaks superimposed on a steadily declining national average $PM_{2.5}$ (Figure 2-22). Averaged over all locations and years from 2001−2016, seasonal average $PM_{2.5}$ concentrations were approximately 12 $\mu g/m^3$ in summer and winter, but declined to approximately 9 $\mu g/m^3$ in the spring and fall (see Figure 2-28).

While monthly average $PM_{2.5}$ concentrations are higher in summer than in winter from 2002−2008, this pattern is reversed from 2009−2015, when monthly average $PM_{2.5}$ concentrations become higher in winter than in summer (see Section 2.5.2.1.1, Figure 2-22). This is a major departure

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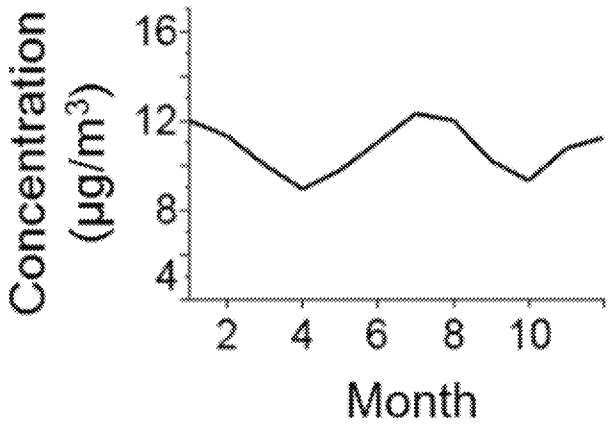
from previous concentration trends. Observations that the highest seasonal average concentrations occurred in summer in the Eastern U.S. and in winter in the Western U.S. with a few exceptions was already clearly established from 1999–2001 data from the newly operational $PM_{2.5}$ network (U.S. EPA, 2004). These early $PM_{2.5}$ network results were in turn consistent with previous studies carried out prior to its implementation, and were confirmed in the 2009 PM ISA (U.S. EPA, 2009). The observed reduction in summer $PM_{2.5}$ concentrations in the East to the extent that summer is no longer the season with the highest national average $PM_{2.5}$ concentrations is a major development, and is a predictable consequence of successful reduction of SO_2 emissions.



Source Permission pending: Chan et al. (2018).

Figure 2-28 National average $PM_{2.5}$ concentration by month 2000–2015.

2.5.2.2.2 $PM_{10-2.5}$

Relatively little had been published on the seasonal variability in $PM_{10-2.5}$ concentrations at the time of the 2009 PM ISA (U.S. EPA, 2009). Figure 2-29 shows three U.S. regions used for comparison of $PM_{10-2.5}$: the U.S. East of the Mississippi and the Northern and Southern portions of the U.S. West of the Mississippi. The regions were divided in this way because previous discussions based on limited data had suggested that PM_{10} was mostly $PM_{2.5}$ in the eastern U.S. and mostly $PM_{10-2.5}$ in the western U.S. (U.S. EPA, 2009, 2004), and these two regions were compared to investigate whether there were also seasonal differences between East and West. However, because results indicated that geographic differences within the western U.S. were greater than observed East-West differences, the western U.S. was further divided into northern and southern portions.

Figure 2-30 shows average concentrations on each day for 4 years from 2011–2014 by region based on data from the IMPROVE network, after dividing the U.S. into these three regions. All regions

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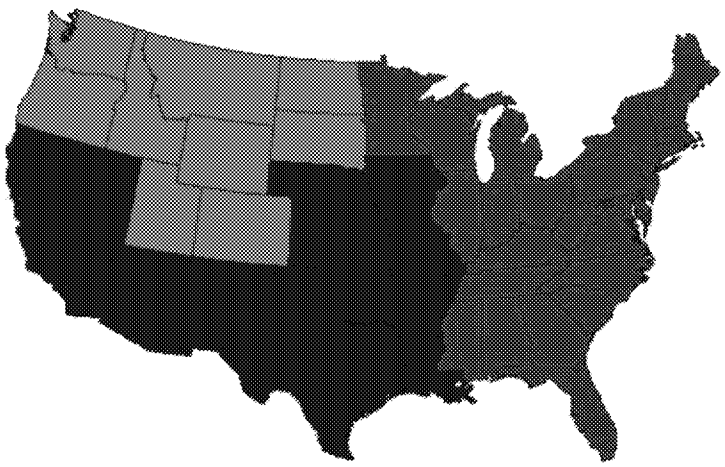
display clear seasonal variations, with the lowest concentrations occurring around January and the highest occurring in the summer months. The highest $PM_{10-2.5}$ concentrations are observed in the Southwest/Central region. Concentrations in this region are much higher than concentrations in the East and a seasonal pattern of high summer and low winter concentrations is apparent. By contrast, average concentrations in the Northwest region stretching all the way from the Pacific to the Dakotas were more similar to those in the East, but with a more pronounced seasonal pattern than either the East or the Southwest. These observations indicate that geographic patterns of $PM_{10-2.5}$ concentrations are more complicated than a simple East-West split, but that there are large areas of the Western U.S. where average $PM_{10-2.5}$ concentrations are similar to the Eastern U.S.



Source Permission pending: U.S. EPA analysis of Air Quality System network data 2011–2015.

Figure 2-29 Regions used for coarse PM comparison.

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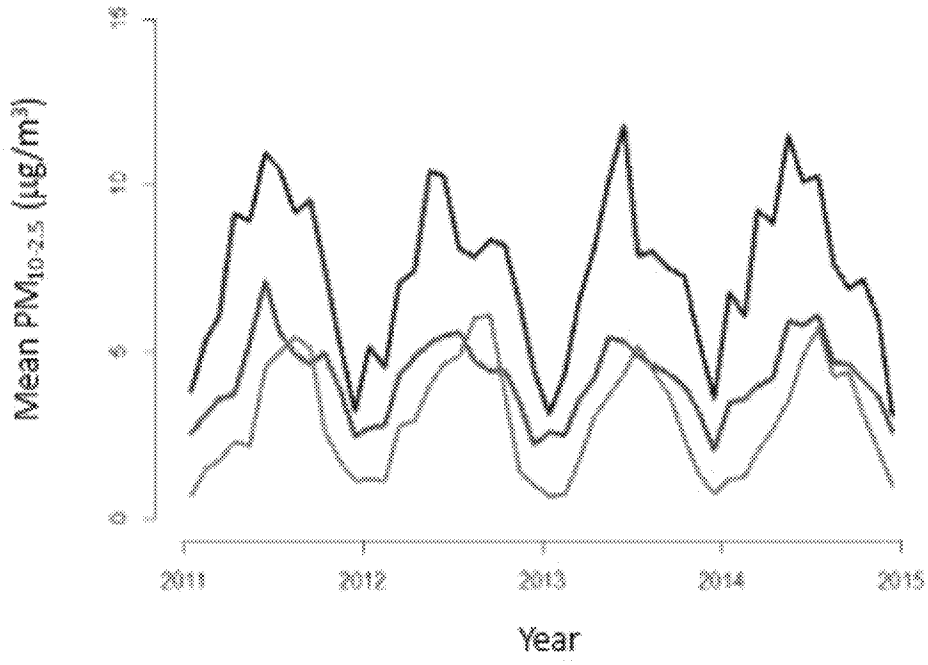

Colors of the lines correspond to the colors of the regions in Figure 2-29, i.e., red is East, green is Northwest, and blue is Southwest/Central.

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2011–2015.

Figure 2-30 Average daily $PM_{10-2.5}$ concentrations over the 4-year period 2011–2014 collected by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network.

The seasonal differences described in Section 2.5.1.1.3 of highest $PM_{10-2.5}$ concentrations in Spring and Fall and lowest concentration in winter (see Table 2-6) are consistent with other recent observations. In Colorado the highest $PM_{10-2.5}$ concentrations were observed in the Spring and Fall (Clements et al., 2014b). The monsoon period in this region is characterized by high wind events that increase $PM_{10-2.5}$ concentrations due to local wind driven soil, especially at rural sites with agricultural activity (Clements et al., 2014b). In Los Angeles $PM_{10-2.5}$ concentrations were 2–4 times higher in summer than in winter (Pakbin et al., 2010). However, organic coarse PM in Southern California was higher in winter than summer, and mostly was due to soil or biota, especially in "semirural" areas like Riverside and Lancaster (Cheung et al., 2012b).

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2.5.2.2.3 Ultrafine Particles

Relatively little has been published about seasonal or hourly differences in UFP concentrations except for localized studies in a few locations suggesting higher concentrations in winter than summer and an inverse relationship between UFP number and temperature (U.S. EPA, 2009). High afternoon concentrations during warmer months were attributed to NPF and high winter and evening UFP concentrations were attributed to lower mixing heights (U.S. EPA, 2009). More recent results indicate urban episodes of high UFP concentrations occur more often in winter than in summer (NYDEC, 2016).

2.5.2.2.4 PM Components

PM composition varies considerably with season. Figure 2-31 shows these changes. Seasonal concentration patterns are for the most part similar to those reported in the 2009 PM ISA (U.S. EPA, 2009) and conclusions from recent analyses of network data (Hand et al., 2013; Hand et al., 2012c) are consistent with patterns that can be observed in Figure 2-31. Sulfate and OC together accounted for the majority of $PM_{2.5}$ mass in many metropolitan areas in the summer, while higher nitrate concentrations were observed in the winter (U.S. EPA, 2009). Urban and rural seasonal variations of ammonium sulfate were similar, and both urban and rural concentrations were substantially higher in the East (Hand et al., 2012c). High winter nitrate concentrations were common in both urban and rural areas, but higher in urban areas (Hand et al., 2012c). Fine soil concentrations, highest in the Southwest, also had similar seasonal patterns for urban and rural sites (Hand et al., 2012c).

The higher OC contributions in fall and winter in the West compared to lower OC concentrations in winter in the Southeast reported in the 2009 PM ISA (U.S. EPA, 2009) are evident in Figure 2-31. EC mass concentration exhibited smaller seasonal variability than OC, particularly in the eastern half of the U.S. Carbonaceous aerosols varied more with season in the West than in the East for both urban and rural sites, although the seasonal patterns were different between Western urban and rural sites (Hand et al., 2013). PBAP often contributes more to PM mass in spring and summer than in fall and winter (U.S. EPA, 2009).

The metals Cu, Fe, Se, Pb, V, and Ni showed less seasonal variability than the sulfate, nitrate, and OC as reported in the 2009 PM ISA (U.S. EPA, 2009). More recently, in Los Angeles, trace element concentrations were higher in drier months of September and October, compared to December and January (Na and Cocker, 2009).

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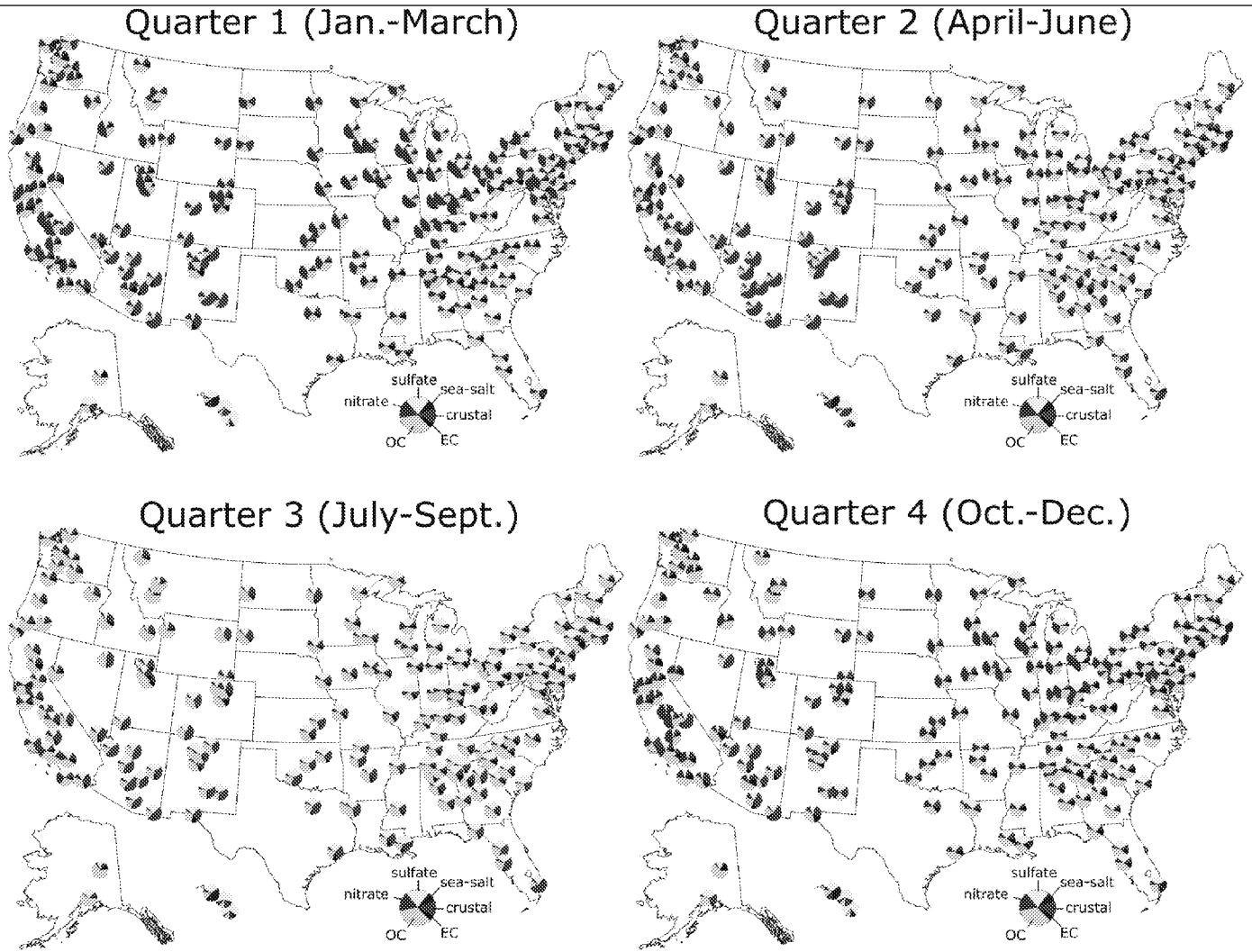

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2013−2015.

Figure 2-31 Ambient $PM_{2.5}$ seasonal composition 2013−2015.

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2.5.2.3 Hourly and Weekday-Weekend Variability

As described in the 2009 PM ISA (U.S. EPA, 2009), hourly $PM_{2.5}$ and PM_{10} measurements are conducted at several hundred network monitoring sites. A two-peaked diel pattern was observed in diverse urban locations and attributed to rush-hour traffic for the morning peak and a combination of rush hour traffic, decreasing atmospheric dilution, and nucleation for the afternoon/evening peak (U.S. EPA, 2009). In most cities, a morning $PM_{2.5}$ peak was present starting at approximately 6:00 a.m., corresponding with the start of the morning rush hour just before the break-up of the planetary boundary layer. Figure 2-32 shows diurnal patterns for multiple cities using more recent data showing rush hour peaks in the morning and evening in most cases, which is consistent with the daily variability in $PM_{2.5}$ concentrations observed in the 2009 PM ISA (U.S. EPA, 2009).

Diurnal variations in $PM_{10-2.5}$ concentrations have also been investigated. In Los Angeles in the summer the highest concentrations of $PM_{10-2.5}$ were observed in midday and afternoon when winds were the strongest. Traffic was responsible for significant resuspension especially during winter nights when mixing heights were lowest at near-freeway sites in urban areas of Southern California (Cheung et al., 2012b).

As described in Section 2.5.1.1.5 (Figure 2-18), for UFP a diel maximum was observed on average during evening hours in diverse geographic locations. An inverse relationship between UFP number and temperature has also been observed, and high afternoon concentrations during warmer months were attributed to photochemical formation and high winter and evening UFP concentrations were attributed to lower mixing heights (U.S. EPA, 2009). Relatively little had been published about hourly differences in UFP concentrations at the time of the 2009 PM ISA except for localized studies in a few locations indicating a diel maximum during evening hours (U.S. EPA, 2009).

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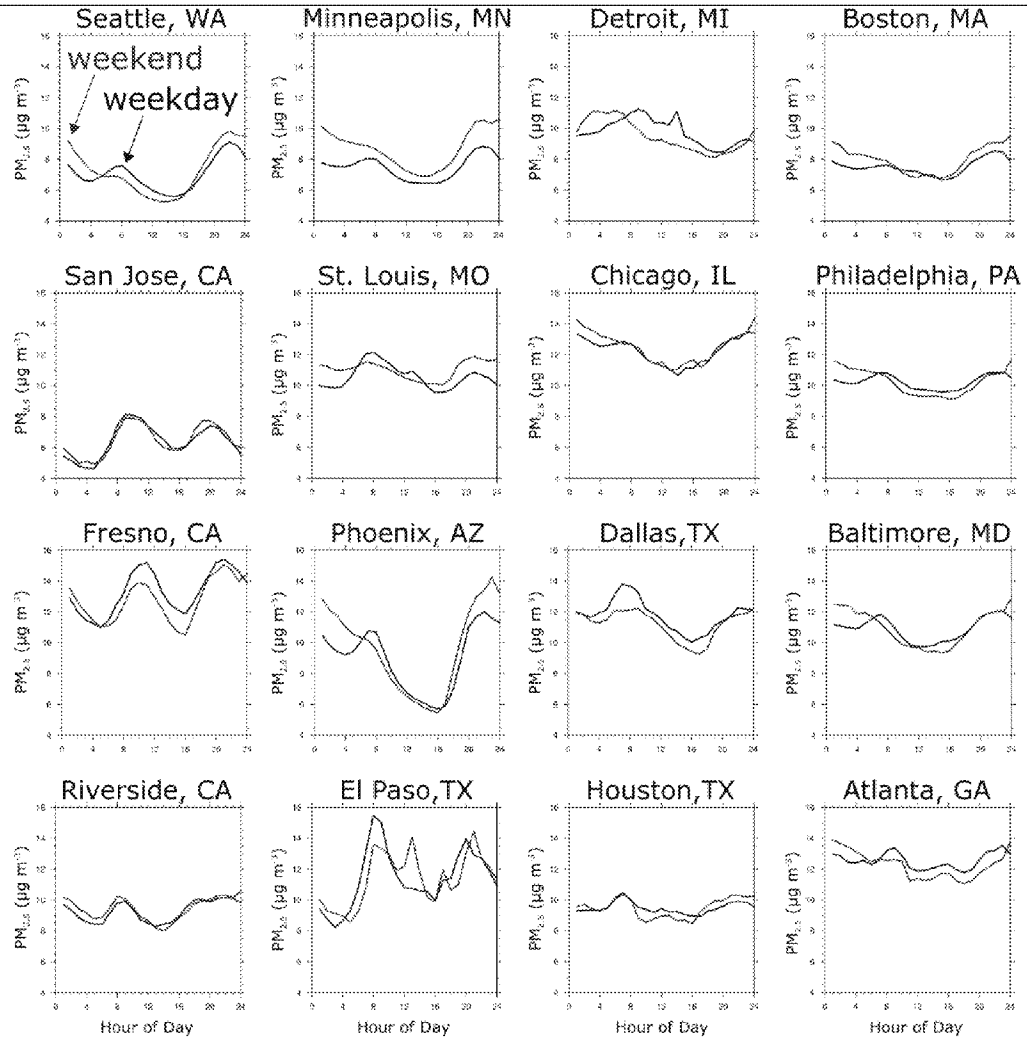

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2012−2015.

Figure 2-32 Diurnal variation of $PM_{2.5}$ concentrations in urban areas

2.5.3 Common Patterns of Particulate Matter Characteristics in the U.S.

In this section the information on sources, particle size distribution and composition from recent research results and monitoring data are combined to describe common patterns of PM characteristics observed in the U.S. across different regional and seasonal conditions. Historically, $PM_{2.5}$ has been highest in the summer and has been largely accounted for by sulfate over a large area that encompasses most of the Eastern U.S., extending into the Great Plains. Figure 2-33 shows how sulfate concentrations have changed in major urban areas of the Eastern U.S. between 2003−2005 and 2013−2015 based on CSN monitors. At all of the locations shown in Figure 2-33 sulfate was the most abundant component measured for the period 2003−2005, accounting for close to half of the overall average $PM_{2.5}$ mass. In contrast, during the period 2013−2015 sulfate accounted for only about a quarter or a third of $PM_{2.5}$ mass.

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For example, the sulfate fraction dropped from 49 to 31% in Washington DC, 51 to 34% in Pittsburgh, 42 to 24% in New York, 43 to 26% in Philadelphia, 44 to 27% in Boston, and 52 to 33% in Cincinnati. In all but five of these locations, mostly in Ohio or the Ohio Valley (Cleveland, Cincinnati, and Dayton, OH, Louisville, KY, Dallas, TX), OC has replaced sulfate as the most abundant component, although OC and sulfate concentrations are very similar in most locations, as shown in Figure 2-31.

In the Eastern half of the U.S., the steep decline in sulfate concentrations has led to major changes in PM composition, seasonal concentration patterns, and size characteristics since publication of the 2009 PM ISA (U.S. EPA, 2009). PM_{10} concentrations in the Eastern U.S. and Midwest previously peaked in summer and was mostly composed of $PM_{2.5}$, with sulfate as the largest single component. More recently, summer concentrations are similar to other seasons, the $PM_{10-2.5}$ and $PM_{2.5}$ fractions are often comparable, and OC is frequently the most abundant single component.

Some finer scale trends within the Eastern U.S. are evident. While OC is becoming the component with the highest concentration throughout the Eastern U.S., in the Southeast annual average OC concentrations are somewhat higher than in the Northeast or Midwest, reaching their highest monitoring concentrations in a large area encompassing most of Alabama, Georgia, and South Carolina (Hand et al., 2011). The origin of summer OC in the Southeast has been intensively studied and is largely SOA due to oxidation of biogenic precursors (Marais et al., 2017; Rattanavaraha et al., 2016; Lewandowski et al., 2013), and urban areas of the Southeast like Atlanta have considerably more biogenic VOC precursors than urban areas of the Northeastern U.S. like New York City (Weber et al., 2007). Integrated modeling and measurement results (Kim et al., 2015), modeling predictions (Marais et al., 2017; Ying et al., 2015), and product concentration measurements (Lewandowski et al., 2013) are also consistent with higher OC concentrations and biogenic SOA at Southeastern sites than in the Northeast or Midwest. OC concentrations in the Southeast are decreasing (Marais et al., 2017).

Another area in the Eastern half of the U.S. stretching from Minnesota and Iowa through Wisconsin, Michigan, Indiana, and Ohio comprises as a region susceptible to high winter nitrate episodes resulting from high emissions of ammonia from animal agriculture combining with atmospheric nitric acid, that lead to mean winter ammonium nitrate concentrations exceeding 4 $\mu g/m^3$ (Pitchford et al., 2009). This region can be distinguished in Figure 2-31 for 2012–2014 by winter nitrate contributions of more than 40% to seasonal average $PM_{2.5}$ mass in Chicago, IL, Minneapolis, MN, Milwaukee, WI, Detroit and Grand Rapids, MI, Indianapolis, IN, Cincinnati and Dayton, OH, Davenport and Des Moines, IA, Omaha, NE, Kansas City, MO and at several other sites in the upper Midwest.

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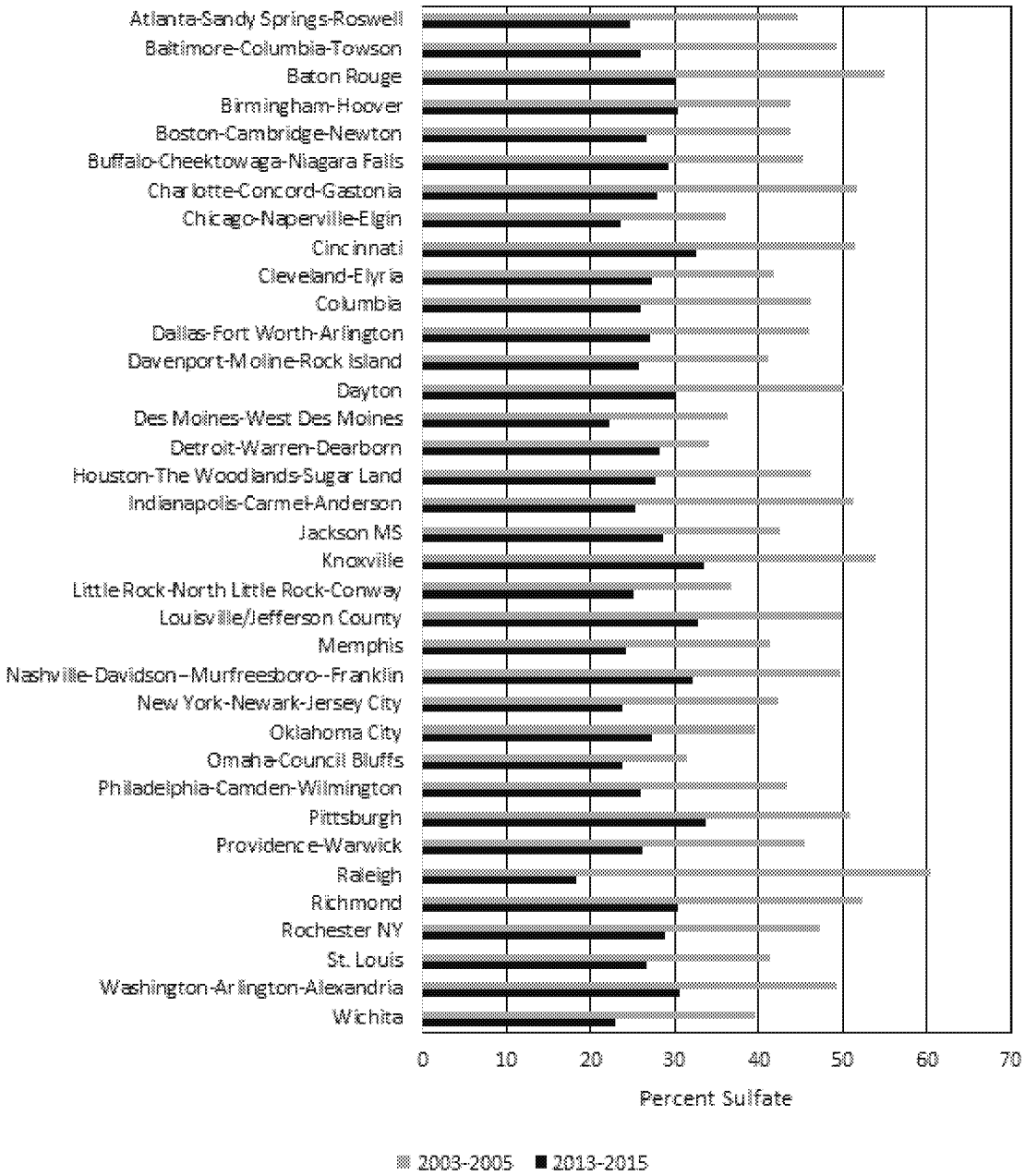

Source Permission pending: U.S. EPA 2016 analysis of Air Quality System network data 2003−2005 and 2013−2015.

Figure 2-33 Sulfate as percentage of $PM_{2.5}$ in eastern urban areas 2003−2005 and 2013−2015.

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While substantial differences in PM size distribution, composition, and other characteristics have been reported between the Eastern and Western U.S. (U.S. EPA, 2009), the diversity of PM characteristics across the West makes it more difficult to identify a set of fundamental PM characteristics that applies to the entire region. In interior urban areas, including Salt Lake City, UT, Reno, NV, Boise, ID, Missoula, MT, and Spokane, WA, $PM_{2.5}$ levels are higher under stable conditions on days with snow cover. In Salt Lake City, UT, Reno, NV, and Missoula, MT, most of the highest concentrations were observed on days with high nitrate concentrations enhanced by colder temperatures and higher relative humidity that occur with snow cover (Green et al., 2015). After multiday periods with stable conditions created by snow cover, $PM_{2.5}$ can build up rapidly in layers or in cold air pools. In one case in Salt Lake City $PM_{2.5}$ concentrations increased by 6 to 10 $\mu g/m^3$ per day over a period of several days (Whiteman et al., 2014; Silcox et al., 2011). This area is also subject to episodically high $PM_{10-2.5}$ concentrations from dust suspension.

Closer to the coast, high PM episodes cannot be explained by snow cover and extreme cold, yet some of the highest $PM_{2.5}$ concentrations in Figure 2-13 and Figure 2-14 are in California and concentrations are also highest in winter. In many California locations, a specific combination of conditions appears to be responsible for the highest PM concentrations. High winter $PM_{2.5}$ concentrations were studied intensively over 12 winters and the existence of several simultaneous conditions for at least 2 days duration were required for concentrations to exceed 35 $\mu g/m^3$, including a ridge of high pressure aloft, persistent easterly flow extending up vertically, orographically channeled winds resulting from stability, and enhanced nocturnal cooling under clear sky conditions (Beaver et al., 2010). Ammonium nitrate and organic PM from diverse combustion sources are the main contributors to $PM_{2.5}$ under winter conditions in California (Young et al., 2016; Zhang et al., 2016; Schiferl et al., 2014). Some of the highest 98th percentile concentrations were reported in California and other monitoring sites in the Western U.S. in Section 2.5.1.1.1 (Figure 2-14).

A common characteristic of PM in both California and the dryer areas of the Western U.S. that contrasts with the Eastern U.S. is the higher fraction of PM_{10} accounted for by $PM_{10-2.5}$, with $PM_{10-2.5}$ accounting for most PM_{10} mass in the West, but $PM_{2.5}$ accounting for most PM_{10} mass in the East (see Table 2-7). Populated areas of the Northwest (Western Oregon and Washington) make an exception to this trend. Table 2-7 shows that in both Seattle, WA and Portland, OR, $PM_{2.5}$ accounts for more than 50% of the PM_{10} mass and concentrations are higher in winter than in summer. Wood smoke is a major source of $PM_{2.5}$ in Portland, OR and Seattle, WA (Kotchenruther, 2016; U.S. EPA, 2009), as well as in smaller urban areas in this region.

$PM_{2.5}$ concentrations averaged over the 11-year period from 1998–2008 over the entire contiguous U.S. were reported to be 2.6 $\mu g/m^3$ higher on days under stagnant conditions than for non-stagnant days (Tai et al., 2010). When all U.S. data over a multiyear period are considered, temperature is positively correlated with $PM_{2.5}$ (Tai et al., 2012a; Tai et al., 2012b), especially in the Eastern U.S. (Tai et al., 2012a). Much of $PM_{2.5}$ variability could be explained by cold frontal passages in the East, maritime

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inflow in the West, and cyclone frequency in the Midwest (Tai et al., 2012b). Other meteorological conditions that have been reported to enhance PM concentrations include sea breezes (Georgoulias et al., 2009) and drought (Wang et al., 2015).

2.5.4 Background Particulate Matter

The definition of background PM can vary depending upon context, but it generally refers to PM that is formed by sources or processes that cannot be influenced by actions within the jurisdiction of concern. Consistent with other recent NAAQS reviews (U.S. EPA, 2014); U.S. EPA, 2015, 4679035}, there are two specific definitions of background PM of interest: natural background and U.S. background. Natural background is the narrowest definition of background, and it is defined as the PM that would exist in the absence of any manmade emissions of PM or PM precursors. U.S. background PM is defined as any PM formed from sources or processes other than U.S. manmade emissions. Approaches to estimating background PM have evolved over the years. Different approaches for estimating background concentrations in the western and eastern U.S. were taken in the 2004 PM AQCD (U.S. EPA, 2004). Data from IMPROVE monitoring sites in the western U.S. thought to be among the least influenced by regional pollution sources exhibited annual mean concentrations of ~3 $\mu g/m^3$. However, even the most remote monitors within the U.S. can be periodically affected by U.S. anthropogenic emissions, and concentrations observed at the most remote sites in the Eastern U.S. were considerably higher than in the western U.S. In the 2009 ISA (U.S. EPA, 2009), estimates of background concentrations were calculated by CMAQ and classified by region and quarter. All quarterly and annual estimates were less than 2 $\mu g/m^3$, with many <1 $\mu g/m^3$. However, episodic contributions from dust storms or wildfires can be much higher. Further details are given by (U.S. EPA, 2009).

As illustrated by this example, background PM concentrations can be best characterized with chemical transport modeling simulations via source apportionment modeling or estimating what the residual PM concentrations would be were the U.S. anthropogenic emissions entirely removed (i.e., "zero-out" modeling). Unfortunately, there has not been a similar national scale effort to update background $PM_{2.5}$ concentration estimates since the 2009 PM ISA. However, there has been considerable research focused on better understanding the sources and processes that influence background contribution to $PM_{2.5}$ in the U.S.

Background contributions to PM can come from a variety of sources. Natural sources include wind erosion of natural surfaces, volcanic production of SO_4^{2-}; primary biological aerosol particles (PBAP); wildfires producing EC, OC, and inorganic and organic PM precursors; and SOA produced by oxidation of biogenic hydrocarbons such as isoprene and terpenes (U.S. EPA, 2009). However, human intervention can be involved in the formation of SOA. For example, the production of SOA from the oxidation products of isoprene and other biogenic VOC's can be enhanced by the presence of SO_2, NO_X, and other anthropogenic pollutants, accounting for as much 50% of SOA from biogenic VOC's

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(Section 2.3.2.3). Other sources of background PM are anthropogenic, principally emissions from outside the U.S. which can be transported into the U.S. The importance of different contributors to background PM varies across the contiguous U.S. (CONUS) by region and season as a function of the complex mechanisms of transport, dispersion, deposition, and re-entrainment.

Background PM can also be viewed as coming from two conceptually separate components: a somewhat consistent "baseline" component and an episodic component. The baseline component consists of contributions that are generally well characterized by a reasonably consistent distribution of daily values each year, although there is variability by region and season. The episodic component consists of infrequent, sporadic contributions from high-concentration events occurring over shorter periods of time (e.g., hours to several days) both within North America (e.g., volcanic eruptions, large forest fires, dust storms) and outside North America (e.g., transport from dust storms occurring in deserts in North Africa and China). These episodic natural events, as well as events like the uncontrolled biomass burning in Central America, are essentially uncontrollable and do not necessarily occur in all years. Section 2.5.4.1 and Section 2.5.4.2 below discuss natural background and intercontinental transport contributions to background PM in the U.S.

2.5.4.1 Natural Background

On average, natural sources including soil dust and sea salt have been estimated to account for approximately 10% of U.S. urban $PM_{2.5}$ (Karagulian et al., 2015). Dust storms are common occurrences in arid regions of the U.S. and the rest of the world. An extreme example is the haboob. During one of these affecting Phoenix in July of 2011, peak hourly average PM_{10} concentrations were >5,000 $\mu g/m^3$ with area wide average hourly concentrations ranging from a few hundred to a few thousand $\mu g/m^3$ (Vukovic et al., 2014). Dust can also make up a substantial fraction of total $PM_{2.5}$ in the Southwestern U.S. This is illustrated in Figure 2-19 (Section 2.5.1.1.6), which shows that at many locations in the Southwestern U.S., crustal material from soil accounts for close to half of the annual average $PM_{2.5}$ mass. Although similar network data do not exist for $PM_{10-2.5}$, the soil contribution to $PM_{10-2.5}$ mass in these locations is likely to be even higher. Dust also accounts for much of the PM that originates from outside the U.S. (Section 2.5.4.2).

Wildfires are a variable contributor to particulate matter emissions. Satellite-based fire detections are combined with ground-based estimates of area burned, fuel availability, and emission factors to quantify PM and precursor emissions at high spatial and temporal resolution (Strand et al., 2012). The gas-phase species emitted from fires can affect oxidation and formation of semivolatile compounds that can condense into the particle phase (Baker et al., 2016). Invasive species, historical fire management practices, frequency of drought, and extreme heat have brought longer fire seasons (Jolly et al., 2015) and more large fires (Dennison et al., 2014). In addition to emissions from forest fires in the U.S., emissions from forest fires in other countries can be transported to the U.S., and transport from Canada, Mexico,

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Central America, and Siberia have been documented (U.S. EPA, 2009). According to the U.S. EPA's National Emission Inventory, wildfire smoke contributes between 10 and 20% of primary PM emissions per year (Section 2.3.1), however these emissions are concentrated at the burn area and mostly during the wildfire season, rather than evenly distributed through the year (Sturtz et al., 2014).

Primary biological aerosol particles (PBAP) such as bacteria and pollen can also contribute substantially to $PM_{10-2.5}$ mass in some locations. These are discussed in more detail in Section 2.3.3.

2.5.4.2 Intercontinental Transport

Intercontinental transport contributes 0.05 to 0.15 μg/m^3 to annual average $PM_{2.5}$ concentrations in the U.S. (Kolb et al., 2010). Large continuous data sets are available to examine the intensity and frequency of intercontinental PM transport events. Ground-based lidar networks and mountain top measurements in Europe, North America, and Asia have been used to establish that intercontinental transport of PM from dust, forest fires, and anthropogenic sources impact local $PM_{2.5}$ and PM_{10} concentrations. Satellites also provide estimates of the amount of PM transported, as well as the altitude at which the transport occurs. Transport at midlatitudes is dominated by westerly winds, which transport East Asian emissions across the North Pacific Ocean to North America. Transport occurs at greater speeds and over longer distances in winter than in summer because the westerly winds are stronger, and greater precipitation in winter in the Western U.S. brings more of the transported PM to the surface. Numerous studies have now documented long-range transport of desert dust from East Asian deserts. Both the frequency of transport events and the overall contribution to PM in the U.S. are reported to be increasing (Kolb et al., 2010; TFHTAP, 2006). By one estimate, 18 Tg/year PM exits Asia between 30 to 60 degrees N latitude, with 4.4 Tg/yr arriving in North America (Yu et al., 2008).

Episodic concentrations as high as 20 μg/m^3 of PM associated with transport to the U.S. from Asia have been estimated (Jaffe et al., 2005), and $PM_{2.5}$ from Asia has been shown to account for a large fraction total $PM_{2.5}$ in polluted urban air (Jaffe et al., 2003). Over longer time periods, long range transport can make a substantial contribution to local PM concentrations in remote areas like the Arctic. However, in regions with local sources, observed trends in PM are usually more closely related to local emission trends than to long-range transport, and at monitoring sites throughout the U.S. intercontinental influences are small (Henze et al., 2009).

On average, Asian dust contributes typically <~1 μg/m^3 to $PM_{2.5}$ at remote sites in western states (Creamean et al., 2014). However, transport of Asian dust shows both strong seasonal and interannual variability. Dust emissions are at a maximum in spring, associated with strong winds following cold fronts as the Siberian High extends southward and before there is sufficient vegetation to stabilize the surface. Based on inverse modeling of Asian dust over the period 2005–2012, Yumimoto and Takemura (2015) suggested that dust emissions, transport and deposition are largest during the La Niña phase of the El Niño-Southern Oscillation cycle. They also found that dust emissions were closely related to a strong

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meridional pressure gradient and a strong winter monsoon. Husar et al. (2001) report that the average PM_{10} concentration at 25 reporting stations throughout the northwestern U.S. reached 65 μg/m^3 during an episode of Asian dust transport during the last week of April 1998, compared to an average of 10–25 μg/m^3 during the rest of April and May. This was accompanied by visual reports of milky-white discoloration of the normally blue sky in nonurban areas along the West Coast. Satellite data have been especially useful for tracking the trans-Pacific transport of Asian dust. Uno et al. (2011) documented the occurrence of multiple large plumes of Asian dust in April of 2010 that had passed over most of the continental U.S. based on space-borne lidar (the Cloud-Aerosol Lidar with orthogonal Polarization) on board the CALIPSO satellite. Three-dimensional, global-scale CTMs have also been used to estimate intercontinental transport of PM pollution (TFHTAP, 2007) and trans-Pacific transport of mineral dust from Asian deserts (Fairlie et al., 2007).

Transport of dust from the Sahara Desert and the Sahel in North Africa (Prospero, 1999a, b), (Chiapello et al., 2005), (Mckendry et al., 2007) can affect the eastern U.S., while transport of dust from the Gobi and Taklimikan deserts in Asia (Vancuren and Cahill, 2002), (Yu et al., 2008) can exert effects in the western U.S. The ability of African dust to substantively affect PM levels in the U.S. was extensively reviewed in the 2004 PM AQCD (U.S. EPA, 2004) and in the 2009 PM ISA (U.S. EPA, 2009). A multidecade record of African dust reaching Miami indicates that the highest loadings are found in July (Prospero, 1999a, b) with concentrations ranging from ~10 to ~100 μg/m^3. Sample collection began in 1974, before network PM_{10} and $PM_{2.5}$ samplers were developed, and no size cut was specified (Prospero, 1999b). Yu et al. (2015) found that the transport of North African dust across the Atlantic Ocean is strongly negatively correlated with precipitation in the Sahel during preceding year. Dust from Africa has shown a decreasing trend of ~ 10% per decade from 1982 to 2008, based on measurements of aerosol optical depth and surface concentrations in Barbados by Ridley et al. (2014), who also suggest that this decrease is due to a corresponding decrease in surface winds over source regions.

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In addition to desert dust, a portion of the PM reaching the U.S. through intercontinental transport is from combustion and industrial sources, and formation of sulfate from SO_2 during transport of air masses to the U.S. from Asia is also well documented. In the Spring in the Northwestern U.S., transport from Asia accounted for 0.16 ± 0.08 μg/m^3 $PM_{2.5}$ sulfate (Heald et al., 2006). Sulfate of Asian origin can account for a large fraction of sulfate in the upper troposphere in western North America, and an increasing fraction of sulfate measured off the northwest coast of the U.S. is of Asian origin. Measurements from an event over the Pacific Ocean were consistent with nearly pure sulfuric acid. Transboundary transport within North America can also be important. Model results suggest that SO_2 emissions in Mexico influence sulfate formation in the U.S. (Henze et al., 2009). Leibensperger et al. (2011) estimated that trans-Pacific transport of SO_2 and NO_X results in a combined increase in background PM[40] in the western U.S. of a few tenths of a μg/m^3.

2.6 Summary

New observations indicate that some fundamental characteristics of atmospheric PM in the U.S. are changing. These range from source emissions and atmospheric formation processes to size distributions, particle composition, and spatial and temporal concentration trends. The most noticeable change in PM or precursor source emissions is the large reduction in SO_2 emissions, mainly from decreased EGU coal combustion. In addition, advances in engine and emissions control technologies have led to continued decreases in automobile emissions. The major urban stationary sources of PM are still industrial processes, construction and road dust, residential wood burning and other fuel combustion, and cooking. The major primary mobile sources are still diesel and gasoline powered highway vehicles as well as off-road vehicles and engines like locomotives, ships, aircraft, and construction and agricultural equipment. $PM_{2.5}$ particles from combustion sources are usually emitted as UFP and grow into larger particles after emission. Secondary $PM_{2.5}$ still accounts for a substantial fraction of the $PM_{2.5}$ mass from both natural and anthropogenic sources (U.S. EPA, 2009). Major $PM_{10-2.5}$ sources are dust suspension, sea spray, and biological materials. Automobile traffic, other combustion sources, and new particle formation are major UFP sources.

Research on atmospheric chemistry has largely focused on better understanding OC sources and SOA formation pathways. Progress in understanding SOA precursors centered on model results of large fractions of SOA from aromatic and monoterpene precursors, observations of gas phase VOC oxidation products continuing to react to form PM, and the discovery of isoprene as a major SOA precursor. Progress related to understanding SOA formation processes was directed toward evidence of cloud processing as well as repeated cycles of volatilization and condensation of semivolatile reaction products as important processes for SOA evolution, investigation of misclassification of SOA as primary organic

[40] PM size was not specified, but secondary PM formed from NO_X and SO_2 is usually nearly all in the $PM_{2.5}$ size range.

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aerosol under typical sampling conditions, and observations of greater SOA yields at high NO_X concentrations. Progress in understanding SOA products involved identification of higher molecular weight particle phase oligomers and organic peroxides as an abundant class of reactive oxygen species (ROS) with high oxidizing potential in SOA, as well as observations of abundant organosulfates and organonitrates in SOA.

Major developments in PM monitoring and monitoring capabilities have taken place, and these have had an important impact on our understanding of PM characteristics. For example, before the availability of network data, the 2009 PM ISA was based on literature results and concluded that $PM_{10-2.5}$ concentrations were higher in the Western U.S. than in the Eastern U.S. (U.S. EPA, 2009). The NCore network was implemented in 2011 and now produces multipollutant concentration and data at 78 stations throughout the U.S. Through NCore, more reliable data on $PM_{10-2.5}$ concentrations are available than were possible before. The first years of NCore data reveal a more complicated concentration pattern than a simple East-West split, with the highest $PM_{10-2.5}$ concentrations observed in the Southwest from California to Texas, and in the Central U.S. from Texas and Louisiana as far north as Nebraska and Iowa. In contrast, there are large areas in the Northwest where average $PM_{10-2.5}$ concentrations and $PM_{2.5}/PM_{10}$ are similar to the Eastern U.S. Rapid advances are taking place in UFP measurement technology, but measurements are more method dependent and network monitoring is in its beginning stages. Network monitoring of $PM_{2.5}$ has expanded to include numerous near road monitoring sites.

Annual mean ambient $PM_{2.5}$ concentrations in the U.S. on average are 4–5 $\mu g/m^3$ lower than they were in the last decade, continuing a downward trend described in the 2009 PM ISA (U.S. EPA, 2009). $PM_{2.5}$ concentrations are highest in the San Joaquin Valley, the Los Angeles-South Coast Air Basin of California, and parts of Utah. In the Eastern U.S. there is a region of higher $PM_{2.5}$ concentrations with annual average concentrations greater than 10 $\mu g/m^3$ stretching from Eastern Iowa and Northern Illinois across Indiana, Ohio, and into to Eastern Pennsylvania. While monthly national average $PM_{2.5}$ concentrations were higher in summer than in winter from 2002–2008, this pattern is reversed from 2012–2015, when monthly average $PM_{2.5}$ concentrations become higher in winter than in summer. Summer $PM_{10-2.5}$ concentrations are generally higher than other seasons, but extreme $PM_{10-2.5}$ events appear to be more likely in the spring. PM_{10} reflects characteristic concentration patterns of both $PM_{10-2.5}$ and $PM_{2.5}$, with the highest concentrations in summer. The decrease in $PM_{2.5}$ concentrations has resulted in smaller $PM_{2.5}/PM_{10}$ ratios, and PM_{10} in the East and Northwest is in the range of 50–60% $PM_{2.5}$, while PM_{10} in the Western U.S. is generally less than 50% $PM_{2.5}$. On urban and neighborhood scales, both spatial and temporal variations are strongly influenced by motor vehicle emissions, with the highest $PM_{2.5}$ and UFP concentrations at rush hour, and the highest concentrations of $PM_{10-2.5}$, UFP, and many $PM_{2.5}$ components near roads with heavy traffic.

Recent changes in $PM_{2.5}$ long-term and seasonal concentration trends are consistent with observed changes in $PM_{2.5}$ composition compared to the 2009 PM ISA (U.S. EPA, 2009), the greatest of which is the reduction in sulfate concentrations, resulting in a smaller sulfate contribution to $PM_{2.5}$ mass

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in 2013–2015 than in the last decade, especially in the Eastern U.S. As a result, at many locations sulfate has been replaced as the greatest single contributor to $PM_{2.5}$ mass by organic matter. Sulfate and OC are the components with the highest contribution to total mass in most eastern locations and OC usually makes the greatest contribution to $PM_{2.5}$ mass in the west, although sulfate, nitrate, and crustal material can also be abundant. The highest nitrate concentrations are found in the west, particularly in California, but with some elevated concentrations in the upper Midwest. Ammonium concentrations follow both nitrate and sulfate spatial patterns because it is mostly present as ammonium sulfate and ammonium nitrate. Larger contributions of OC to $PM_{2.5}$ mass are observed in the Southeast and the West than in the Central and Northeastern U.S. A large fraction of organic PM can be water soluble. Crustal elements and biological material account for large fraction of $PM_{10-2.5}$ mass. There is still little information on the composition of UFP, but urban UFP is often rich in OC and EC.

Background PM originates from natural and international sources. Natural sources include windblown dust, wildfires, and sea salt. International contributions include intercontinental transport of dust, wildfire smoke, and pollution as well as transboundary transport of these contributors from Canada, Mexico. Background PM usually makes a relatively small contribution to urban annual average $PM_{2.5}$ concentrations. However, it is an important contributor to $PM_{2.5}$ concentrations in the southwestern U.S., and impacts $PM_{2.5}$ concentrations elsewhere on an episodic basis. Background contributions to $PM_{10-2.5}$ can be substantial, as it is generally dominated by dust and sea salt. Less is known about background contributions to UFP.

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CHAPTER 3 EXPOSURE TO AMBIENT PARTICULATE MATTER

Overall Conclusions regarding Exposure to Ambient PM

- Recent and existing evidence indicate that exposure error typically produces *underestimation* of health effects in epidemiologic studies of short-term and long-term PM exposure. Bias away from the null can sometimes occur for long-term exposure studies if a monitor or model underestimates population exposure.
- New developments in PM exposure assessment methods, including hybrid spatiotemporal models that incorporate satellite observations of AOD, land use variables, surface monitoring data from FRMs, and/or CTMs, have reduced bias and uncertainty in health effect estimates by improving the spatial resolution and accuracy of exposure predictions.
- High correlations of $PM_{2.5}$ with some gaseous copollutants necessitate evaluation of the impact of confounding on health effect estimates.
- There is typically more uncertainty for health effect estimates for exposure to $PM_{10-2.5}$ and UFP, because their concentrations tend to be more spatially variable than $PM_{2.5}$ concentrations and concentration data for $PM_{10-2.5}$ and UFP are less frequently available and/or more uncertain.

3.1 Introduction

Assessment of exposure to ambient PM builds from the characterization of concentrations and atmospheric chemistry presented in CHAPTER 2. The primary conclusions from CHAPTER 2 were that $PM_{2.5}$ concentrations continue to decrease over time with few areas exceeding the level of the current NAAQS, sulfates comprise a smaller proportion of total $PM_{2.5}$ throughout the country including in the eastern half of the country, $PM_{10-2.5}$ contributes most substantially to PM_{10} in the southwestern U.S. but is highly variable across urban areas, and substantial uncertainty still exists regarding UFP sources, composition, and concentrations.

This chapter presents new developments in exposure assessment methodology and interpretation of epidemiological study results given strengths and limitations of the exposure assessment data. The chapter describes concepts and terminology relating to exposure (Section 3.2), methodological considerations for use of exposure data (Section 3.3), and exposure assessment and interpretation of epidemiologic study results (Section 3.4). This chapter focuses on the ambient component of personal exposure to PM, because the NAAQS pertains to ambient PM. However, studies using total personal PM measurements or indoor PM concentrations to represent exposure can also inform the understanding of the relationship between exposure and health effects and so are included as supporting evidence if ambient PM exposure can be deduced from the information provided in the studies. This chapter focuses on studies of exposure among the general population. Exposure of groups potentially at increased risk of PM-related health effects, based for example on socioeconomic status and race, is addressed in CHAPTER 12. Intake of PM based on ventilation rate, and in relation to physical activity, is described in CHAPTER 4. The information provided in this chapter will be used to help interpret the evidence for the

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health effects of PM exposure presented in the health chapters that follow (CHAPTER 5, CHAPTER 6, CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11).

3.2 Conceptual Overview of Human Exposure

The 2009 PM ISA (U.S. EPA, 2009b) provided a conceptual model of exposure to form a distinction between ambient PM exposure and total personal exposure. This section illustrated that exposure is integrated over time and across the microenvironments in which a person spends time. This section also introduced the concept of an infiltration factor that depends on both penetration of PM indoors and the ventilation and deposition characteristics that influence indoor PM concentration. That discussion is currently updated and presented in Section 3.2.2.

This ISA contains two new sections to orient the reader to concepts relevant to exposure. Section 3.2.1 introduces terminology that is used throughout the chapter when describing exposure assessment studies. Section 3.2.3 highlights facets of exposure assessment that are particularly relevant to PM.

3.2.1 Exposure Terminology

A variety of metrics and terms are used to characterize air pollution exposure. They are described here at the beginning of the chapter to provide clarity for the subsequent discussion.

The *concentration* of PM is defined as the mass of the pollutant in a given volume of air (e.g., $\mu g/m^3$). Concentrations observed in outdoor locations accessible to the public are referred to as ambient concentrations. The term exposure refers to contact at the interface of the breathing zone with the ambient concentration of a specific pollutant over a certain period of time (Zartarian et al., 2005), in single or multiple locations. For example, contact with a concentration of 10 $\mu g/m^3$ $PM_{2.5}$ for 1-hour would be referred to as a 1-hour exposure to 10 $\mu g/m^3$ $PM_{2.5}$, and 10 $\mu g/m^3$ is referred to as the *exposure concentration*. As discussed in CHAPTER 4, dose incorporates the concept of intake into the body (via inhalation).

A location where exposure occurs is referred to as a *microenvironment*, and an individual's daily exposure consists of the time-integrated concentrations in each of the microenvironments visited during the day. Ambient air pollution may penetrate indoors (see Section 3.4.1.1 on infiltration), where it combines with air pollution from indoor sources (*nonambient air pollution*) to produce the total measured indoor concentration. Exposure to the ambient fraction of total indoor concentration, together with exposure to ambient concentrations in outdoor microenvironments such as parks, yards, sidewalks, and bicycles or motorcycles, is referred to as ambient exposure (Wilson et al., 2000). *Total personal exposure to ambient PM* is the concentration of PM emitted from ambient sources or formed in the atmosphere that

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is encountered by an individual over a given time. This differs from overall total personal exposure, which may also include exposure to nonambient air pollution. Personal exposure to ambient PM is influenced by several factors, including:

- Time-activity in different microenvironments (e.g., vehicle, residence, workplace, outdoor);
- climate (e.g., weather, season);
- characteristics of indoor microenvironments (e.g., window openings, draftiness, air conditioning); and
- microenvironmental emission sources (e.g., roadways, construction equipment, indoor gas stoves) and concentrations.

Because personal exposures are not routinely measured, the term *exposure surrogate* is used in this chapter to describe a quantity meant to estimate or represent exposure, such as $PM_{2.5}$ concentration measured at an ambient monitor (Sarnat et al., 2000). A *fixed-site monitor* (i.e., a monitor with a fixed position) is a type of *ambient monitor* used to estimate population average exposure concentrations and their trends over neighborhood- and urban-scales for epidemiologic studies.

When surrogates are used for exposure estimation in epidemiologic studies, exposure error or exposure misclassification can result. *Exposure error* refers to the bias and uncertainty associated with using concentration metrics to represent the actual exposure of an individual or population (Lipfert and Wyzga, 1996). Exposure misclassification refers to exposure error that occurs when exposure conditions such as location, timing, or population grouping are incorrectly assigned. *Exposure misclassification* due to exposure assignment methods and spatial and temporal variability in pollutant concentrations may be either differential (i.e., systematic), or nondifferential (i.e., random). *Differential misclassification* refers to the situation where exposure errors differ between groups. An example of differential misclassification is the use of geocoding to estimate air pollution exposure by proximity to roadways, because concentrations decrease with distance from roadways and are different upwind and downwind of a major roadway (Lane et al., 2013; Singer et al., 2004). *Nondifferential misclassification* refers to the situation where exposure characterization has the same probability of being misclassified to a similar degree across all groups.

Exposure misclassification and exposure error can result in bias and reduced precision of the effect estimate in epidemiologic studies. *Bias* refers to the difference between the population-average measured and true exposure, while precision is a measure of the variation of measurement error in the population (Armstrong et al., 1992). Bias toward the null, or attenuation of the effect estimate, indicates an underestimate of the magnitude of the effect, and is characteristic of nondifferential measurement error. Bias away from the null can occur through differential exposure measurement error, such as may occur when an exposed person or group of people are located far from a source that is captured by a fixed-site monitor (Armstrong et al., 1992).

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Exposure error has two components: (1) exposure measurement error derived from uncertainty in the metric being used to represent exposure and (2) use of a surrogate parameter of interest in the epidemiologic study in lieu of the true exposure, which may be unobservable. *Classical error* is defined as error scattered around the true personal exposure and independent of the measured exposure. Classical error results in bias of the epidemiologic health effect estimate. Because variation in the measurements tends to be greater than variation in the true exposures, classical error typically biases the health effect estimate towards the null (no effect of the exposure). This would cause the health effect estimate to be underestimated. Classical error can also cause inflation or reduction of the standard error of the health effect estimate. For example, classical error may occur when a fixed-site monitor measuring exposure concentration is imprecise. *Berkson error* is defined as error scattered around the measured exposure surrogate (in most cases, the ambient monitoring measurement) and independent of the true exposure (Goldman et al., 2011; Reeves et al., 1998). Pure Berkson error is not expected to bias the health effect estimate. Berkson error tends not to cause bias in the health effect estimate. For example, Berkson error may occur when personal monitors used in a panel study capture ambient and nonambient exposures, if the objective of the study is to evaluate the effect of ambient exposures on health and the ambient and nonambient exposures are independent of each other.

Definitions for *classical-like* and *Berkson-like errors* were developed for modeled exposures. These errors depend on how exposure metrics are averaged across space. Classical-like errors can add variability to predicted exposures and can bias health effect estimates in a manner similar to pure classical errors, but they differ from pure classical errors in that the variability in estimated exposures is also not independent across space. Szpiro et al. (2011a) defined Berkson-like and classical-like errors as errors sharing some characteristics with Berkson and classical errors, respectively, but with some differences. Specifically, Berkson-like errors occur when the modeled exposure does not capture all of the variability in the true exposure. Berkson-like errors increase the variability around the health effect estimate in a manner similar to pure Berkson error, but Berkson-like errors are spatially correlated and not independent of predicted exposures, unlike pure Berkson errors. Berkson-like error can lead to bias of the health effect estimate in either direction (Szpiro and Paciorek, 2013).

The influence of these types of exposure errors on health effect estimates for specific short-term and long-term exposure study designs is evaluated in Section 3.4.5. This review of the influence of error on exposure estimates used in epidemiology studies informs evaluation of confounding and other biases and uncertainties when considering the health effects evidence in CHAPTER 5, CHAPTER 6, CHAPTER 7, CHAPTER 8, CHAPTER 9, CHAPTER 10, and CHAPTER 11.

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3.2.2 Conceptual Model of Total Personal Exposure

A conceptual model of personal exposure is presented to highlight measurable quantities and the uncertainties that exist in this framework. An individual's time-integrated total exposure to PM can be described based on a compartmentalization of the person's activities throughout a given time period:

$$E_T = \int_1^n C_j dt$$

Equation 3-1

where E_T = total exposure over a time-period of interest, C_j = airborne PM concentration at microenvironment j, n = total number of microenvironments, and dt = portion of the time-period spent in microenvironment j. Total exposure (E_T) can be decomposed into a model that accounts for exposure to PM of ambient (E_a) and nonambient (E_{na}) origin of the form:

$$E_T = E_a + E_{na}$$

Equation 3-2

Indoor combustion, such as cooking, smoking, or candle burning, as well as cleaning, and other activities are nonambient sources of PM (see Section 3.4.1.2, indoor-outdoor [I/O] relationships on indoor PM) that are specific to individuals and result in variable nonambient exposures across the population. Assuming steady-state outdoor conditions, E_a can be expressed in terms of the fraction of time spent in various outdoor and indoor microenvironments (U.S. EPA, 2006; Wilson et al., 2000):

$$E_a = \Sigma f_o C_o + \Sigma f_i F_{inf,i} C_{o,i}$$

Equation 3-3

where f_o = fraction of the relevant time period (equivalent to dt in Equation 3-1) in outdoor microenvironments; f_i = fraction of the relevant time period (equivalent to dt in Equation 3-1) in indoor microenvironments; C_o = PM concentration in outdoor microenvironments; $C_{o,I}$ = PM concentration in outdoor microenvironments adjacent to an indoor microenvironment i; and $F_{inf,i}$ = infiltration factor for indoor microenvironment i. Equation 3-3 is subject to the constraint $\Sigma_{fo} + \Sigma_{fi} = 1$ to reflect the total exposure over a specified time period, and each term on the right hand side of the equation has a summation because it reflects various microenvironmental exposures. Here, "indoors" refers to being inside any aspect of the built environment, [e.g., homes, schools, office buildings, enclosed vehicles (automobiles, trains, buses), and/or recreational facilities (movie theaters, restaurants, bars)], while "outdoors" refers to outdoor microenvironments (e.g., parks, yards, sidewalks, and bicycles or motorcycles). Assuming steady state ventilation conditions, the infiltration factor (F_{inf}) is a function of the penetration (P) of PM into the microenvironment, the air exchange rate (a) of the microenvironment, and the rate of PM loss (k) in the microenvironment:

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$$F_{inf} = \frac{Pa}{(a+k)}$$

Equation 3-4

In epidemiologic studies, the ambient PM concentration, Ca, is often used in lieu of outdoor microenvironmental data to represent these exposures based on the availability of data. Thus, it is often assumed that $C_o = C_a$ and that the fraction of time spent outdoors can be expressed cumulatively as f_o; the indoor terms still retain a summation because infiltration differs for different microenvironments. If an epidemiologic study employs only C_a, then it is assumed that exposure to ambient PM, E_a given in Equation 3-3, is re-expressed solely as a function of C_a:

$$E_{\mathrm{a}} = \left(f_{\mathrm{o}} + \Sigma f_{\mathrm{i}} F_{\mathrm{inf,i}}\right) C_{\mathrm{a}}$$

Equation 3-5

Equation 3-5 encapsulates several facets of the relationship between ambient concentration and E_a. First, C_a represents all ambient PM concentrations combined. Measurements and models to quantify C_a may assign one uniform PM concentration in the region of study (e.g., Section 3.3.1.1), or it might be modeled to represent how it varies outdoors across space (Section 3.4.2.2). Second, exposure is related to both concentration encountered and time spent in a given microenvironment. Outdoor exposure is directly influenced by ambient concentration and time spent outdoors. Indoor exposure occurs where infiltration of ambient PM into the envelope of an enclosed space (e.g., building, bus) likely reduces ambient PM exposure by filtering out a fraction of the ambient PM, but the influence of ambient concentration and time of exposure is still present. The components of indoor and outdoor exposure to ambient PM to comprise total ambient PM exposure, E_a. Further combining these factors with human activity level influences dose (Section 4.1.7).

Certain factors influence whether Equation 3-5 is a reasonable approximation for Equation 3-3, including the spatial variability of outdoor PM concentrations due to spatial distribution of sources; meteorology, topography, oxidation rates, and the design of the epidemiologic study. These equations also assume steady-state microenvironmental concentrations. Errors and uncertainties inherent in using Equation 3-5 in lieu of Equation 3-3 are described in Section 3.4. with respect to implications for interpreting epidemiologic studies. Epidemiologic studies often use concentration measured at an ambient monitor to represent ambient concentration; thus α, the ratio between personal exposure to ambient PM and the ambient concentration of PM, is defined as:

$$\alpha = \frac{E_a}{C_a}$$

Equation 3-6

Combining Equation 3-5 and Equation 3-6 yields:

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$$\alpha = f_o + \Sigma f_i F_{inf,i}$$

Equation 3-7

where α varies between 0 and 1. If a person's exposure occurs in a single microenvironment, the ambient component of the microenvironmental PM concentration can be represented as the product of the ambient concentration and F_{inf}. Time-activity data and corresponding estimates of F_{inf} for each microenvironmental exposure are needed to compute an individual's α with accuracy (U.S. EPA, 2006). In epidemiologic studies, α is assumed to be constant in lieu of time-activity data and estimates of F_{inf}, which can vary spatially (between homes) and temporally (within a home) based on building and meteorology-related air exchange characteristics.

The conceptual model presented in Equation 3-1 through Equation 3-7 establish a framework for considering the influence of exposure measurement error on statistical models used in epidemiology studies. Exposure measurement error occurs when there is an absence of information for the variables in this framework, so assumptions must be made regarding ambient exposures. If important local outdoor sources and sinks exist but are not captured by ambient monitors, then the ambient component of the local outdoor concentration may be estimated using dispersion models, land use regression (LUR) models, chemical transport models (CTMs), satellite data, or some combination of these techniques, which are described in Section 3.3.2.

3.2.3 Exposure Considerations Specific to PM

The inhalation exposure route relevant for PM is influenced by sources, chemistry, particle size distribution, meteorology, and ambient concentrations, as described in detail in Chapter 2 and briefly summarized here.

The polydisperse size distribution (Section 2.2) and composition (Section 2.3) of PM interact to influence several aspects of exposure. UFP dominates the number concentration (NC) distribution of PM, while $PM_{2.5}$ typically dominates the mass distribution. Combustion via energy production, mobile sources, and industrial processes is the main primary anthropogenic source of UFP and $PM_{2.5}$. Brake, tire, and clutch wear can also contribute to primary UFP, $PM_{2.5}$, and $PM_{10-2.5}$. Secondary production of NO_3^-, NH_4^+, and SO_4^{2-} are also major contributors to $PM_{2.5}$, and the magnitude of those contributions varies by region, time of day, and season. UFP will also grow to the accumulation mode following emissions on time scales of hours to days. Road and construction dust are important anthropogenic sources of $PM_{10-2.5}$ in urban areas, while agricultural dust is an anthropogenic source of $PM_{10-2.5}$ in rural areas. Biogenic $PM_{10-2.5}$ from pollen can also be a substantial contributor to overall $PM_{10-2.5}$.

The size distribution influences transport and dispersion of PM, therefore affecting spatial and temporal variability of PM concentration and hence exposure (U.S. EPA, 2009b). UFP has a short lifetime because it either readily evaporates or undergoes rapid growth into the accumulation mode via

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agglomeration of UFP into larger particles, condensation or adsorption of vapors onto UFP, or reaction of gases in or on the particles (Section 2.2). $PM_{2.5}$ will tend to follow the wind unless evaporating, participating in a surface reaction, and/or accumulating to a larger size. Particle growth may enhance deposition. $PM_{10-2.5}$ in dust can settle out of the air at a faster rate than $PM_{2.5}$. Resuspension by vehicle-generated turbulence, tire motion, or other activities may occur for particles of any size but are more likely for $PM_{10-2.5}$, which forms more readily via mechanical generation (Section 2.3.3). As a result, spatial and temporal variability of PM exposure concentration tends to be greater for UFP and $PM_{10-2.5}$ compared with $PM_{2.5}$ (Section 2.5).

Size distribution will also affect what fraction of the ambient air penetrates indoors (U.S. EPA, 2009b). Because $PM_{2.5}$ navigates changes in direction more easily, more $PM_{2.5}$ tends to infiltrate indoors compared with $PM_{10-2.5}$, which impacts onto building envelope surfaces more easily. UFP is more likely to diffuse onto building envelope surfaces compared with $PM_{2.5}$, so it would be expected that a lower proportion of UFP would infiltrate indoors compared with $PM_{2.5}$.

In summary, variability and uncertainties in accounting for PM emissions, chemistry, transport, and dispersion (noted here and described in detail in CHAPTER 2) leads to variability and uncertainties in estimates of exposure concentrations. For PM, uncertainties extend to characterization of the statistical distribution of particles by size and concentration (spatially and temporally). Because they have shorter lifetimes compared with $PM_{2.5}$, spatial and temporal variability is more pronounced for the lower (UFP) and upper ($PM_{10-2.5}$) segments of the particle size distribution compared with the accumulation mode ($PM_{2.5}$). Such uncertainties may complicate estimation of exposure concentrations using models such as CTMs (Section 3.3.2.4) or satellite-based methods where a relationship between $PM_{2.5}$ and surface measurements is derived (Section 3.3.3). Errors associated with these factors are described further in Section 3.4.2, and their influence on epidemiologic study results is considered in Section 3.4.5.

3.3 Methodological Considerations for Use of Exposure Data and Models

This section describes methods for estimating human exposure to PM, along with their strengths and limitations, which are important to understand when developing associations between PM exposure and health endpoints in epidemiologic analyses. The 2009 PM ISA (U.S. EPA, 2009b) and other literature [e.g., Madrigano et al. (2013); Hubbell (2012); Tagaris et al. (2009)] presented information about ambient and personal monitoring, as well as models for data averaging, spatial interpolation, LUR, CTM, and dispersion models. The current section extends that presentation by updating the assessment with discussion of new methodology and a more detailed consideration of features, strengths, and limitations of measurement and modeling techniques for PM exposure assessment.

For epidemiologic analyses, accurately assigning air pollutant exposure concentrations to individuals is difficult given the limited spatial and temporal resolution of the available observations.

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Applications can vary in scale, from personal (Baxter et al., 2013; Brown et al., 2012; Dons et al., 2012; Kaur and Nieuwenhuijsen, 2009) to national (Fann et al., 2012; Bell et al., 2011b) to global (Lelieveld et al., 2015; Brauer et al., 2012; Lim et al., 2012). In some studies, personal monitoring has been used, but study limitations (e.g., expense, recruiting subjects to participate) typically constrain the size of the population studied in panel studies (Baxter et al., 2013; Ozkaynak et al., 2013; Jerrett et al., 2005a; Sarnat et al., 2000). Thus, methods are employed that use the limited observational data available from ambient air quality monitoring regulatory networks (Solomon et al., 2011) and special, often intensive studies that may be designed to provide data for exposure assessment and/or spatial characterization (Vedal et al., 2013; Hansen et al., 2006; Edgerton et al., 2005; Jerrett et al., 2005b; Butler et al., 2003; Hansen et al., 2003). In addition, health studies are taking advantage of satellite data [e.g., Madrigano et al. (2013); Liu et al. (2009)], mobile monitoring data [e.g., Levy et al. (2014); Bergen et al. (2013)], and models [e.g., Jerrett et al. (2016); Turner et al. (2016); Villeneuve et al. (2015); Pope et al. (2014)].

Modeling PM exposure concentrations can be challenging because PM may contain a mixture of components and is found in a continuum of sizes (Section 2.2). Approaches for modeling PM exposure concentration can generally be used for different sized particles ($PM_{10-2.5}$, $PM_{2.5}$, UFP) and components, though additional considerations may be involved. For example, there are very limited observational data on UFP for cross-validation (Section 2.5); $PM_{2.5}$ composition data from ambient monitoring networks are typically available every few days (e.g., every third or every sixth day) using 24-hour integrated measurements. Different observational techniques for $PM_{10-2.5}$, $PM_{2.5}$, and UFP have different biases and uncertainties, and composition may influence biases and uncertainties within a given size fraction. Some observed components (e.g., OC) are composed of multiple compounds that behave differently in the environment.

There are a range of approaches used to model PM exposure that are applied for specific purposes, and their uses depend upon available data. Ozkaynak et al. (2013) developed a hierarchy of methods based upon complexity, ranging from using ambient monitoring data as an exposure surrogate to human exposure models accounting for time-activity data and microenvironmental exposure concentrations (Figure 3-1). This list can be extended to include source apportionment models. The amount and complexity of model input data increases with increasing complexity of the models. Increasing the complexity of the exposure modeling methods may reduce exposure error in some cases (Sarnat et al., 2013b).

This section includes discussions of surface measurements (including fixed-site and personal monitoring [Section 3.3]), modeling approaches (increasing in complexity from data averaging techniques through microenvironmental models [Section 3.3.2]), and satellite-based methods (Section 3.3.3). Each of these approaches has strengths and limitations, and several new studies discussed in Section 3.3.2.4.3 and Section 3.3.3 blend observations and air quality model results to reduce exposure measurement error. An analysis of the relative strengths and limitations of these methods for application in epidemiologic studies is provided in Section 3.3.5.

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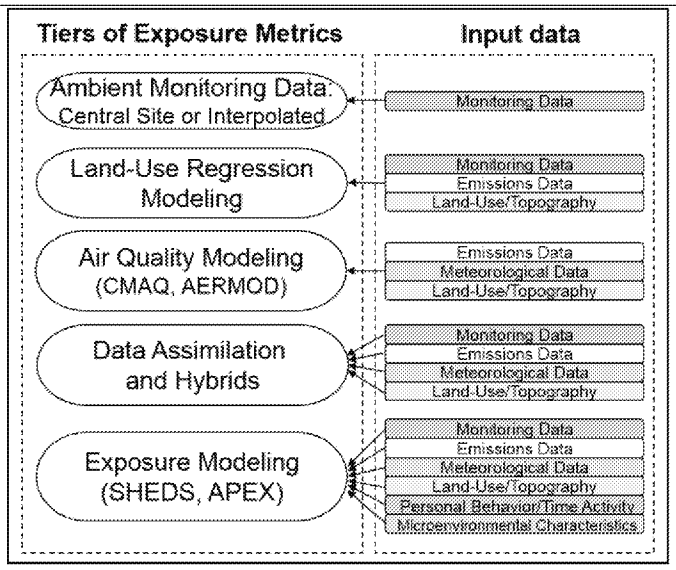

Source: Permission pending, Adapted from Ozkaynak et al. (2013).

Figure 3-1 Tiers of exposure models relevant to epidemiology studies and input data types for each exposure model tier.

3.3.1 Surface Measurement

The 2009 PM ISA (U.S. EPA, 2009b) discussed the use of ambient PM concentration data measured at FRMs and FEMs and used as surrogates for PM exposures, and main points are summarized in Section 2.4. The technology for measuring ambient PM at fixed-site monitors has largely stayed the same. More attention is given in Section 2.4.3 to measuring UFP concentrations. New insights to help interpret $PM_{2.5}$, $PM_{10-2.5}$, and UFP concentration data for use in exposure assessment studies are provided in Section 3.4.1.1.

The 2009 PM ISA (U.S. EPA, 2009b) described developments in using personal monitors for exposure assessment. Specifically, developments in light scattering continuous monitoring instrumentation, passive sampling, cascade impactor sampling for $PM_{10-2.5}$ and $PM_{2.5}$, and use of GPS for estimating time-activity were presented. Since then, new developments have been made in active sampling of $PM_{10-2.5}$, $PM_{2.5}$, and UFP. Important developments include reducing the size and increasing portability and battery life of samplers. These are described in Section 3.3.1.2.

3.3.1.1 Ambient Monitoring

Ambient PM data from FRM or FEM from individual sites continue to be used widely in health studies as a surrogate for PM exposure concentration. (Pope et al., 2009; Zanobetti and Schwartz, 2009) provide a number of reasons for the continued use of fixed-site monitor data as exposure surrogates:

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(1) instrument error is typically small compared to spatiotemporal modeling error, (2) an ambient monitor may provide a comprehensive set of measurements, (3) the need to capture temporal variation is typically greater than the need to capture spatial variation in short-term exposure studies, and (4) ambient monitor data provide a useful reference for comparing population exposure concentration estimates in long-term exposure studies. The ambient monitor approach is the least data intensive approach among all exposure concentration estimation methods because it only requires data from a single monitor to represent exposures to a large area (on the order of 100 km^2).

Differences in sampler design for $PM_{2.5}$, $PM_{10-2.5}$, and UFP influence the quality of exposure concentration data available for epidemiologic studies of each respective size cut. For $PM_{2.5}$ samplers, quality assurance testing has demonstrated that $PM_{2.5}$ concentration measurements are replicable [(U.S. EPA, 2004), Section 2.4.1.1], lending confidence to their frequent application in exposure assessment studies. In contrast, $PM_{10-2.5}$ exposure concentration has been measured in three ways [dichotomous samplers, differencing using concentrations from collocated PM_{10} and $PM_{2.5}$ monitors, and subtracting area-wide (e.g., county-wide) $PM_{2.5}$ concentration from area-wide PM_{10} concentration] with large differences in quality assurance (Section 2.4.2). It is expected that dichotomous samplers would produce the most accurate measure of $PM_{10-2.5}$ concentration for use as an exposure surrogate, because dichotomous samplers are designed for isokinetic flow appropriate for each PM cut point. However, a systematic study comparing all three methods has not yet been performed. Differences in spatial variability of $PM_{2.5}$ and $PM_{10-2.5}$ (Section 2.5) coupled with low-moderate correlation (Section 3.4.3.1) suggest that area-wide differences would provide the least accurate measure of $PM_{10-2.5}$ concentration for use in exposure assessment studies. UFP is usually measured by condensation particle counters (CPC) (Section 2.4.3.1) and at times by inertial impaction (Section 2.4.3.3). Testing of CPCs has shown that CPCs may operate at 95% counting efficiency. However, concentrations measured by UFP samplers are also more susceptible to negative bias due to larger evaporative losses compared with $PM_{2.5}$ or $PM_{10-2.5}$ concentration measurements. Hence, there is generally higher confidence in $PM_{2.5}$ concentration measurements than in $PM_{10-2.5}$ and UFP concentration measurements used as exposure surrogates.

3.3.1.2 Personal Monitoring

Methods for personal PM monitoring were described in the 2009 PM ISA (U.S. EPA, 2009b). At that time, filter-based personal monitors were used most frequently. Developments at the time of the 2009 PM ISA included size selectivity of personal samples using a Personal Cascade Impactor Sampler that can sample down to a cut point of 250 nm (Singh et al., 2003), a mini-cyclone with the capability of sampling down to 210 nm (Hsiao et al., 2009), and a two-stage cascade impactor for $PM_{10-2.5}$ sampling (Case et al., 2008). A passive monitor had also been adapted for $PM_{10-2.5}$ sampling (Ott et al., 2008; Leith et al., 2007) based on a passive sampler developed earlier that can be used for user-defined size fractions including $PM_{2.5}$ (Wagner and Leith, 2001a, b). Light-scattering detection devices for continuous monitoring, such as the Personal DataRam (pDR, Thermo Scientific, Waltham, MA), the DustTrak (TSI,

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Inc., Shoreview, MN), and the SidePak (TSI, Inc., Shoreview, MN) for PM_{10} or $PM_{2.5}$ mass concentration and the P-Trak (TSI, Inc., Shoreview, MN) or personal CPC Model 3007 (TSI, Inc., Shoreview, MN) for UFP count concentration were also described in the 2009 PM ISA. The P-Trak samples between 20 nm and 1 μm, and the CPC samples between 10 nm and 1 μm. However, it is anticipated that the majority of particles are smaller than 100 nm when measuring NC (see Preface). Additionally, the 2009 PM ISA detailed new methodologies used by investigators to enhance personal sampling by incorporating videotape (Sabin et al., 2005) or Global Positioning Systems (GPS) (Westerdahl et al., 2005) into their sampling protocols to estimate personal exposure by using simultaneous measures of exposure concentration and time-activity data. Techniques discussed in the 2009 PM ISA are widely in use, and development of new samplers have largely built upon these techniques. Table 3-1 lists these new techniques with sampling size fraction, speciation, mechanism, and error characteristics.

Table 3-1 New or innovative methods for personal sampling of PM exposure concentrations published since the 2009 PM ISA.

Reference	Active or Passive Sampling	Sampler	Size Fraction	Species	Mechanism	Error Characteristics
Thornburg et al. (2009)	Active	Coarse Particulate Exposure Monitor (CPEM)	$PM_{10-2.5}$, $PM_{2.5}$	NA	Three-stage impactor	$PM_{10-2.5}$: −23% (R^2 = 0.81) $PM_{2.5}$: −3% (R^2 = 0.91) compared with a dichotomous $PM_{10-2.5}$ sampler
Volckens et al. (2016)	Active	Ultrasonic Personal Aerosol Sampler (UPAS)	$PM_{2.5}$	NA	Miniature piezoelectric pump with a cyclone for 2.5 μm size cut plus additional sensors for air flow, sunlight, temperature, pressure, relative humidity, and acceleration	−1.4% compared with a $PM_{2.5}$ FRM
Ryan et al. (2015b)	Active	Personal UFP Sampler (PUFP)	UFP	NA	Water-based CPC plus GPS for location	+16% (R^2 = 0.99)

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Table 3-1 (Continued): New or innovative methods for personal sampling of PM exposure concentrations published since the 2009 PM ISA.

Reference	Active or Passive Sampling	Sampler	Size Fraction	Species	Mechanism	Error Characteristics
Nash and Leith (2010)	Passive	Algorithm to modify output from the Wagner-Leith passive sampler to UFP	UFP	Yes	Model of deposition flux developed the passive sampler's size range	6% compared with SMPS
Cai et al. (2014); Cai et al. (2013)	Active	Modification to the Microaethalometer (AethLabs, Berkeley, CA)	$PM_{2.5}$	BC	Reduced humidity and temperature fluctuations through addition of a diffusion dryer	53 ± 238% difference in 1-min readings between the original and diffusion dryer inlet on 97−100% RH day and 5 ± 33% difference between original and diffusion dryer inlet on 65% RH day. The differences reduce to approximately 1% when data are averaged over an hour.
Hagler et al. (2011);Cheng and Lin (2013)	Active	Algorithm to modify output from the Microaethalometer (AethLabs, Berkeley, CA)	$PM_{2.5}$	BC	Introduced a data cleaning algorithm to reduce erroneous fluctuations in the signal (i.e., noise)	Comparison between 1-min data with optimized noise reduction algorithm was comparable to 5-min data averaged with noise
Sameenoi et al. (2012)	Active	Microfluidic electrochemical sensor to detect oxidative potential of PM	Any	ROS	Incorporated DTT assay into Particle into Liquid Sampler (PILS)	Comparison with traditional DTT assay: $R^2 = 0.98$
Sameenoi et al. (2013)	Active	Microfluidic paper-based analytical device (μPAD) to detect oxidative potential of PM	Any	ROS	Collected $PM_{2.5}$ and PM_{10} on filters, desorbed, then pipetted onto μPAD	Comparison with traditional DTT assay: bias = 10.5%, $R^2 = 0.98$

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Table 3-1 (Continued): New or innovative methods for personal sampling of PM exposure concentrations published since the 2009 PM ISA.

Reference	Active or Passive Sampling	Sampler	Size Fraction	Species	Mechanism	Error Characteristics
Landreman et al. (2008)	Active	Expose rat macrophages to collected aerosol sample to detect oxidative potential of PM	Any	ROS	Collected $PM_{2.5}$ onto filters, desorbed, then pipetted onto a 96-well plate seeded with rat macrophages	Response corresponded to spikes for samples exposed to different numbers of macrophages (not quantitative)

BC = black carbon; DTT = dithiothreitol; ROS = reactive oxygen species.

Prevalent field usage of continuous personal PM monitors using optical techniques necessitates validation of these instruments, since calibration is not possible given that ambient PM does not have replicable optical properties. Wallace et al. (2011) tested the 6 pDR and 14–16 DustTrak (number varied with tests) for $PM_{2.5}$ (with a size-selective inlet), and 14 P-Trak personal samplers for particle number to measure UFP exposure concentrations to establish operational parameters (MDL, bias, precision, drift) for each sampler compared with the median. MDL for the DustTrak and pDR were estimated to be 5 μg/m^3 and 5.5 μg/m^3, respectively (not detected for the P-Trak), and relative precision was within 10% for all four monitors. The pDR measurements were 60% higher than collocated personal gravimetric samples from the field tests ($R^2 = 0.7$), and the DustTrak measurements were 164% higher than personal gravimetric measurements ($R^2 = 0.9$). The authors pointed out that the higher readings from the light-scattering instruments relative to the gravimetric measurements are due in part to the lower density of ambient PM relative to the density of the aerosol standard used for laboratory calibration. Another factor Wallace et al. (2011) noted to influence the performance of light-scattering personal PM monitors is relative humidity (RH). High RH results in sorption of water to particles and an increase in volume and mass detected by the instrument. Quintana et al. (2000) found that pDRs produced much higher readings than a gravimetric TEOM instrument when RH was above 85%, but that pDR readings tracked the TEOM readings relatively well at RH values below 60%. Since indoor RH is generally maintained below 60%, the influence of RH is likely to mainly affect outdoor light-scattering measurements, particularly in morning, evening, and overnight hours when RH is highest. Optical personal samplers are subject to errors given the inability to calibrate the monitors for ambient characteristics. The characterization work described above has been done for optical sampling of $PM_{2.5}$, so uncertainties are greater for the $PM_{10-2.5}$ and UFP size fractions. Instrument error and replicability and the factors that affect them must be evaluated for each use in panel studies.

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3.3.2 Modeling

At the time of the 2009 PM ISA (U.S. EPA, 2009b), fine-scale exposure prediction models were still relatively nascent in their development. Methods reviewed include time-weighted microenvironmental models and stochastic exposure models for estimation of PM exposure and dispersion models, LUR, and GIS-based modeling approaches for estimation of PM exposure concentration, and attention was given to the models' limitations in adequately capturing spatial variability of PM concentration, particularly for more variable UFP and $PM_{10-2.5}$. Since the 2009 PM ISA, more approaches to spatial averaging of concentrations used for estimating exposure concentrations (Section 3.3.2.1), and new developments in spatiotemporal interpolation of exposure concentration surfaces (Section 3.3.2.2), LUR (Section 3.3.2.3), and dispersion models (Section 3.3.2.4.2) have appeared in the peer-reviewed literature. Additionally, there has been growing use of chemical transport models (CTMs) in exposure assessment studies (Section 3.3.2.4.1) in recent years. Table 3-2 provides an overview of the modeling approaches discussed in this section.

The models discussed in the following sections are typically validated by the study authors using surface monitoring data, but model validation is not performed consistently across the literature. Table 3-3 lists performance measures that have been utilized in the recent PM exposure modeling literature. Model performance is typically evaluated for bias or error using both absolute and relative (or normalized) metrics.

Table 3-2 Comparison of models used for estimating exposure concentration or exposure.

	Type of Model						
Factors[a]	Data averaging	IDW/ Kriging	LUR/ ST	CTM and Hybrid	Dispersion	Satellite and Hybrid	Microenvironmental
Type of model	C	C	C	C	C	C	E
Distance from source	X	X	X	X	X	X	X
Emission rate			X	X	X	X	X
Terrain or land use			X	X	X	X	X
Dispersion				X	X	X	X
Chemistry				X	X	X	X

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	Type of Model						
Factors[a]	Data averaging	IDW/ Kriging	LUR/ ST	CTM and Hybrid	Dispersion	Satellite and Hybrid	Microenvironmental
Human activity							X
Infiltration							X
Inhalation							X

C = concentration model, CTM = chemical transport model, E = exposure model, IDW = inverse distance weighting, LUR = land use regression, ST = spatiotemporal models.

[a]Factors that may be available in each model are checked.

Table 3-3 Statistical measures used for air quality model performance evaluation.

Performance Measures	Definition[a]
Mean bias (MB)	$\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)$
Mean error (ME)	$\frac{1}{N}\sum_{i=1}^{N}\lvert P_i - O_i\rvert$
Root mean square error (RMSE)	$\sqrt{\frac{1}{N}\sum_{i=1}^{N}(P_i - O_i)^2}$
Coefficient of determination (R^2)	$\frac{\left\{\sum_{i=1}^{N}(O_i - \overline{O})(P_i - \overline{P})\right\}^2}{\sum_{i=1}^{N}(O_i - \overline{O})^2 \sum_{i=1}^{N}(P_i - \overline{P})^2}$

[a]P_i and O_i are prediction and observation at the ith monitoring site, respectively; N is the number of monitoring sites.

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3.3.2.1 Data Averaging

Averaging measurements from all monitors in a study area is frequently used to mitigate some of the errors associated with using data from a single ambient monitor to estimate exposure concentrations for a population. There are many averaging approaches in use to provide more representative exposure concentration estimates than those derived from a fixed-site ambient monitor. For example, Strickland et al. (2011) compared nearest fixed-site monitor concentrations of $PM_{2.5}$ and $PM_{2.5}$ components (SO_4^{2-}, OC, EC) averaged over 24 hours with concentrations averaged over three monitors (unweighted). They found that $PM_{2.5}$ and $PM_{2.5}$–SO_4^{2-} mass concentrations were within 8% of each other, with strong correlations between the concentration obtained by a fixed-site monitor and with that obtained by a population-weighted average Spearman $R = 0.969$. Reported $PM_{2.5-OC}$ concentrations had a Spearman correlation of $R = 0.847$, but more spatially varying $PM_{2.5-EC}$ had a Spearman correlation of $R = 0.831$. Goldman et al. (2012) had similar findings when comparing nearest monitor with unweighted averaging. Strickland et al. (2013) compared unweighted averages across monitors with concentrations measured at fixed-site monitors and concentrations estimated to be the "true" exposure concentrations at grid cells within the study domain. The fixed-site monitor produced $PM_{2.5}$ concentrations with the largest biases of −31.3%, in comparison with the unweighted average (−9.0%). Biases for $PM_{2.5}$ components (SO_4^{2-}, NO_3^-, NH_4^+, EC, OC) were similar for both the fixed-site monitor and unweighted average. In the unweighted averaging technique studied by Strickland et al. (2013), temporal variability may be dampened, leading to Berkson errors. As described below, more spatial heterogeneity inherent to the exposure concentration field implies greater Berkson errors.

Spatial averaging techniques include area-weighting and population-weighting (Vaidyanathan et al., 2013). Such schemes require some type of spatial modeling of data before averaging. For example, area and population-weighting might involve use of a regression model of PM or PM component concentration and population density, land use, or emission estimates to develop exposure concentration estimates at grid locations. Concentrations for census tracts, zip codes, or counties can then be averaged and weighted by the associated areas or populations. In such schemes, the objective of the spatial modeling is to develop more representative area or population estimates.

Population-weighted averaging is designed to reduce bias in the health effect estimate by giving greater weight to the locations where more people live. As part of the study referenced above, Strickland et al. (2013) compared population-weighted averages across monitors with concentrations measured at fixed-site monitors and concentrations estimated to be the "true" exposure concentrations at grid cells within the study domain. The population-weighted average produced $PM_{2.5}$ concentrations with biases of −8.1% in comparison with the true $PM_{2.5}$ exposure concentrations. Biases for $PM_{2.5}$ components (SO_4^{2-}, NO_3^-, NH_4^+, EC, OC) were similar for both the fixed-site monitor and unweighted average. Strickland et al. (2011) compared nearest fixed-site monitor concentrations of $PM_{2.5}$ and $PM_{2.5}$ components (SO_4^{2-}, OC, EC) averaged over 24 hours with concentrations averaged using population-weighted averages. They found that $PM_{2.5}$ and $PM_{2.5}$–SO_4^{2-} mass concentrations were within 8% of each other, with correlations

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among the three spatial representations ranging from Spearman $R = 0.963–0.995$. Reported $PM_{2.5-OC}$ concentrations had Spearman correlations of $R = 0.891$, but more spatially varying $PM_{2.5-EC}$ had Spearman $R = 0.804$. Goldman et al. (2012) had similar findings when comparing nearest monitor, unweighted, and population-weighted averaging. These results suggest that population-weighted averaging may provide a small improvement over unweighted averaging for estimation of exposure concentration.

Spatial averaging approaches may influence exposure measurement error (Goldman et al., 2010) and associations between short-term $PM_{2.5}$ exposure and health outcomes (Goldman et al., 2012). In the latter study, the authors noted improved population-weighted R^2 values (relative to the fixed-site ambient monitoring method) between exposure concentration metrics estimated using data averaging methods and the simulated "true" ambient concentration field. For example, the R^2 values increased from 0.25 for a fixed-site ambient monitoring method to approximately 0.38 for data averaging methods.

Various methods can be chosen for temporal averaging, such as straight arithmetic averaging or methods that account for site-specific variability and that also account for the lack of some observations during the period. Temporal averaging is used to estimate exposure concentrations over different time intervals. Hourly and daily measures are averaged to provide metrics of interest (e.g., daily, weekly, monthly, seasonal, and annual). Darrow et al. (2011) tested different averaging intervals and found that 1-hour daily max $PM_{2.5}$ concentrations had high correlation with 24-hour average (Spearman $R = 0.82$) and moderate correlations (Spearman $R = 0.75$ and 0.68) with commuting time (7:00–10:00 and 16:00–19:00) and daytime (8:00–19:00) average $PM_{2.5}$ concentrations, respectively. As with the development of spatial averages, the objective of temporal averaging is to minimize error that might be introduced due to missing data from a time-series, so that diurnal, weekly, seasonal, or annual trends can be well characterized.

Spatial and temporal averaging methods provide a mechanism for interpolating where data are missing over space or in a time-series, respectively. The literature shows that averaging techniques produce some bias when compared with true exposure concentrations, but averaging techniques do present an improvement over using data from a single fixed-site monitor.

3.3.2.2 Spatial Interpolation Methods

The single fixed-site ambient monitor and methods that average concentration data across monitoring sites in an area both lead to exposure concentration estimates with no spatial variation. When spatially resolved estimates of PM exposure concentration are desired, a variety of approaches are available for two-dimensional interpolation of observations ranging from smoothing techniques (described here) to statistical modeling techniques involving additional data (Section 3.3.2.4). Various spatial interpolation methods exist that use multiple monitors to provide spatially varying fields. Such

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methods include: inverse distance weighting (IDW), inverse distance squared weighting (ID2W) (Hoek et al., 2002), and kriging (Mercer et al., 2011; Whitworth et al., 2011).

IDW, in which ambient PM concentration at a receptor point is calculated as the weighted average of ambient PM concentration measured at monitoring locations, is a commonly used simple interpolation method [e.g., Tai et al. (2010)]. Several variations of IDW have been used to estimate exposure based on ambient PM concentration surfaces. The weighting factor is an inverse function of distance between the receptor and the monitor. For example, Brauer et al. (2008) and MacIntyre et al. (2011) estimated exposure to ambient $PM_{2.5}$ and other industrial pollutants within 10 km of point sources using an IDW sum of ambient $PM_{2.5}$ concentration and the three closest monitors within 50 km. Often, the weighting factor is the inverse distance raised to some power, and a higher power is applied to increase the weight on monitors that are closer to the receptor. Rivera-González et al. (2015) applied an ID2W model and compared the results with a citywide average, use of the nearest monitor, or kriging for development of an ambient $PM_{2.5}$ concentration surface. The results from IDW were correlated with the other city-wide averaging, nearest monitor, and ordinary kriging (Pearson $R = 0.83–0.99$), and the mean ambient $PM_{2.5}$ concentration estimated with IDW was within 5% of the mean computed with the other methods. Neupane et al. (2010) compared estimates of the ambient $PM_{2.5}$ concentration surface calculated using IDW with a $PM_{2.5}$ concentration surface calculated using both bicubic spline interpolation. Bicubic spline interpolation produced a lower mean ambient $PM_{2.5}$ concentration and larger IQR compared with IDW. Because there is no reference value in these studies, it is difficult to conclude that IDW presents any substantial improvement in prediction accuracy compared with other methods. These findings indicate that the results of IDW are comparable to methods that average concentrations across monitors and to methods that smooth concentration surfaces when estimating $PM_{2.5}$ concentration.

Kriging is a set of well-established methods that use observed covariance for geostatistical interpolation [e.g., Beelen et al. (2009)]. Recent developments have been made to improve kriging techniques. Pang et al. (2010) developed a space-time Bayesian Maximum Entropy (BME) model and compared it with ordinary kriging (OK). OK assumes linearity between data points, and it also assumes that the data are normally distributed. BME is not restricted to linearity or normality and so can draw on different sources of information, such as space-time relationships between variables and probability distributions describing the concentration dataset, to address missing data. Pang et al. (2010) found that estimation errors were 2–4 times larger for OK compared with BME. The ability to apply nonlinear models to address missing data thus provide BME-kriging approaches greater accuracy in modeling $PM_{2.5}$ concentration surfaces.

Berkson-like error in the estimated exposure concentration may arise from smoothing inherent to spatial interpolation models, such as IDW and kriging (see Section 3.2.1 for definition of Berkson-like error). The potential for Berkson-like error may be evaluated by cross-validation across receptor locations distributed over space, and the statistical performance of spatial interpolation methods may vary from study to study. When an interpolation model is fit using a relatively sparsely distributed monitoring

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network, Berkson-like errors in estimated exposure concentration can be substantial (Alexeeff et al., 2015; Whitworth et al., 2011). All of the spatial interpolation approaches will produce spatially smoothed pollutant exposure concentration fields from monitoring data. However, spatial and temporal variabilities not captured by monitors are also not captured by these approaches.

If the quantity of data is small in each given site, or if the quality of the data obtained at the monitors is low, then classical-like error may arise (Szpiro et al., 2011a). If there are few observations, all of the interpolation methods suffer. This includes kriging, which depends on developing a variogram. With few observations at the monitoring locations, there is limited information to determine the functional coefficients used for kriging (e.g., the nugget, sill, and range). Weighting schemes for the interpolation models may amplify these errors (Wong et al., 2004).

3.3.2.3 Land Use Regression and Spatiotemporal Modeling

Direct spatial interpolation of PM exposure concentration and methods that employ static parameters to capture spatial variance can lead to excessive spatial autocorrelation when spatial variability of PM is high (Krewski et al., 2009). $PM_{2.5}$ tends to have less spatial heterogeneity than $PM_{10-2.5}$ or UFP (Section 3.4.2) given secondary production (U.S. EPA, 2009b), but high concentrations can still occur near primary sources. Statistical approaches that utilize data that vary over space and time can address this limitation. Geographic information system (GIS) models are being used to incorporate land use, emissions data, and geographic covariates into PM exposure concentration estimates. Two types of models are covered in this section, LUR and spatiotemporal models. LUR models regress observed PM concentrations on land use (and sometimes additional geographic) covariates and then use the model to predict exposure concentrations where PM is not measured (Hoek et al., 2008a; Ryan and Lemasters, 2007). Spatiotemporal models tend to incorporate kriging or autocorrelation into the response variable, which is then fit to the land use and geographic covariates [e.g., Sampson et al. (2013)].

3.3.2.3.1 Land Use Regression

LUR is an empirical approach to estimate exposure concentrations, often at very high resolution in more densely populated locations, by relating observed concentrations to the detailed information on land use. The basic approach is to develop an equation, via regression, relating observed pollutant concentrations (Hoek et al., 2008a; Ryan and Lemasters, 2007) to land use characteristics and other inputs:

$$Y(s_i, t_j) = \beta_0(s_i, t_j) + \sum_k \beta_{1,k}(s_i, t_j) X_k(s_i, t_j) + \epsilon(s_i, t_j)$$

Equation 3-8

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Here, $Y(s_i,t_j)$ is the observed concentration at location (monitor) s_i (where i is a monitor location) and time t_j, β_0 and $\beta_{1,k}$ are the regression coefficients (intercept and slopes that are potentially spatially and temporally varying, but may also be constant in time and space), X are the independent variables (e.g., land use or meteorological parameters that may vary in time and/or space), k is the index indicating type of land use, and ϵ is the residual error term. β_0 is also called the additive bias and $\beta_{1,k}$ the multiplicative bias. Other forms of LUR models are also used. While the regression equation often is linear in the independent variables (as shown above), it can include nonlinear and mixed terms, particularly if there is specific knowledge of the relationship between a concentration and a variable that would suggest a specific functional form. The resulting regression equation can then be used to predict exposure concentrations at other times (t) and locations (s) where observations are not available.

Recent studies demonstrate typical LUR model performance, performance evaluation, and variability between cities. Eeftens et al. (2012) evaluated the application of LUR models in 20 cities in Europe for $PM_{2.5}$, PM_{10}, $PM_{2.5}$ absorbance, and $PM_{10-2.5}$. First, the models for the various cities had substantially different independent variables used in the final models, as well as coefficients associated with similar independent variables, demonstrating the location-specific nature of the models. Second, the in-sample R^2 of the various city models varied between 35 and 89% for $PM_{2.5}$ and between 32 and 81% for $PM_{10-2.5}$. Evaluation using a leave one out cross-validation (LOOCV) produced R^2 levels of 21 to 79% for $PM_{2.5}$ and 3 to 73% for $PM_{10-2.5}$. R^2 was not consistent between each city. Wang et al. (2014) expanded on the same model for $PM_{2.5}$ in thirty-six European cities. They found a LOOCV R^2 of 81% (RMSE = 2.38 μg/m^3) for cities where the model was fit. However, Wang et al. (2014) tested transferability of the model to areas where the model was not fit, and R^2 dropped to 42% (RMSE = 1.14 μg/m^3). Estimation of $PM_{10-2.5}$ in the LUR can be accomplished using the difference between the PM_{10} and $PM_{2.5}$ LUR models, since each model was trained using PM_{10} and $PM_{2.5}$ concentration data. However, low LOOCV R^2 for $PM_{10-2.5}$ in select cities may have been related to how measured $PM_{10-2.5}$ concentration was calculated for the validation dataset. If reference $PM_{10-2.5}$ concentration was calculated by the difference of two collocated monitors rather than by a dichotomous sampler, flow rate differences could cause some error in the reported $PM_{10-2.5}$ concentrations. If $PM_{10-2.5}$ was calculated by the difference between concentrations measured by PM_{10} and $PM_{2.5}$ monitors that were not collocated, then errors would likely be larger.

Several features of LUR have the potential to limit the accuracy of modeled exposure concentrations. Beckerman et al. (2013a) noted that two major limitations with LUR are variable selection and how to best deal with unbalanced repeated measures, potentially involving arbitrary decisions in the model building process. They used a generalized linear model with a deletion/substitution/addition machine learning algorithm to model $PM_{2.5}$, resulting in an out-of-sample R^2 of 0.65 based on fivefold cross-validation (n-fold cross-validation means that 1/n of the data are reserved for validation with the rest used for model training, and the process is repeated n times). The ability of an LUR method to relate air pollutant concentrations to specific land uses, and thus estimate high resolution exposure concentration fields, is directly dependent on having sufficient numbers of observations in time and/or space to develop

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the regression equation with reasonable uncertainties in each of the coefficients (Wang et al., 2014). The sparseness of the routine monitoring networks may incur Berkson-like error in the exposure estimates. More intensive studies may be conducted where additional monitoring data are available (sometimes called saturation monitoring if the additional monitors lead to extensive spatial coverage). Saturation sampling can also lead to introduction of classical-like error in the exposure predictions if different measurement methods are used and differences in the methods are not fully understood (Vedal et al., 2013; Levy et al., 2010).

A related weakness of LUR is its limited generalizability when the monitor and study participant locations are different. The developed regression equations are usually restricted to the study region (typically city-scale) alone and may not be directly applied to another region, due largely to the empirical nature of LUR (Wu et al., 2011; Jerrett et al., 2005a). Local PM data are required to calibrate LUR models, and measurements must be available that estimate the spatial patterns of exposure concentrations. For example, Patton et al. (2015) found during estimation of UFP exposure concentrations in Boston urban neighborhoods that models fit to one neighborhood did not necessarily provide robust estimates of particle NC for another neighborhood, and acceptable model performance required calibration with local data. Hoek et al. (2008a) also reviewed the performance of the LUR model regarding their application for $PM_{2.5}$ given differences between where the model was fit and where it was used for predictions. R^2 values for the developed LUR models for $PM_{2.5}$ ranges from 0.17 to 0.69, with substantially lower out-of-sample R^2 in evaluation (0.09–0.47, with fewer studies performed evaluation/cross-validation). This suggests that comparing performance statistics between cities, even when using one method (in this case, LUR) can yield very different performance and that using cross-validation reduces performance, but to a degree that is not predicable from the full model R^2. This work was extended by Wang et al. (2015) to show the association between the LOOCV R^2 and a health outcome (forced vital capacity: FVC). For models of $PM_{2.5}$, Wang et al. (2015) note that cross-holdout validation, where the model is rebuilt after removing data from a site and retraining the model using the same variables, may be more appropriate than traditional LOOCV for assessing LUR performance, particularly when there are a small number of training sites, because it makes use of all data in the model evaluation process instead of leaving out a portion of the data. In summary, LUR models can have relatively good validation ($0.4 < R^2 < 0.7$), even for spatially variable $PM_{10-2.5}$, but good validation will only occur when the model is used to predict concentrations in the same geographic area where it was fit.

Although LUR models have been used to estimate long-term (e.g., annual) average PM exposure concentrations within large metropolitan areas by using variables such as road type, traffic count, land cover, and topography (Gulliver et al., 2011; Hoek et al., 2008a) and can be applied to current or historical conditions (Hystad et al., 2013), LUR has been used less frequently for time-series exposure studies. Land use variables (e.g., elevation, road-type, distance to road, land cover) usually do not vary in time. Temporal variation in the model is gained by including both the available observations and other temporally-varying inputs, such as meteorological parameters. As part of the New York City Community Air Survey (NYCCAS) in which $PM_{2.5}$ samples were collected from 150 sites across the five boroughs of

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New York City, Ross et al. (2013) built a LUR for application in a birth defects exposure study and developed a temporal adjustment procedure to increase the temporal resolution of $PM_{2.5}$ exposure concentration estimates to 2 weeks. This was accomplished by multiplying an LUR derived for one year by the ratio of 2-week averages to annual averages. Validation of the method using data from a second year of measurements produced out-of-sample R^2 of 0.83 ($R^2 = 0.88$ if two outliers were removed from the dataset). Dons et al. (2013) aimed to fit a LUR model of black carbon (BC) concentration to hourly data for a time-activity exposure study. However, they observed that many variables became insignificant when inputting hourly data into an annual model. Dons et al. (2013) instead built a LUR for hourly data using static and dynamic variables in different models. They found that LOOCV R^2 varied from 0.13 to 0.78. Higher R^2 but also higher RMSE were observed during the late morning to evening hours for the model with dynamic variables. These studies demonstrate that LUR can be extended to study temporal variability of $PM_{2.5}$ and BC, but caution must be used for application in time-series studies since model accuracy is sometimes low.

Recently, LUR has been applied to predict spatial distribution of $PM_{2.5}$ components. As part of the NYCCAS study, Ito et al. (2016) speciated the collected $PM_{2.5}$ samples and built a LUR model to predict $PM_{2.5}$ components concentrations across New York City. The temporal adjustment described above from Ross et al. (2013) was applied in the Ito et al. (2016) study, as well. LOOCV was used to test the models, and models for $PM_{2.5}$ mass and several components (Ca, Ni, V, and Zn) produced $R^2 > 0.8$. Several other components produced R^2 in the range of 0.6–0.7 (Cu, Fe, K, S, and Si), and others produced $R^2 \leq 0.5$ (Al, Br, Mn, Pb, and Ti). Spatial coefficient of variation (CV) was calculated for each component model, and high spatial variability did not always correspond to low LOOCV. For example, Ni had a spatial CV of 0.70 and LOOCV R^2 of 0.85, while Mn had a spatial CV of 0.68 and LOOCV R^2 of 0.36. The LUR models were then applied to a source attribution analysis in which 50–1,000 m buffers were placed around sources, and then annual average concentrations for each component modeled by the LUR were compared to the sources within those buffers.

In summary, new developments for LUR include adaptation of LUR models for short time resolutions and for spatially variable size fractions (UFP, $PM_{10-2.5}$) of PM and $PM_{2.5}$ components (e.g., Ca, Cu, Fe, K, Ni, S, Si, V, Zn). At the same time, several studies have improved characterization of errors and uncertainties in LUR modeling and how best to quality assure those models. Several studies drew attention to poor validations produced when LUR models were fit to one geographic area and then applied to another. Similarly, lack of spatial correlation between predicted concentrations at the model receptors and actual exposure concentrations of study participants can lead to Berkson-like error, and incompatibility of methods to model and measure PM can lead to classical-like errors (see error type definitions in Section 3.2.1).

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3.3.2.3.2 Spatiotemporal Modeling

A GIS-based spatiotemporal model provides a useful tool for large-scale spatiotemporal analysis. GIS-based mapping such as kriging utilizes the covariogram for statistical smoothing but may lead to invalid spatial features due to insufficient data for characterizing spatial variation. Generalized additive models that describe regional and small-scale spatial and temporal (monthly) gradients (and corresponding uncertainties) were developed for $PM_{10-2.5}$ and $PM_{2.5}$ over the U.S. for 1998–2007 for use in health studies (Yanosky et al., 2014). Model validation was higher for $PM_{2.5}$ (out-of-sample $R^2 = 0.77$, normalized mean bias factor, NMBF = −1.6%) compared with $PM_{10-2.5}$ (out-of-sample $R^2 = 0.52$, NMBF = −3.2%). Bias increased and precision decreased for $PM_{10-2.5}$ compared with $PM_{2.5}$. Spatial covariates, including elevation, urbanized land use within 1 km, county-level population density, distance to roadways of moderate to heavy traffic, and point-source emissions density were all determined by the authors to be important predictors of $PM_{2.5}$, although the authors did not present data for the relative contribution of each variable to the model. Yanosky et al. (2009) developed spatially and temporally resolved concentration fields of $PM_{2.5}$ and $PM_{10-2.5}$ to be used as exposure concentration estimates in long-term exposure studies for the northeastern and Midwestern U.S. Out-of-sample R^2 for the $PM_{2.5}$ model was 0.77 with precision of 2.2 μg/m^3 for 1999 to 2002, compared with out-of-sample R^2 for the $PM_{10-2.5}$ model of 0.39 with precision of 5.5 μg/m^3. The IDW method was applied as an alternative to compare with a semiempirical model. For a $PM_{2.5}$ concentration field developed for 1999 to 2002, cross-validation results for IDW show reasonable performance with out-of-sample $R^2 = 0.60$ (and cross-validation results for IDW were not available for $PM_{10-2.5}$).

Recent studies have attempted to estimate spatially resolved $PM_{2.5}$ exposure across larger regions of the U.S. for application in epidemiologic studies. For example, Sampson et al. (2013) developed a model combining universal kriging that builds from regional partial least squares regression LUR models with categorical variables describing land use, population, emissions, vegetative index, roadway type, impervious surfaces, and proximity to features. Results of cross-validation with 10-fold cross-validation produced out-of-sample $R^2 = 0.52–0.63$ at the national scale and $R^2 = 0.84–0.88$ at the regional scale. Keller et al. (2015) applied this model to $PM_{2.5}$ and BC prediction in the six MESA Air cities (Baltimore, MD, Chicago, IL, Los Angeles, CA, New York City, NY, St. Paul, MN, and Winston-Salem, NC) and obtained out-of-sample R^2 of 0.82–0.91 for $PM_{2.5}$ and 0.79–0.99 for BC (using both AQS and MESA Air monitors for cross-validation). Bergen et al. (2013) applied a similar method for four $PM_{2.5}$ components: EC, OC, silicon, and sulfur, and the out-of-sample R^2 ranges from 0.62 to 0.95. Kim et al. (2015) examined $PM_{2.5}$ component networks for suitability of the data inputs for applying spatiotemporal models for PM component exposure concentrations, and they found that the Chemical Speciation Network (CSN) and Interagency Monitoring of Protected Visual Environments (IMPROVE) networks were too sparse to fit the model. They found that the greater density of the National Particle Component Toxicity (NPACT) study network, set up outside study participants' homes, would be needed to fit the model. Additionally, differences among the three networks with respect to averaging times, quality assurance, and pump flow rates, complicates the ability to combine networks into one database for fitting the model.

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Recent developments in spatiotemporal modeling have enabled modeling of larger geographic regions and to overcome some of the limitations of kriging. In some cases, these models have been fit with good accuracy and precision. However, differences in model calibration in different regions introduce model errors, and sparse networks have been found insufficient for model fitting.

3.3.2.4 Mechanistic Models

Improvements in computational resources have led to mechanistic models (see Section 2.4.7 for a description) that are more amenable to exposure assessment studies, because they provide finer spatial resolution over larger domains and can include more components, more sources, and longer time periods compared with previous versions of CTMs (Garcia-Menendez et al., 2015; Ivey et al., 2015; Li et al., 2015; Turner et al., 2015; Hu et al., 2014d; Burr and Zhang, 2011; Civerolo et al., 2010; Wagstrom et al., 2008). Such models computationally solve the atmospheric-diffusion-reaction equations that describe the transport and physical and chemical transformations of pollutants (Seinfeld and Pandis, 2006). Turbulent diffusion is typically treated by using atmospheric dispersion coefficients or diffusivities. Mechanistic models may be used to characterize exposure concentrations where monitoring data are limited or not available.

3.3.2.4.1 Chemical Transport Model Applications for Exposure Concentration Estimation

CTMs commonly utilized for exposure concentration modeling in the U.S. include the Community Multiscale Air Quality (CMAQ) model, Particulate Matter-Comprehensive Air Quality Model with Extensions (PM-CAMx), and the University of California at Davis/California Institute of Technology (UCD/CIT) CTM (Gaydos et al., 2007; Byun and Schere, 2006; Kleeman and Cass, 2001; Russell et al., 1988) at the urban-to-regional scales and global models such as the Goddard Earth Observing System CTM (GEOS-Chem) and Comprehensive Air Quality Chemistry Model (CAM-Chem) (Garcia-Menendez et al., 2015; Bey et al., 2001). The European Air Pollution Dispersion and Chemistry Transport Model (EURAD-CTM) has been used in Europe for PM and related exposure concentration modeling (Weinmayr et al., 2015; Nonnemacher et al., 2014), and GEM-MACH is being used in Canada (Peng et al., 2017). More specialized models may also be used to model specific sources, such as forest fires (Rappold et al., 2014).

CTMs are typically applied over grid sizes of 1 km or more, depending upon the application (while grid resolutions of less than 10 km are used over urban areas, continental scale applications typically are done at about 10–40 km, and global scale applications with larger grids yet). Nested grids are used to achieve a range of resolutions in many applications (Isakov et al., 2007; Byun and Schere, 2006; Zhang et al., 2004). In some applications, CTMs are coupled directly (i.e., on-line) to a meteorological model to provide meteorological fields, commonly WRF and CMAQ (Mathur et al.,

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2010). Inputs include meteorological parameters (e.g., wind speed and direction, temperature, relative humidity, etc.) throughout the vertical layers of the atmosphere up to and including portions of the stratosphere and source emissions. The model outputs are the pollutant concentrations, and how they vary in space and/or time (Figure 3-1). The resulting fields are then used for epidemiologic studies and other studies of air quality. The ambient concentration fields are also used as inputs to microenvironmental models for estimating exposure (Baxter et al., 2013; Jones et al., 2013; Georgopoulos et al., 2005; Burke et al., 2002).

CTM models have been used for estimation of exposure concentrations, including for use in epidemiologic studies, both in North America and abroad (Ostro et al., 2015; Weinmayr et al., 2015; Anenberg et al., 2014; Marshall et al., 2014; Nonnemacher et al., 2014; Silva et al., 2013; West et al., 2013; Lim et al., 2012; Tagaris et al., 2010). For studies covering a large geographic area, CTM models can provide location-specific estimates without gaps in coverage. Issues with using CTM models relevant for exposure assessment studies are discussed below. Hu et al. (2015) used the UCD/CIT model to develop a 9-year set of simulated pollutant concentration fields, which were then used by Ostro et al. (2015) to assess the associations of $PM_{2.5}$ and UFP with health in a cohort epidemiologic study. When evaluating the model against monitoring data, they observed low error for $PM_{2.5}$ mass compared with error for individual components, such as SO_4^{2-}. In general, errors were higher when matching observations and simulated values on a daily basis compared with monthly and annual averaging periods, suggesting that model results are more accurate over longer averaging times. They did not report RMSEs or R^2. They noted one advantage of using model results over ambient monitoring was the availability of PM component concentrations every day, versus one out of three. Hou et al. (2015) extended the application of CMAQ to the study of human health effects by using the emissions input data to calculate the sensitivity of $PM_{2.5}$ concentrations to EGU and non-EGU emissions from four regions of the U.S. The sensitivities were then used to estimate changes in mortality as a function of $PM_{2.5}$ exposure concentrations and sensitivity of mortality to regional EGU and non-EGU emissions. Bravo et al. (2012) simulated $PM_{2.5}$ over the eastern U.S. using a 12 km× 12 km grid with a normalized mean bias of 2.1% over the course of a year. However, $PM_{2.5}$ concentrations were underestimated by up to 27% in summer and by up to 32% in late fall. In a related study, Mannshardt et al. (2013) compared results using observations and CMAQ-estimated exposure concentration fields in a study of $PM_{2.5}$ and O_3 associations on emergency hospital emissions in three counties of New York City for 2002−2006. CMAQ was run for the eastern U.S. using 12 km grids and used as input to a human exposure model, SHEDS-PM.

Results from CTMs can be biased and subject to various errors due to inputs and model parameterizations, but factors leading to simulation errors continue to be identified and reduced [e.g., Yu et al. (2014); Barsanti et al. (2013); Baek et al. (2011); Foley et al. (2010)]. For example, PM chemistry modules in CMAQ have been added and revised to address limitations in modeling secondary organic PM formation and nitrate chemistry. Nonetheless, biases and errors persist that may have weekly and seasonal trends due to limitations in emission inventory specifications and chemical and meteorological inputs, respectively. Nolte et al. (2015) compared MOUDI measurements of PM size distribution with

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predictions of size distribution (ranging from 0.05 to 20 μm) for several PM components (SO_4^{2-}, NO_3^-, NH_4^+, Na^+, Cl^-, Mg_2^+, Ca_2^+, K^+) at different sites. Nolte et al. (2015) observed discrepancies between the modeled and monitored size distributions where the emissions data were not accurate. Typically, where data were omitted from the NEI, modeled size fractions were negatively biased so that exposure concentrations would be underestimated for those size fractions. Differential bias may also be observed across regions in space. Many such biases can be corrected for using adjustment factors based on comparisons of simulation results with observational data.

The dearth of ambient UFP observations, given that necessary instrumentation is not standard to routine monitoring networks (Section 2.4.5), has limited development and validation of CTMs at this size fraction. UFPs are derived from both direct emissions as well as atmospheric nucleation, and they coagulate on shorter time scales than larger particles (Section 2.3.4). Their concentrations can vary rapidly, and there is an observed steep spatial gradient in NC near sources, e.g., within a few hundred meters of highways (Karner et al., 2010; Zhou and Levy, 2007), suggesting finer resolution modeling should be used when using models to estimate exposure fields for UFPs. The lack of emissions information on UFPs also complicates CTM development. Hu et al. (2014a) and Hu et al. (2014b) developed source-based CTMs to predict $PM_{0.1}$ mass concentration surfaces for estimation of exposure concentrations that were used in an epidemiologic study by Ostro et al. (2015). The model included emissions, advection, diffusion, wet deposition, and dry deposition, but it omitted gas-to-particle phase chemistry, gas-to-particle phase conversion, nucleation, and coagulation. Hu et al. (2014b) used a 4 km × 4 km grid, which creates uncertainties because it is larger than the spatial scale over which UFPs evolve. They noted the need for either fine grid resolution or a subgrid scale model such as large eddy simulation to capture finer-scale dynamics. Hu et al. (2014b) reported Pearson $R = 0.92$ for comparison of $PM_{0.1}$ mass concentration predictions with measurements and Pearson $R = 0.94$ for comparison of $PM_{0.1}$ EC mass concentration predictions with measurements. Bias was not reported, but the authors noted that model performance degrades for $PM_{0.1}$ mass concentration >4 μg/m^3 or <1 μg/m^3 and for $PM_{0.1}$ EC mass concentration >1 μg/m^3 or <0.2 μg/m^3. Using SEARCH data to evaluate CMAQ performance for application in epidemiologic studies, Park et al. (2006) found that CMAQ did not capture UFP dynamics well, finding biases of an order of magnitude and more in NC. Elleman and Covert (2010, 2009a), and Elleman and Covert (2009b) also found that CMAQ did not accurately predict UFP numbers. They linked the biases to the treatment of particle nucleation, emissions estimates, and how the size distribution is captured. Stanier et al. (2014) developed a nonlinear, Lagrangian trajectory model designed to capture the size distribution of UFPs, and applied it to simulate UFPs in the Los Angeles area for a period when more detailed observations existed. They were able to reproduce NC within a factor of two 94% of the time at the four sites being used in the evaluation. In a comparison of 12 different nucleation parameterizations, Zhang et al. (2010a) found that the predicted NC of Aitken mode particles can vary by three orders of magnitude. These recent efforts illustrate that the large uncertainties in UFPs are still a great limitation in applying CTMs to model UFP exposure concentration.

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Several new developments in CTM have made the technology more amenable for application in exposure assessment, such as improvements to the model through bias correction methods. However, several limitations still exist, including large grid sizes, uncertainties regarding emissions inputs, and uncertainties in modeling UFP. Specific modeling decisions must therefore be evaluated when CTMs are employed in epidemiologic studies.

3.3.2.4.2 Dispersion Modeling Applications for Exposure Concentration Estimation

Dispersion modeling has been performed to develop relatively fine resolution PM exposure concentration fields (Jerrett et al., 2005a). Dispersion models describe the relationship between emissions, meteorology and the resulting pollutant concentrations using algebraic relationships (e.g., the Gaussian Plume Equation), but they typically have limited ability to model chemistry (if any) (Holmes and Morawska, 2006). Examples of dispersion models include AERMOD, Research LINE-Source Model (or R-LINE), Community LINE-source Model (C-LINE), and California LINE Source Dispersion Model (CALINE) (Barzyk et al., 2015; Snyder et al., 2013; Cimorelli et al., 2005; Perry et al., 2005; Benson, 1992).

Model intercomparison has more recently focused on near-road dispersion modeling. Heist et al. (2013) conducted an intermodel comparison of AERMOD, CALINE, ADMS, and R-LINE for tracer (SF6) dispersion and found that the more recently developed ADMS and R-LINE exhibited lower error and better validation compared with CALINE and AERMOD. The models were each compared with results from a tracer study in Idaho Falls, ID (for open field and constructed barrier conditions) under different convective mixing conditions and near Highway 99 in Sacramento, CA and showed that ADMS, R-LINE, and both versions of AERMOD performed better than the CALINE models for both sites (Table 3-4). ADMS and R-LINE were further compared for near-neutral, weakly stable, convective, and moderately-to-strongly stable convective mixing conditions. At low concentrations (<1 pbb), both models exhibited a tendency for positive bias except for the moderately-to-strongly stable conditions, where both models exhibited some negative bias with more scatter. Chen et al. (2009) tested the performance of three dispersion models, CALINE4, CAL3QHC and AERMOD, at Sacramento, CA and London, U.K. regarding their application in modeling near road $PM_{2.5}$ concentrations. All three models produced R^2 values ranges from 0.85 to 0.90 comparing with measurement data (without adding background concentrations) in Sacramento, CA. However, the models perform less well at London, U.K. with R^2 value at around 0.03 without background concentrations due to the influence of street canyons on receptor performance.

Dispersion models are typically applied over smaller domains (near-source to urban) than CTMs (urban to global). For example, AERMOD is designed for simulating "near source" dispersion from point and area sources, and is most useful for assessing source impacts within 20 km of the source (Silverman et al., 2007), although it has been evaluated for distances up to 50 km for certain applications (Perry et al.,

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2005). R-LINE is used for line source modeling, and was originally evaluated by Snyder et al. (2013) for distances of 200 m, though applications have applied it to urban scale (Batterman et al., 2014). While AERMOD is designed to simulate point and area/volume sources, it has been used to estimate the impacts of road networks by approximating road segments as area or volume sources (Isakov et al., 2014; Chen et al., 2009). Rowangould (2015) proposed a new dispersion modeling method for urban environments by breaking the city into coarse and fine grid cells (depending on the roadway density) and modeling dispersion from roadway sources in each roster in parallel. No validation was presented in the Rowangould (2015) paper.

Table 3-4 Comparison of dispersion models with data from a tracer study in Idaho Falls, ID and a near road study in Sacramento, CA and an UFP study in Somerville, MA and Chinatown in Boston, MA.

	Idaho Falls, ID		Sacramento, CA		Somerville, MA		Boston, MA	
Model	NMSE	*R*	NMSE	*R*	NMSE	R^2	NMSE	R^2
CALINE3	NR	NR	2.26	0.29	NR	NR	NR	NR
CALINE4	1.94	0.76	0.86	0.47	0.06	0.54	0.02	0.78
AERMOD-V	1.26	0.84	0.28	0.77	0.11	0.57	0.02	0.81
AERMOD-A	1.25	0.82	0.31	0.72	NR	NR	NR	NR
ADMS	1.14	0.88	0.20	0.78	NR	NR	NR	NR
R-LINE	0.96	0.85	0.34	0.75	0.13	0.58	0.02	0.81

NMSE = normalized mean squared error; NR = not reported, *R* = correlation (not specified if Pearson or Spearman); R^2 = coefficient of determination.

Sources: Data reproduced with permission of Heist et al. (2013); data reprinted with permission from Patton, AP, Milando, C, Durant, JL, Kumar, P. Assessing the suitability of multiple dispersion and land use regression models for urban traffic-related ultrafine particles. Environ Sci Technol. 2017;51:384-392. Copyright (2017) American Chemical Society. (Patton et al., 2017).

Several studies have used dispersion models at urban or neighborhood scales to estimate exposure concentrations. For example, Isakov et al. (2014) applied both AERMOD and R-LINE in Detroit, MI to estimate exposure concentrations to $PM_{2.5}$, EC, OC and pollutant gases at homes and schools of children with asthma participating in the Near Road Exposure of Urban Air Pollutants Study (NEXUS). CMAQ and kriging of observations were used to define regional air pollutant levels. Comparison between model results and measurement show reasonable performance with Pearson *R* range from 0.78 to 0.94 (daily average $PM_{2.5}$ concentrations) at different monitor sites. Simulated concentrations of PM are often used in conjunction with other estimates of regional PM because dispersion models are the more limited in spatial extent and so not designed for PM transport over large distances. For example, in an Atlanta application

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(Dionisio et al., 2013; Sarnat et al., 2013b), a variety of approaches were used to estimate exposure concentrations. One approach used AERMOD to model impacts of traffic emissions and added the resulting concentrations to background concentrations (developed from observations) to construct a high-resolution PM field for use in an epidemiologic study. Sarnat et al. (2013b) used the fine-scale resolution to help identify potential health disparities linked to socioeconomic status that were not apparent when using a single fixed-site monitor. Maroko (2012) used AERMOD to simulate $PM_{2.5}$ impacts from point sources in the New York City area to assess environmental justice issues. Dispersion models can also be used to simulate components of PM, assuming that they do not undergo a chemical reaction in the atmosphere. For example, Colledge et al. (2015) used AERMOD to estimate particulate manganese exposure in two Ohio towns.

A recent development in dispersion modeling is the inclusion of UFP when modeling PM dispersion in the vicinity of a road. Patton et al. (2017) evaluated CALINE4, R-LINE, and AERMOD for UFP transport near roads in the greater Boston, MA area (Somerville, MA and Chinatown, within Boston). They found similar performance among all three models (Table 3-4). Stanier et al. (2014) recognized that it is challenging to model UFP emitted from mobile sources, because the UFP size distribution rapidly evolves upon emission from vehicle tailpipes. They fit emissions factors based on existing data for cruising and acceleration of heavy-duty and light-duty vehicles, estimating across a size distribution down to 7 nm and correcting for coagulation and deposition. The emissions factors were incorporated into a dispersion term in the model. Modeled particle NC was compared with measured concentration at two sites within the Los Angeles, CA metropolitan area and showed underestimation of the model (below a factor of 1:2) at one location and modeled data within a factor of two at the other site. Stanier et al. (2014) propose that the model is suitable for estimating spatially resolved UFP exposure concentrations on a daily basis.

Dispersion modeling continues to be used in exposure assessment studies, often in conjunction with CTMs to provide fine-scale spatial resolution. Recent improvements have been made in modeling dispersion of traffic-related air pollution and applying dispersion models at urban scales. However, dispersion models are still limited when applied in dense urban environments since dispersion models are not designed to deal with complex built topography (Kakosimos et al., 2010), and they are limited in their ability to represent UFP transport because they are not designed to capture size-specific UFP dynamics (Stanier et al., 2014).

3.3.2.4.3 Hybrid Approaches

Although spatiotemporal and LUR models have been applied to estimate long-term (e.g., monthly and annual) spatially-resolved ambient PM exposure concentrations, these techniques are typically not as successful for short-term (e.g., hourly and daily) applications as they do not include the impacts of changing source emissions and meteorology. PM data from ambient monitors provide accurate information on temporal trends at monitoring sites but little information on spatial patterns.

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Emissions-based models provide spatial information consistent with emissions, chemistry, and meteorology but subject to limitations in the accuracy of these inputs as well as in the ability of models to simulate air pollution physical and chemical processes. "Hybrid" approaches that combine observational data with emissions-based model results are being developed and used to provide better estimates of single component and mixtures along with estimates of the associated uncertainties. These approaches range from rescaling model results to correction for known biases to combining observational and simulation data and optimizing spatiotemporal exposure concentration estimates.

Fusion of Model Outputs for Exposure Concentration Estimation

As noted above, CTMs by themselves typically have spatial resolution of 4 km or greater due to computational limitations, but they provide regional variations in PM (and PM component levels) and capture the formation of secondary PM, while dispersion models provide near-source impacts with a finer resolution. Given these complementary characteristics, it is natural to couple them (though care must be taken to not double count emissions) (Isakov et al., 2009).

Several recent studies have merged CMAQ with dispersion models. For example, Beevers et al. (2013) combined CMAQ results with the ADMS (a dispersion model) in London, England. They found that the combination could capture the spatial and temporal variations in air quality, with a mean bias of 0.6 $\mu g/m^3$ when comparing the model to monitors at five sites. Similarly, Zhai et al. (2016) combined R-LINE results with CMAQ-data fusion fields to estimate $PM_{2.5}$ exposure concentration fields for Atlanta, GA for a birth cohort study, with Pearson $R^2 = 0.72$ between the model and monitoring data with LOOCV normalized RMSE = 0.50 and normalized mean bias of 12%. A combined AERMOD-CMAQ application to New Haven, CT, was conducted (Lobdell et al., 2011; Isakov et al., 2009) to develop local scale (census block level) PM exposure concentrations in a base year (2001) and future years (2010, 2020 and 2030 to assess pollutant control programs). They noted the uncertainties due to model inputs, with coefficients of variation (standard deviation of concentration/mean concentration) ranging from 10–70% within different census tracts, but no estimates of model uncertainty with respect to $PM_{2.5}$ were provided. They linked their results to the HAPEM (Ozkaynak et al., 2008) and SHEDS (Isakov et al., 2009) exposure models, as described further in Section 3.3.4.

Another method of addressing the low spatial resolution of a CTM is to combine the model results with dispersion model results and LUR modeling output for exposure concentration. Wang et al. (2016) combined CTM with LUR using a hierarchical spatiotemporal modeling technique in which the 2-week average LUR-derived $PM_{2.5}$ concentration is modeled as a function of spatiotemporal trends and spatiotemporal residual terms, where the trend terms can be decomposed into an average and a spatially-varying trend (Keller et al., 2015). Wang et al. (2016) incorporated the CTM predictions into the spatially-varying trend term. The advantage of combining these two models is that the CTM is a mechanistic model employing principles of transport, dispersion, and atmospheric chemistry with finer temporal resolution (daily for this study), while the LUR offers fine-scale spatial resolution. The LUR

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was fit to fixed-site $PM_{2.5}$ monitoring data in AQS and from the MESA Air study and incorporated a variable for long-term average concentration derived from the CAL3QHCR near-road line source dispersion model. Wang et al. (2016) found that addition of the CTM to the spatiotemporal model of Keller et al. (2015) only produced a marginal improvement in the prediction ability of the model for capturing $PM_{2.5}$ exposure concentrations. Di et al. (2016b) combined GEOS-Chem simulations, based on a 28 km × 25 km grid, with land use and meteorological variables to improve resolution to 1 km × 1 km across the northeastern U.S. Di et al. (2016b) compared the model results with monitoring data when the GEOS-Chem model was used alone and when it was combined with land use and meteorological variables. Out-of-sample R^2 for $PM_{2.5}$ improved from 0.47 for GEOS-Chem alone to 0.85 for the hybrid model. Out-of-sample R^2 ranged from 0.13–0.33 for $PM_{2.5}$ components (EC, OC, NO_3^-, SO_4^{2-}, NH_4^+, dust, sea salt) for GEOS-Chem alone, and R^2 improved to 0.41–0.83 across the $PM_{2.5}$ components for the hybrid model.

Fusion of Chemical Transport Model Predictions with Surface Observation Data

To take greater advantage of the strengths of observational data and model simulations, various data fusion approaches have been developed and applied. Such model-data fusion approaches used in estimating exposure concentration fields for health studies have frequently used CMAQ.

Downscaling approaches have been used frequently in recent years to correct biases in CTM output. Berrocal et al. (2009) proposed a downscaling approach combining monitoring and CMAQ modeling data to improve the accuracy of spatially resolved O_3 model data. Specifically, a Bayesian model was developed to regress CMAQ model estimates of O_3 concentration on monitoring data, and then the regression model was used to predict concentrations using the CMAQ model results as an input field. Although the downscaling method was originally developed for to model O_3 concentration, this technique has since been applied for modeling $PM_{2.5}$ concentration surfaces and found to have low NMB (0.95%) with mean correlation between model output and monitoring data of 0.97 (Bravo et al., 2017). Berrocal et al. (2010) extended the approach to include two pollutants (ozone and $PM_{2.5}$) in a single modeling framework. Predictive mean absolute error (PMAE) for $PM_{2.5}$ concentration in the bivariate model was 2.3 μg/m^3, compared with observations at 65 monitoring sites. PMAE for $PM_{2.5}$ was 2.4 μg/m^3 for the comparison of the single-pollutant model with the monitoring sites. Berrocal et al. (2012) also added smoothing processes that incorporate spatial autocorrelation and correction for spatial misalignment between monitoring and modeled data. Bentayeb et al. (2014) applied a similar data assimilation method in which local measurements and elevation data were combined with CTM output in a geostatistical forecasting model. This algorithm was applied for $PM_{2.5}$, PM_{10}, NO_2, SO_2, C_6H_6, and O_3. For the years 1989–2008, correlation between assimilated $PM_{2.5}$ concentration and local observations at 2 km resolution ranged from Pearson $R = 0.12$ to 0.85, with correlations decreasing with year. Bentayeb et al. (2014) explained the low correlations by a small number of $PM_{2.5}$ monitoring stations producing anomalous data and low correlations between emissions and concentration data.

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