CHAPTER 3: SPATIAL VARIABILITY OF AVERAGE MONTHLY PATTERNS IN FINE AEROSOL SPECIES CONCENTRATIONS AND PARTICULATE EXTINCTION COEFFICIENTS

INTRODUCTION

In this section, the spatial variability in the seasonal patterns in aerosol composition and concentration are explored for the IMPROVE and STN networks. Additionally, the spatial variability in the seasonal patterns in particulate extinction coefficients is discussed and contrasted to those in aerosol concentrations for the IMPROVE network. Aerosol composition is influenced by both the nature of pollutant emissions and atmospheric characteristics that determine dispersion, transformation, and deposition. It seems likely that these influential factors would be similar during the same times of the year for relatively compact groupings of monitoring sites. Thus it may be reasonable to examine temporal and spatial patterns in annual and monthly averages on a regional basis. Regional groupings of sites in the IMPROVE network have been developed based on examination of seasonal patterns in aerosol composition and concentration in context of the sites' geographical locations and the expected spatial extent of regional fine aerosols [Sisler et al., 1993; Sisler et al., 1996; Malm et al., 2000; Malm et al., 2004]. The rural sites included in this analysis have been grouped into 26 previously defined regions; the three urban IMPROVE sites are analyzed individually. Variations in seasonal composition among sites within the regional groupings were not explicitly examined in this assessment but can be significant, especially for regions with significant terrain features and large emissions gradients and for sites at substantially different elevations. For example, Shining Rock and James River Face wilderness areas are both in the Appalachian region, but at James River Face the January sulfate concentration is about a factor of 1.8 greater than at Shining Rock, and the January organics are about a factor of 3 greater (see Appendix C). Only the 160 sites meeting the completeness criteria outlined in section 3.3 are included.

For the purposes of comparison, STN sites meeting the completeness criteria described in section 3.3 were grouped into regions based upon qualitative analysis of the aerosol data from 2000 through 2004. Stacked bar charts of monthly averaged data for the 69 sites with complete data for the five reconstructed fine mass (RCFM) components were examined for comparability in monthly patterns of aerosol composition and concentrations. Sites in the same geographic area that also had similar patterns in each of the five RCFM components, expressed both as concentrations and as fractional contribution to RCFM, were grouped into regions. The 69 sites were grouped into 27 regions, 14 of which contain only a single site. Quantitative comparability between sites in terms of monthly values or seasonal patterns was not explored.

Monthly and annual average aerosol species concentrations for the IMPROVE rural regions and urban sites are presented graphically in Figures 3.1, 3.3, and 3.5 as stacked bar charts of ammonium sulfate, organic and light-absorbing carbon, ammonium nitrate, and soil concentrations by month with the last bar representing the annual average. The graphics have been scaled to the maximum monthly concentrations. The STN regions are similarly summarized in Figures 3.2, 3.4, and 3.6. The fractional contributions to fine mass are shown in Figures 3.7, 3.9, and 3.11 for IMPROVE and in Figures 3.8, 3.10, and 3.12 for the STN. In these cases the y axis is the percent contribution to RCFM. Similar graphs of particulate extinction for IMPROVE and the STN are in Figures 3.13–3.21, where the y axis is either extinction in Mm^{-1} or percent contribution to reconstructed particulate extinction. Similar charts for the IMPROVE sites (data not regionally averaged) are in Appendix C. The regional seasonal and annual average reconstructed fine mass and extinction budgets are presented in tabular form in Appendix B.

3.1 SPATIAL VARIABILITY OF AVERAGE MONTHLY PATTERNS IN FINE AEROSOL SPECIES CONCENTRATIONS

In this section, the spatial variability in the seasonal patterns of each of the major aerosol types is explored. In particular, regional differences and urban-rural differences in the timing of maximum and minimum concentrations and fractional contributions are examined. Additionally, the spatial variability in the degree of seasonality as measured by the contrast between the maximum and minimum concentrations is also explored.

3.1.1 Fine Particle Ammonium Sulfate Mass

In general, ammonium sulfate concentrations tend to be higher during the summer and early fall months (June–September). The regional monthly maximum occurred during these months in ~70% of the IMPROVE and STN regions. The regions with maximum concentrations in the remainder of the year were primarily urban. The following urban regions had winter maxima: Boise, Minneapolis-St. Paul, Denver, Missoula, and the Wasatch Front. Spring maxima occurred in the urban regions of North Dakota, Florida, northwestern Nevada, central Minnesota, urban as well as rural Alaska and the rural regions of Hells Canyon, the northern Great Plains, the northern Rockies, and the Virgin Islands. The monthly minimums were less varied and occurred during winter in over 85% of the regions. The exceptions were almost exclusively urban, with Boise, urban Alaska, Denver, Minneapolis-St. Paul, Upper Michigan 2, and North Dakota all having fall minimums and Missoula having a spring minimum. The only rural region to not have a winter minimum was Hawaii, where the minimum occurred in summer.

Not surprisingly, because SO_2 emissions are highest there, the highest monthly ammonium sulfate concentrations occurred in the central eastern United States, where maximum rural and urban concentrations were comparable. Including both STN and IMPROVE regions, the maximum monthly concentration of ~11.4 $\mu g/m^3$ occurred in August in the IMPROVE Appalachian and Washington, D.C., regions and the STN Washington, D.C.-Philadelphia corridor region. The lowest monthly concentrations, 0.2–0.3 $\mu g/m^3$, occurred in the western rural regions of Oregon and northern California, the Great Basin, Alaska, the Sierra Nevada, and the Northwest. In general, in the eastern contiguous United States the regional maximum monthly concentrations ranged from 3 to 11.4 $\mu g/m^3$, and the minimums were 1.4–3.9 $\mu g/m^3$. In the western contiguous United States, including the low sulfate concentration regions of the northern Great Plains and Boundary Waters, the maximum monthly concentrations ranged from 4.1 to 6.6 $\mu g/m^3$ (0.8–2.8 $\mu g/m^3$, excluding the urban Wasatch Front in Utah and Los Angeles and San Diego in California) and the minimums ranged from 0.2 to 1.6 $\mu g/m^3$.

Ten of the 57 regions did not exhibit a distinct seasonal cycle in ammonium sulfate concentrations. These exceptions were Minneapolis-St. Paul, Boundary Waters, the northern

Great Plains, North Dakota, the Sacramento and San Joaquin valleys, and the Virgin Islands, which qualitatively appeared to have minimal seasonality and had ratios of the highest to lowest monthly ammonium sulfate concentrations of less than 2. The ratios of the maximum to the minimum monthly concentration ranged from 1.4 in Denver to 7.2 in the Sierra Nevada, with a median value of 2.5. The highest ratios, above 4.5, were all found in California in the Sierra Nevada, southern California, Los Angeles, San Diego, Oregon, northern California, and Death Valley regions. A higher degree of seasonality was present in a greater proportion, ~2/3 as compared to ~1/3, of the rural regions as compared to the urban regions. An above-median ratio was considered indicative of a higher relative degree of seasonality as compared to other regions.

There was greater variability in the timing of the maximum and minimum monthly ammonium sulfate percent contributions to RCFM as compared to the variability in the timing of maximum and minimum ammonium sulfate concentrations. The maximum occurred during summer in ~60% of the regions, and the minimum occurred in winter in ~75% of the regions. Many of the rural and urban regions in the southwestern, north-central, and southeastern United States exhibited minimal seasonality in the percent contribution of ammonium sulfate to RCFM. The highest percent contributions, 50–70%, occurred exclusively in the IMPROVE network. The regions with these high ammonium sulfate contributions included both regions with high (in the top quartile) ammonium sulfate concentrations–Washington, D.C., the mid-South, Southeast, Northeast, Ohio River valley, East Coast, and Appalachia—and those with low to moderate concentrations (in the bottom 25–50th quartiles)—Alaska, Hawaii, and the Virgin Islands.

3.1.2 Fine Particle Organic Carbon Mass

There was greater regional variability in the seasonality of organic mass by carbon concentration (OMC) as compared to ammonium sulfate. A double peak structure with both a summer and winter peak in OMC was observed in the seasonal cycle of OMC in some regions (see the STN Nevada region for an example). In general, the OMC mass concentrations are at a minimum in winter or spring, with ~80% of the regions having their minimum during these seasons. Approximately 70% of rural regions had winter minima. Hawaii and the Virgin Islands were the only rural regions to exhibit summertime minima. All of the western urban regions had spring minima, with the exception of Phoenix where the minimum occurred in summer. The eastern urban regions were more varied, with spring minima observed in ~40% of the regions, winter minima in ~30% of the regions, and either June or fall minima observed in several regions.

Maximum OMC concentrations tended to occur in summer; ~60% of the regions had maxima during this period, but maxima occur in all seasons, depending on region. While summer and fall monthly maxima occur in both urban and rural areas, spring maxima occur exclusively in rural regions, and winter maxima occur exclusively in urban regions and in the heavily polluted Columbia River Gorge region. Most western urban regions had OMC maxima between November and January; the exceptions were Los Angeles with an October maximum and Missoula and western Nevada with August maxima. In contrast, eastern urban areas typically had maxima between July and September. The exceptions to the eastern norm were Florida with a January maximum, Duluth-Superior with a June maximum, and the Southeast with a November maximum. The rural regions typically had maxima occurring between May and August; the regions with maxima outside of this timespan included the central Great Plains with an April maximum, the mid-South and the Virgin Islands with September maxima, and the Columbia River Gorge with a November maximum.

The highest monthly average OMC concentration of 28.39 $\mu g/m^3$ was in Missoula, Montana, during August. High concentrations were also found during the winter in the urban Sacramento and San Joaquin valleys, STN Phoenix, and San Diego at 23.74, 17.85, and 17.25 $\mu g/m^3$, respectively. The lowest minimum monthly concentrations of OMC were in the rural regions of Alaska, the Virgin Islands, Death Valley, the central Rockies, and Hawaii at 0.17, 0.26, 0.39, 0.46, and 0.48 $\mu g/m^3$, respectively. Overall, the maximum monthly concentrations ranged from 0.6 to 7.7 $\mu g/m^3$ in rural regions and from 5.3 to 28.4 $\mu g/m^3$ in urban regions, and the minimums ranged from 0.2 to 2.6 $\mu g/m^3$ in the rural areas and 3.0 to 9.5 $\mu g/m^3$ in urban areas.

A number of urban regions, including the Southeast, mid-South and Ohio River valley, Puerto Rico, and eastern Texas-Gulf coast regions, exhibited minimal seasonality with maximum to minimum OMC concentration ratios of less than 1.5. The maximum to minimum ratios ranged from 1.3 in the urban Southeast to 30 in rural Alaska, with a median value of 2.3. The highest ratios, those greater than 5, occurred in the following rural western regions: the central and northern Rockies, northern Great Plains, Death Valley, Oregon and North California, the Sierra Nevada, and Alaska. Similar to the patterns observed in ammonium sulfate concentrations, a higher degree of seasonality was present in a greater proportion, about two-thirds as compared to about one-third, of the rural regions as compared to the urban regions.

Approximately half of the regions scattered around the United States, including Alaska and Hawaii, exhibited minimal seasonality in the percent contribution of OMC to RCFM. Again, a lack of seasonality was much more common in urban regions than rural regions, with approximately two-thirds of the urban regions showing fairly constant contributions of OMC to RCFM throughout the year. The percent contributions of OMC tended to be at a minimum in winter and spring and at a maximum in summer, with a high degree of variability depending on region. The maximum monthly percent contribution ranged from 24% to 89% and the minimum ranged from 5% to 64%.

3.1.3 Fine Particle Light-Absorbing Carbon Mass

Similar to the OMC concentrations, maxima and minima in the monthly light-absorbing carbon concentrations were regionally variable. Western urban regions had spring or summer light-absorbing carbon (LAC) minima. The eastern urban regions were variable in the timing of the monthly minimum concentrations, with minima occurring in every season. However, spring minima were marginally dominant, occurring in ~40% of the eastern urban regions. The monthly minima also occurred in every season in the rural regions; springtime was again marginally dominant, with ~40 of the rural regions having their minima in this season. Peak LAC concentrations occurred more or less equally in summer, fall, and winter, with timing depending upon region; rural west Texas was the only region to have had a springtime maximum. Summer maxima were typical of rural regions, fall maxima were typical of eastern urban regions, and winter maxima were typical of western urban regions. Approximately one-third of the regions had minimum monthly OMC and LAC concentrations in the same month, and about half of the regions had maximums in the same month. The regions with temporally matched OMC and LAC minima were equally split between urban and rural, whereas about two-thirds of the regions with matched maxima were rural.

The maximum monthly LAC concentrations ranged from 0.06 to 2.66 $\mu g/m^3$ and the minimums ranged from 0.03 to 1.93 $\mu g/m^3$. The highest maximum monthly concentrations occurred at the urban sites Denver, Los Angeles, Phoenix (IMPROVE and STN), and Puerto Rico at 1.90, 1.94, 1.95 and 2.19, and 2.66 $\mu g/m^3$, respectively. The lowest minimum monthly concentrations occurred in rural Alaska, Virgin Islands, Hawaii, and Death Valley at 0.03, 0.04, 0.04, and 0.05 $\mu g/m^3$, respectively

The ratio of the maximum to minimum monthly LAC concentrations ranged from 1.3 in the urban Northeast to 6.8 in rural Alaska. The highest ratios, those greater than 4, occurred in Oregon and northern California, the Sierra Nevada, Death Valley, urban and rural Alaska, Phoenix, and the northern Rockies. The lowest ratios, those less than 1.4, occurred in the urban Northeast, urban and rural mid-South, North Dakota, Upper Michigan 2, and Puerto Rico. Approximately 35% of the regions exhibited minimal seasonality in LAC; all were in the central or eastern United States.

The timing of the minima and maxima in percent contribution of LAC to RCFM varied depending on region, with contributions dipping in spring or summer and peaking in fall or winter in most regions. The maximum monthly percent contributions ranged from 3% to 26% and the minimums ranged from 0.6% to 15%. Approximately a quarter of the regions exhibited minimal seasonality in the percent contribution of LAC; most were in the central and eastern United States.

3.1.4 Fine Particle Ammonium Nitrate Mass

While ammonium sulfate and rural organic carbon concentrations tend to peak during the summer months, ammonium nitrates are typically highest during the winter season, because the cooler winter season temperatures favor particulate nitrate over gaseous nitric acid equilibrium. The monthly maximums occurred in November–March in most regions with the following exceptions: maximums in late spring in the Great Basin, southern California, Hawaii, and Puerto Rico; in summer in Death Valley, the Virgin Islands, Oregon and northern California, and rural Alaska; and in October in Los Angeles, San Diego, and the IMPROVE Puget Sound site. If the assumption of ammonium nitrate is valid for these regions, then the warm season nitrate maxima are interesting, particularly in hot regions such as the tropics, Death Valley, and southern California. The minimums in monthly ammonium nitrate occurred between June and September in all regions with the following exceptions: the minima occurred in spring in San Diego; in late fall in Death Valley and the Northwest; and in summer in southern California, Los Angeles, Oregon and northern California, rural Alaska, the Great Basin, and the Virgin Islands.

The highest monthly average concentrations occur in the urban regions of Los Angeles, Sacramento, and the Wasatch Front at 19.9, 16.9, and 12.6 $\mu g/m^3$, respectively. The highest rural monthly averages occurred in the central Great Plains, Ohio River valley, and the mid-South at 3.1, 3.2, and 5.0 $\mu g/m^3$, respectively, and were approximately 4 to 6 times smaller than those in Los Angeles. The lowest monthly maximums, 0.2–0.3 $\mu g/m^3$, occurred in rural Alaska, Hawaii, Oregon and northern California, the Great Basin, and the northern Rockies. These regions also had some of the lowest monthly minimum ammonium nitrate concentrations at less than 0.15 $\mu g/m^3$.

The only regions that exhibited minimal seasonality in ammonium nitrate concentrations were Hawaii and Puerto Rico. The highest ratios between the maximum and minimum concentrations within a region occurred in the Boundary Waters, Hells Canyon, Boise, Missoula, central Minnesota, and Wasatch Front regions, where the ratios were between 11 and 33. The lowest ratios, less than 2, occurred in Puerto Rico, Hawaii, the Northwest, and the Great Basin. In contrast to the patterns observed in ammonium sulfate and OMC concentrations, a higher degree of seasonality was present in a greater proportion of the urban regions as compared to the rural regions. About two-thirds of urban sites had above-median ratios as compared to about one-third of rural regions with above-median ratios.

In most regions, on a fractional basis nitrates make their greatest contribution to RCFM from November to March, with most of the maximum contributions occurring during the winter months. The exceptions were Los Angeles and Hawaii, where the maximum ammonium nitrate contributions occurred during summer. The minimum nitrate contributions occurred in June–September, except in Hawaii, San Diego, and Los Angeles, which all had wintertime minima. The maximum monthly percent contributions ranged from 5% to 51% and the minimums ranged from 2% to 30%. The largest percent contributions, 40–50%, occurred in regions of California, the Midwest, and urban Idaho and Utah. The only regions with minimal seasonality were Puget Sound and Portland, Florida, and Puerto Rico.

3.1.5 Fine Particle Soil Concentrations

Several regions displayed minimal seasonality in fine soil concentrations; all were urban and included Puget Sound and Portland, Los Angeles, Denver, IMPROVE Phoenix, and STN Northeast. However, they were the anomaly, with most regions having clear seasonality, with minimum monthly soil concentrations in fall and winter and maximum concentrations in spring or summer. San Diego, where soil concentrations were at a minimum in June and at a maximum in November, was an exception to this general pattern. The Northwest and the Sacramento and San Joaquin valleys regions also had fall maxima. There was a west-to-east gradient in the timing of the soil maxima, with spring maxima more common in the West (including Hawaii and Alaska) and summer maxima more common in the East (including the Virgin Islands and Puerto Rico).

The highest maximum monthly soil concentrations, 3–4.5, $\mu g/m^3$, were found in the arid Southwest regions of Death Valley, southern Arizona, STN Phoenix, and urban west Texas and in the eastern noncontiguous United States regions of Puerto Rico and the Virgin Islands. The lowest maximum monthly soil concentrations, less than 0.5 $\mu g/m^3$, occurred in the rural noncontiguous United States regions of Alaska and Hawaii and in the Boundary Waters and upper Michigan regions. The northwestern United States had the lowest minimum monthly soil concentrations, less than 0.1 $\mu g/m^3$, in the regions of rural Alaska, the Northwest, Oregon and northern California, and the northern Rockies. Not surprisingly, the regions with the highest minima, 1–2.5 $\mu g/m^3$, were found in the arid southwestern regions of southern Arizona, IMPROVE/STN Phoenix, and urban west Texas.

The ratio of maximum to minimum soil concentrations was greatest in the Virgin Islands where it was 20. The ratios were also quite high, 10–15, in the northwestern United States regions of the Northwest, Oregon and northern California, the Great Basin, and the northern Rockies. The lowest ratios, less than 2, occurred in the urban regions of Puget Sound and Portland, IMPROVE Phoenix, Los Angeles, Denver, northwest Nevada, the Northeast, and Washington, D.C. Similar to the patterns observed in ammonium sulfate and OMC concentrations, a higher degree of seasonality was present in a greater proportion of the rural regions as compared to the urban regions. The breakdown was similar, with about two-thirds of rural sites having an above-median ratio as compared to about one-third of urban regions meeting the same criteria.

In most regions, on a fractional basis soil makes the greatest contribution to RCFM during spring and summer, with approximately half of the regions having maximum soil contributions in April. Minimum soil contributions occurred primarily from November through February; the only exceptions were San Diego, Los Angeles, Alaska, and Death Valley, where the minimums occurred during summer. Maximum fractional contributions ranged from 4% in upper Michigan to 70% in the Virgin Islands. Minimum contributions ranged from 1% in the Sacramento and San Joaquin valleys to 27% in southern Arizona.

The broad-scale regional and temporal trends in the soil concentrations are indicative of large-scale transport mechanisms rather than local wind-blown mechanisms. A number of researchers have documented the impact of Saharan dust during the spring–summer months in the Virgin Islands and in the southeastern United States [Perry et al., 1997; Prospero et al., 2002]. The elevated spring–summer soil and the increasing southeast gradient over the eastern United States are consistent with this region being impacted by North African dust. It is also known that the western United States is periodically impacted by large dust plumes originating in Asia [Husar et al., 2001; VanCuren and Cahill, 2002] during the spring season. The widespread elevated springtime dust from northern Nevada to Texas is an indication of long-range-transport dust, particularly in the mountainous regions where local origins of dust are expected to be low.

In addition to potential Asian dust influences, the western United States is affected by local dust sources. The western United States and Mexico have three large dust source regions [Prospero et al., 2002], one located west and southwest of Salt Lake City, Utah, a second defined by the Salton Trough of southernmost California and northern Mexico, and the third in Mexico just south of the United States-Mexico border in the southern Mimbres Basin. The dust activity from these source regions begins in April–May and peaks in June–July, with dust extending from west Texas to the Mogollon Rim and to the Great Basin. The Owens Valley in California and eastern Washington are also important dust sources. The timing and locality of the highest fine soil aerosol concentrations are similar to those of these North American dust sources.

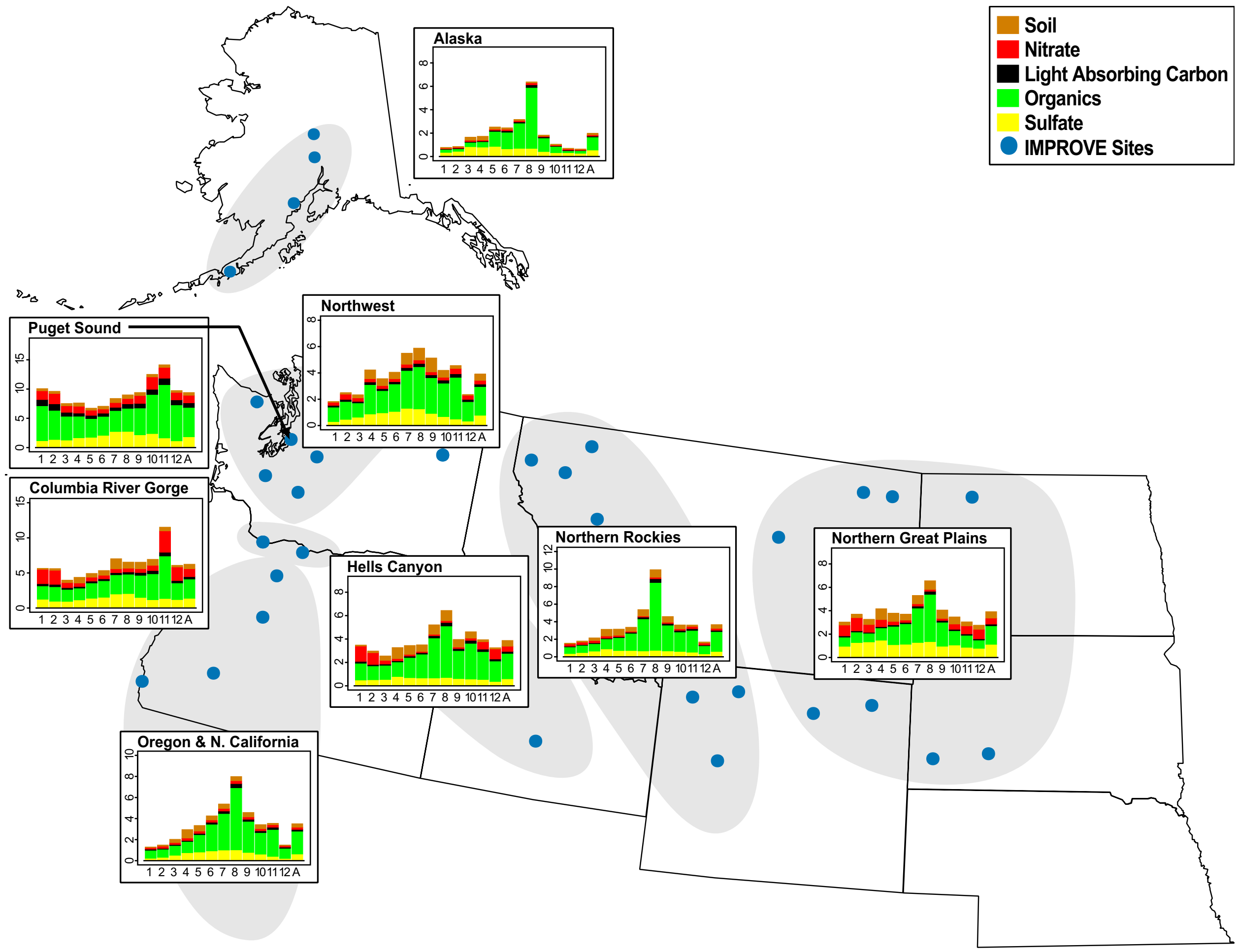

Figure 3.1. Map of stacked bar charts of monthly mean concentrations (µg/m^3) of fine aerosol species in the northwestern U.S. regions of the IMPROVE network.

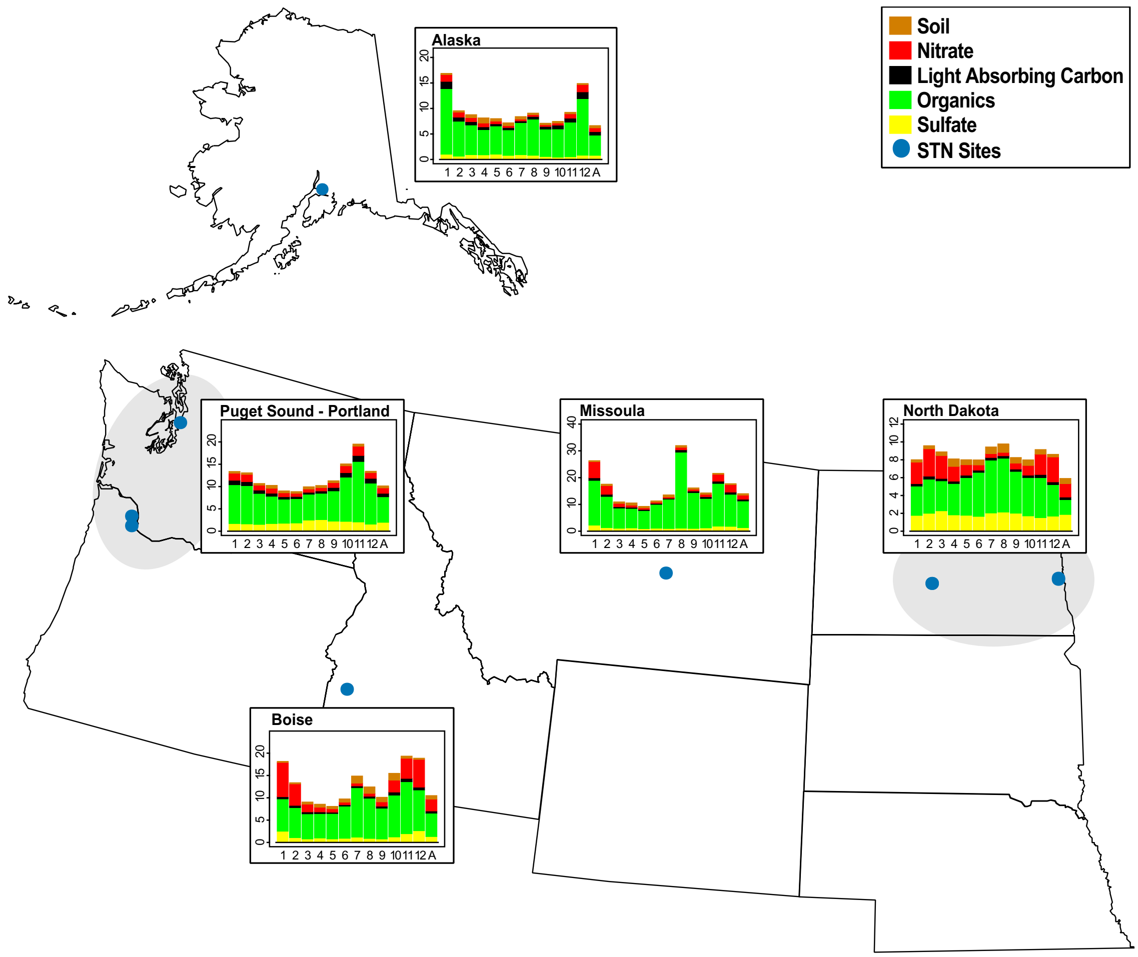

Figure 3.2. Map of stacked bar charts of monthly mean concentrations (µg/m^3) of fine aerosol species in the northwestern U.S. regions of the STN network.

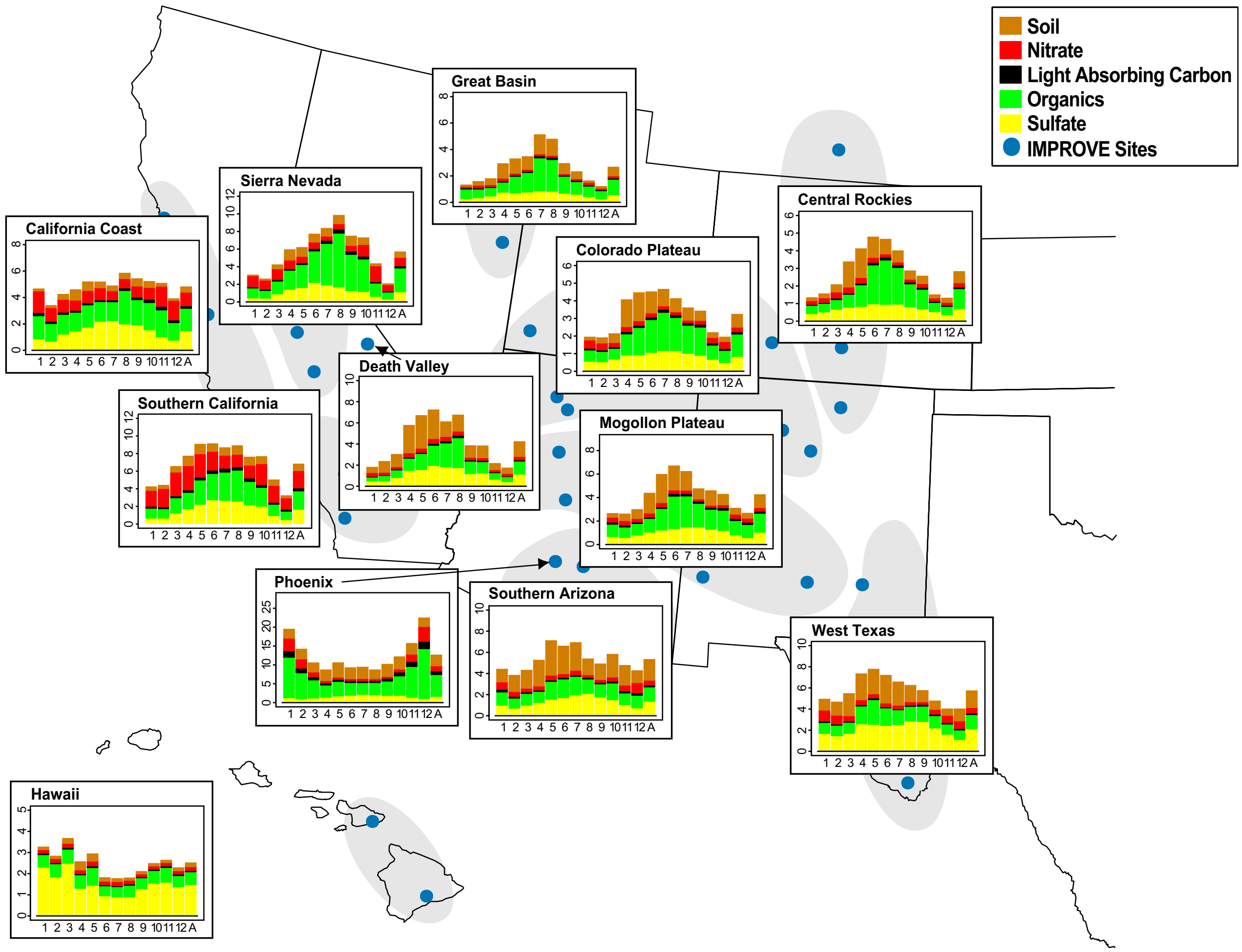

Figure 3.3. Map of stacked bar charts of monthly mean concentrations (μg/m^3) of fine aerosol species in the southwestern U.S. regions of the IMPROVE network.

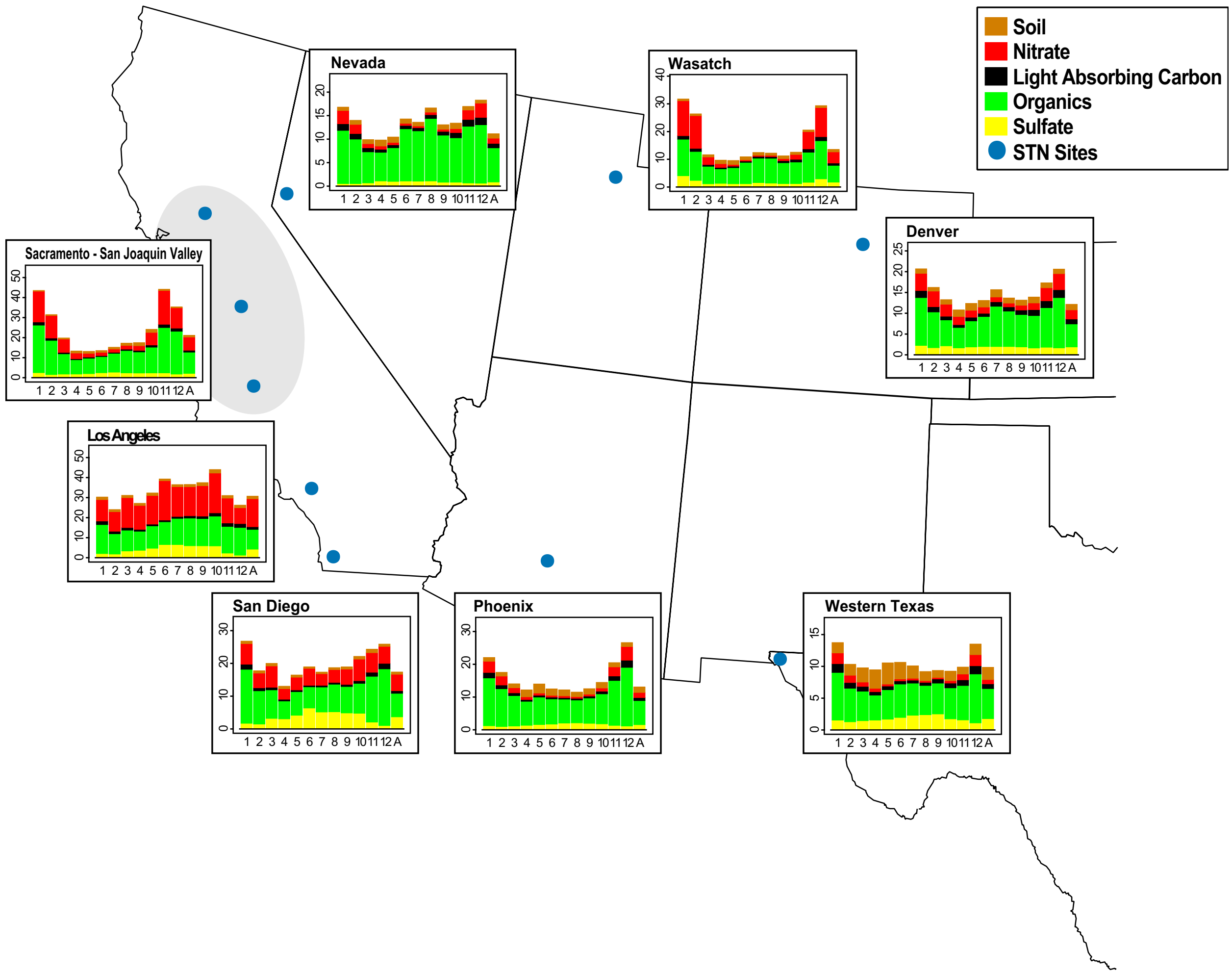

Figure 3.4. Map of stacked bar charts of monthly mean concentrations (μg/m^3) of fine aerosol species in the southwestern U.S. regions of the STN network.

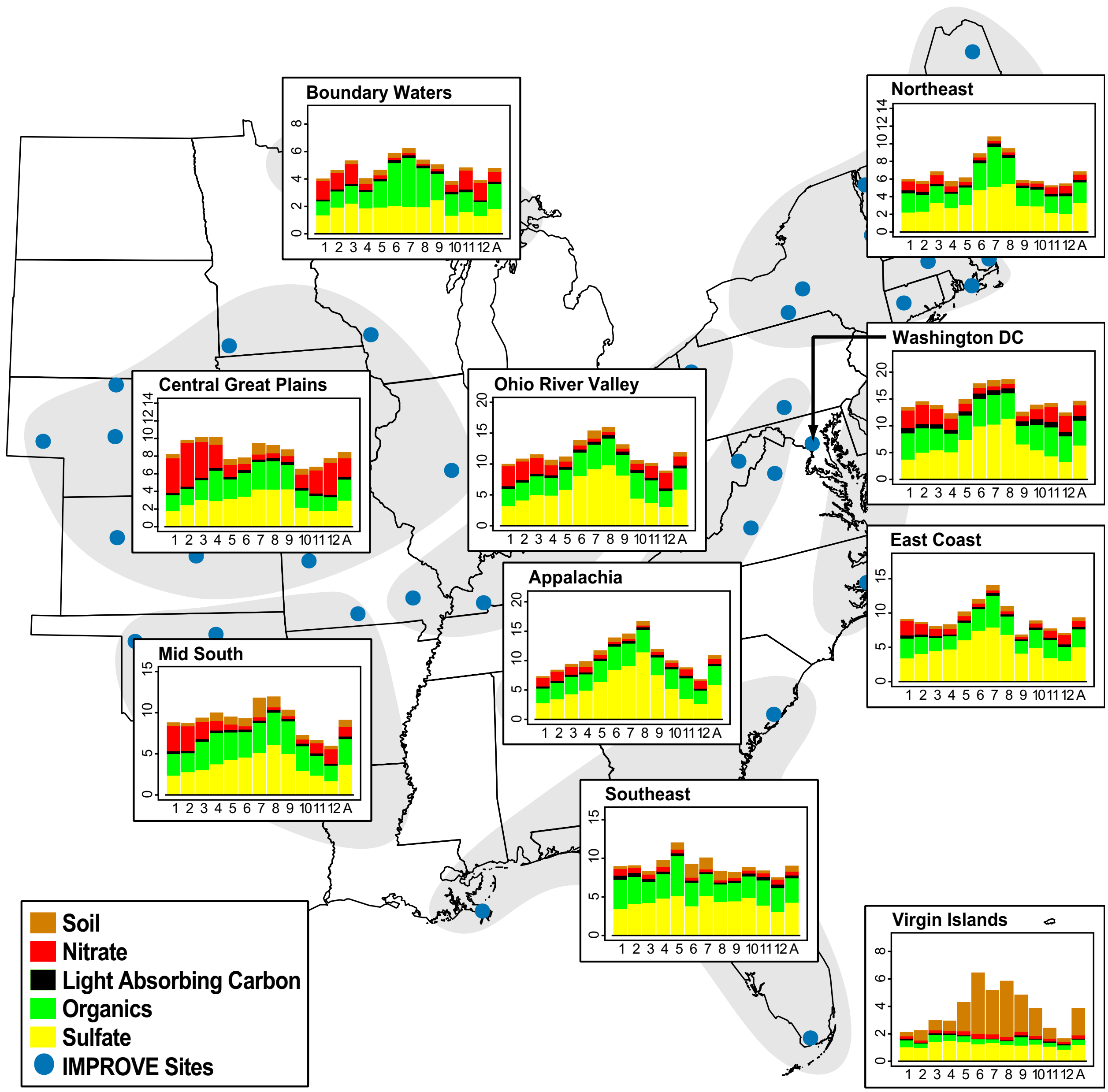

Figure 3.5. Map of stacked bar charts of monthly mean concentrations (μg/m³) of fine aerosol species in the eastern U.S. regions of the IMPROVE network.

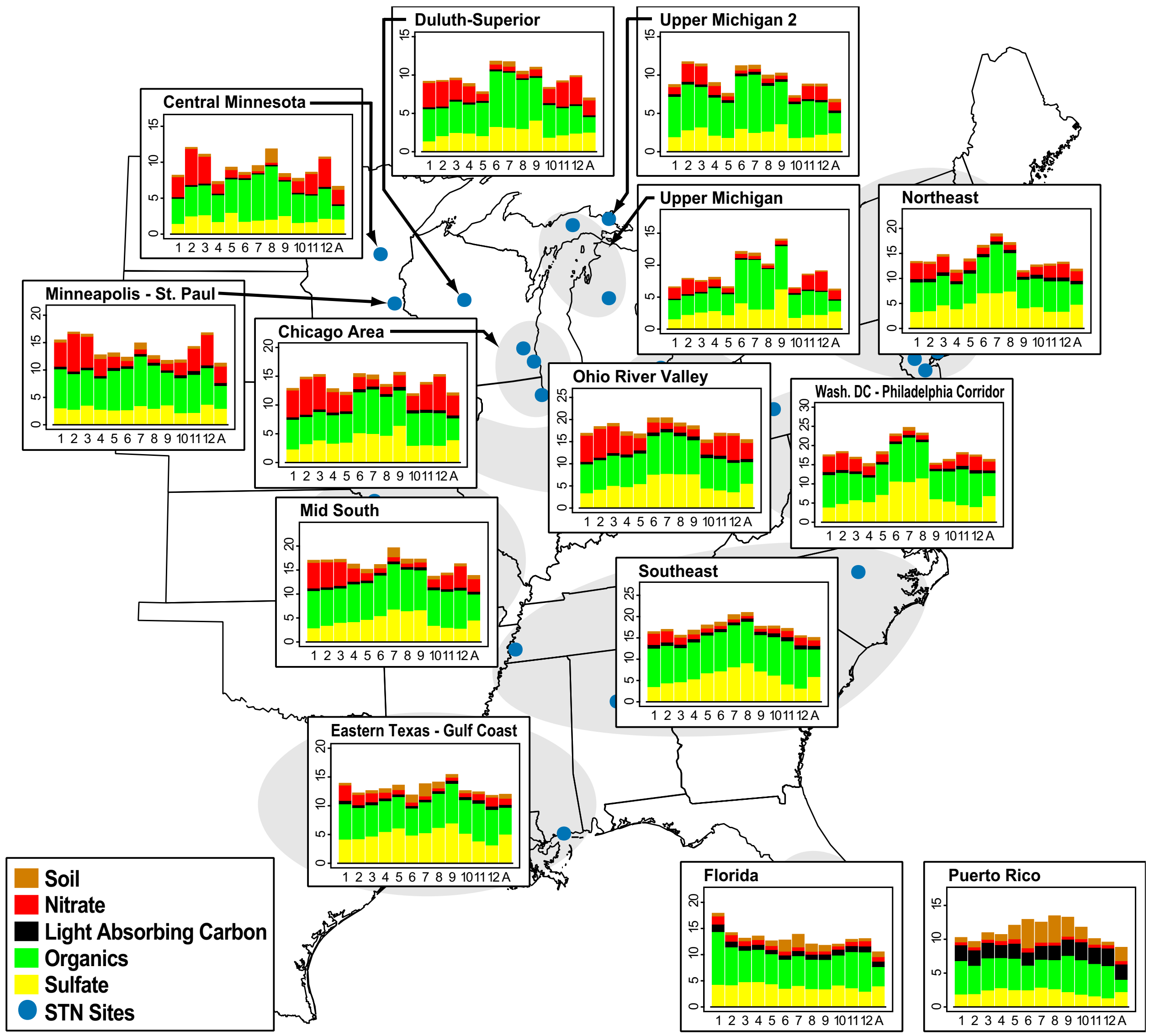

Figure 3.6. Map of stacked bar charts of monthly mean concentrations (μg/m^3) of fine aerosol species in the eastern U.S. regions of the STN network.

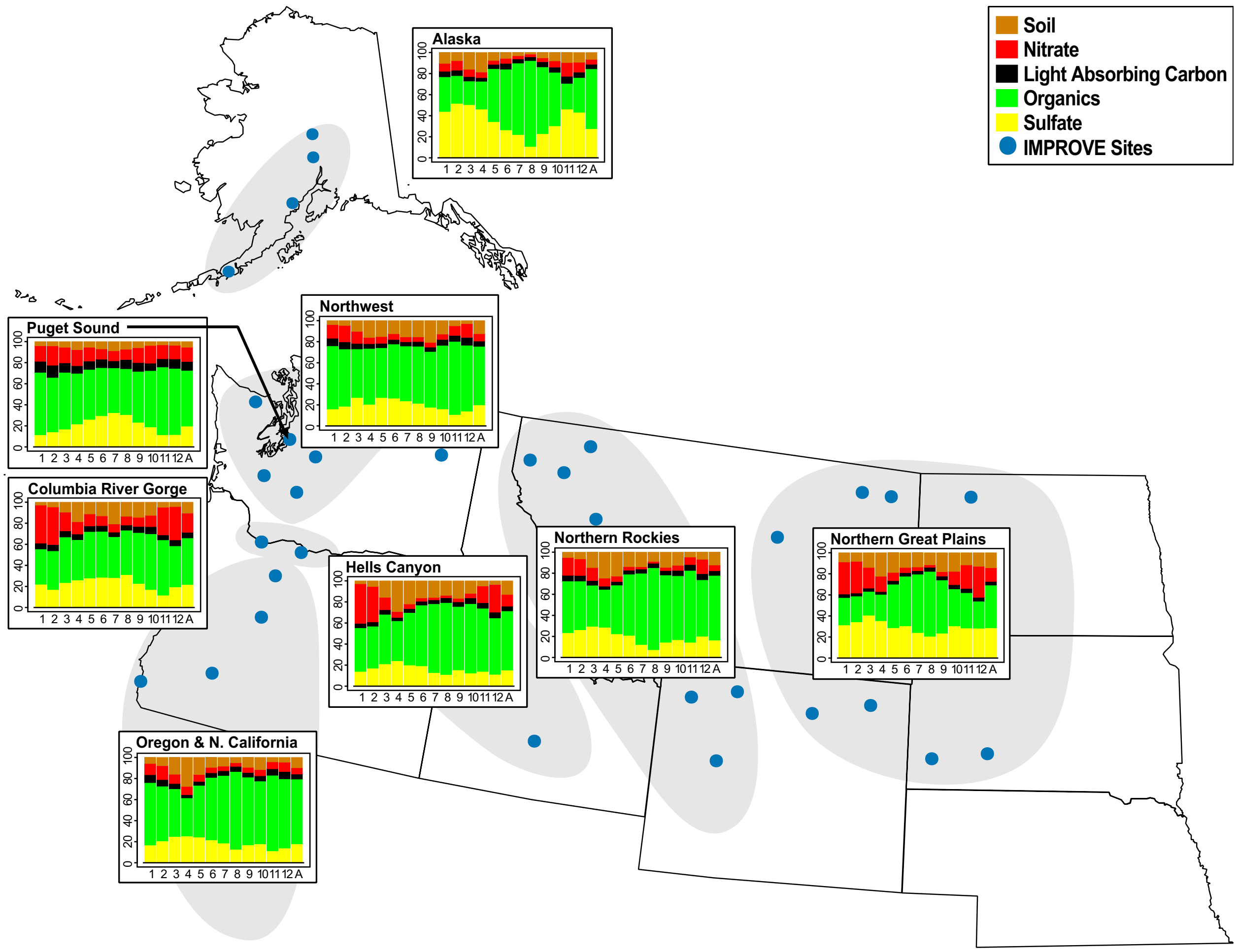

Figure 3.7. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the northwestern U.S. regions of the IMPROVE network.

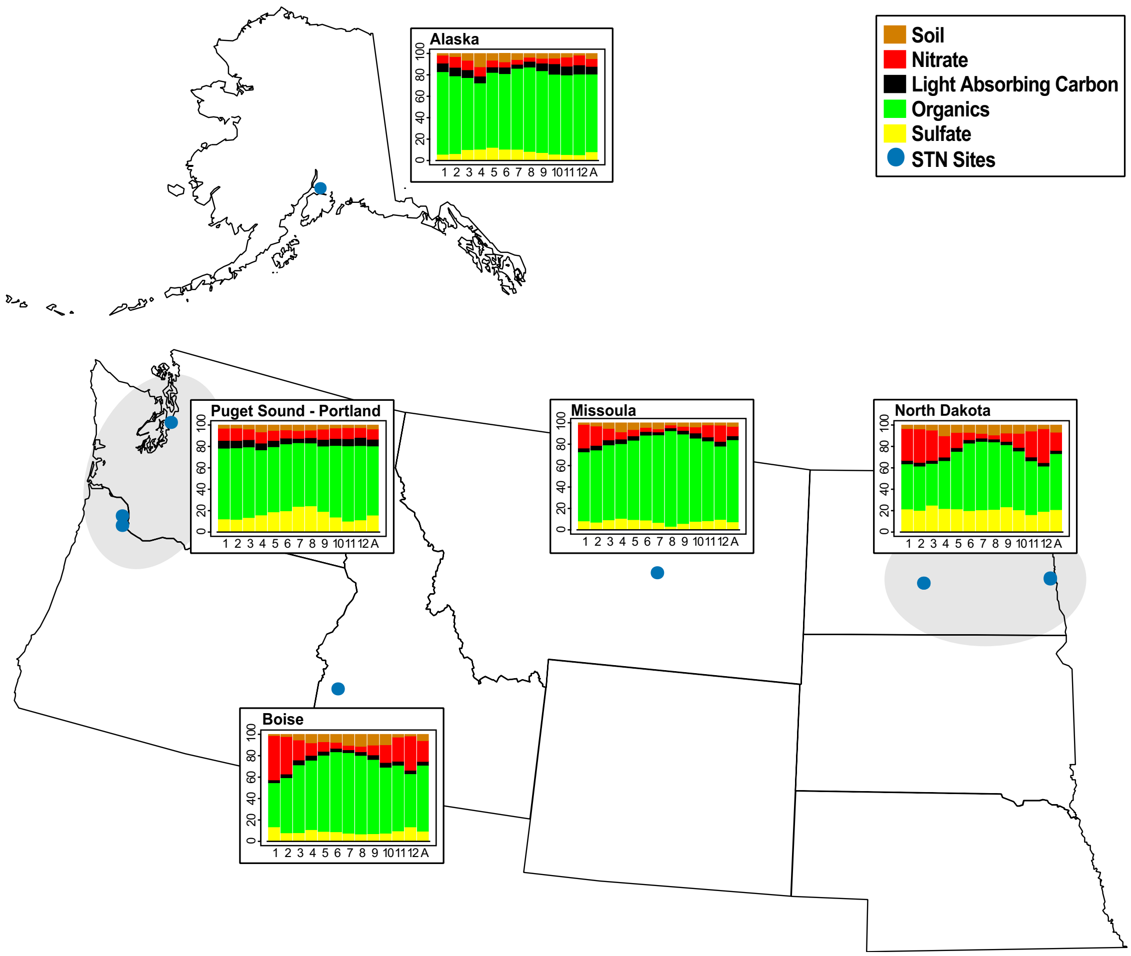

Figure 3.8. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the northwestern U.S. regions of the STN network.

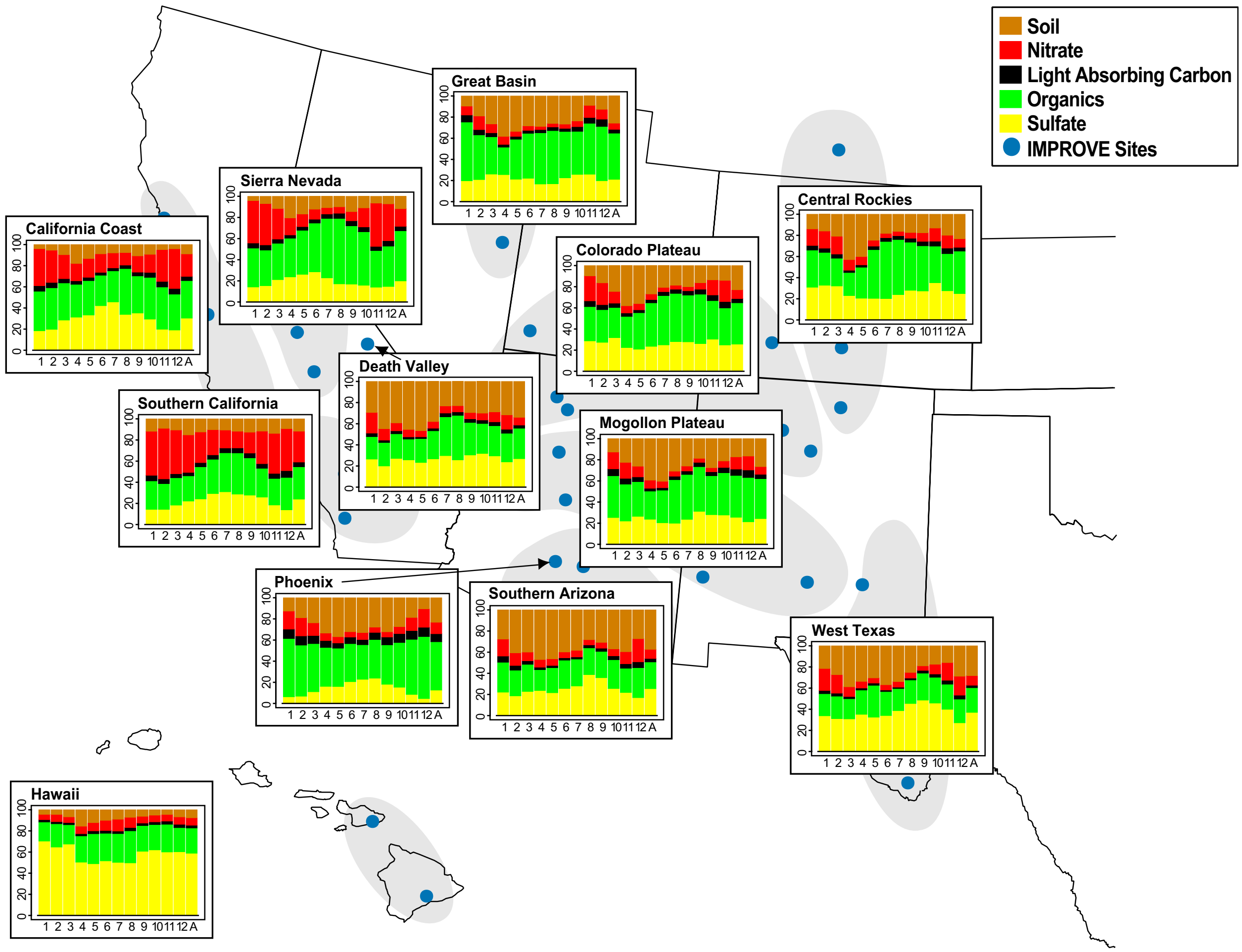

Figure 3.9. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the southwestern U.S. regions of the IMPROVE network.

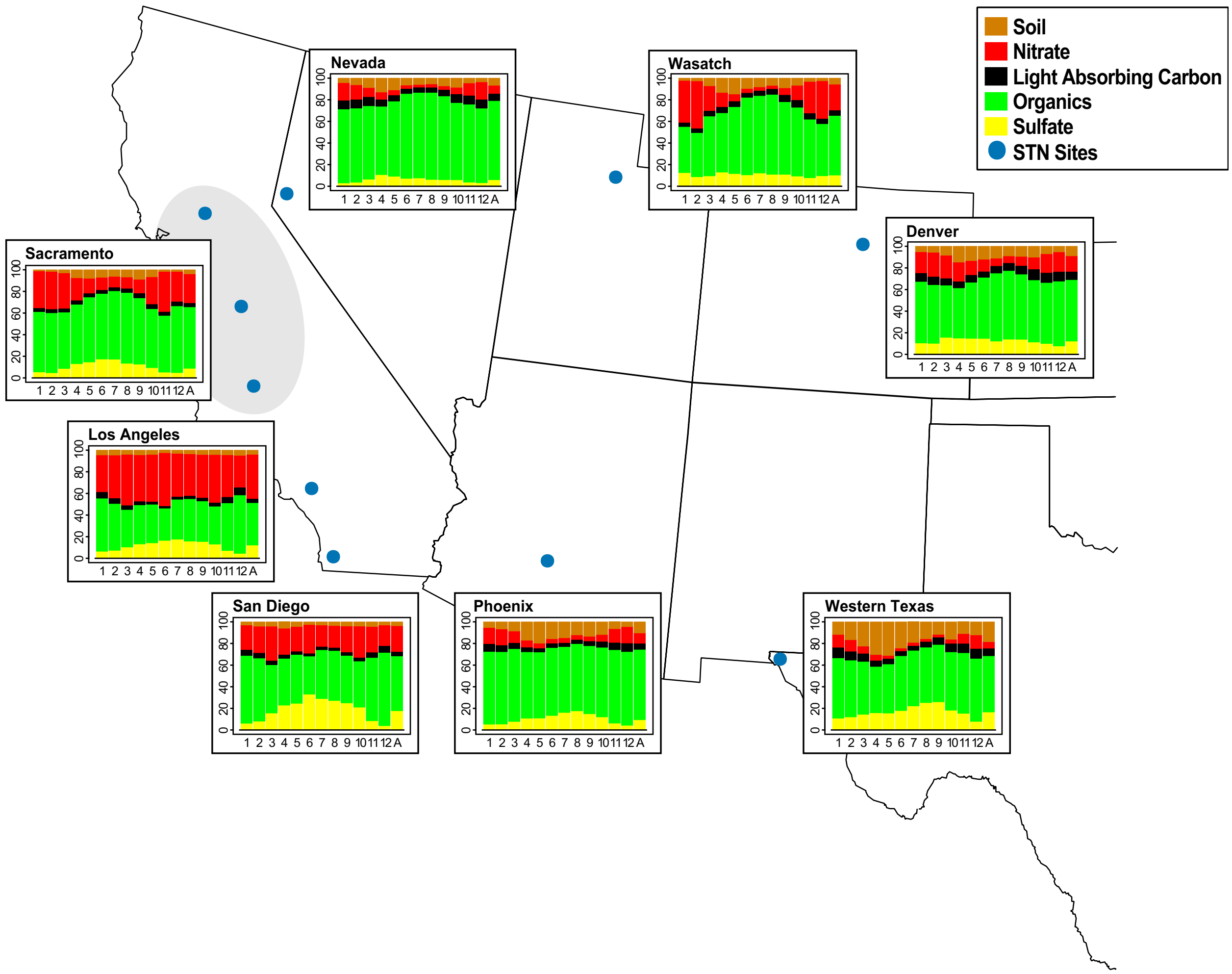

Figure 3.10. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the southwestern U.S. regions of the STN network.

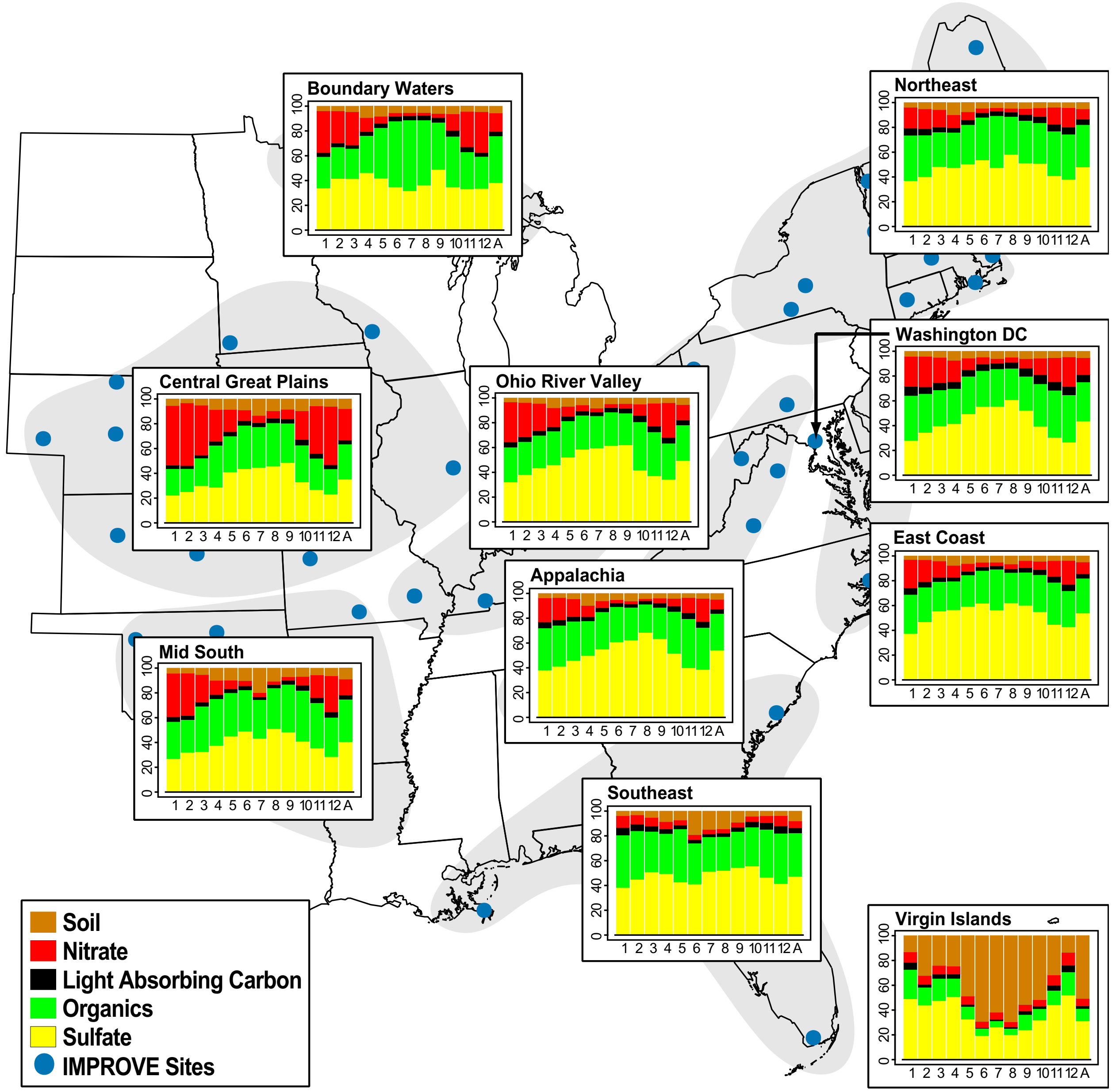

Figure 3.11. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the eastern U.S. regions of the IMPROVE network.

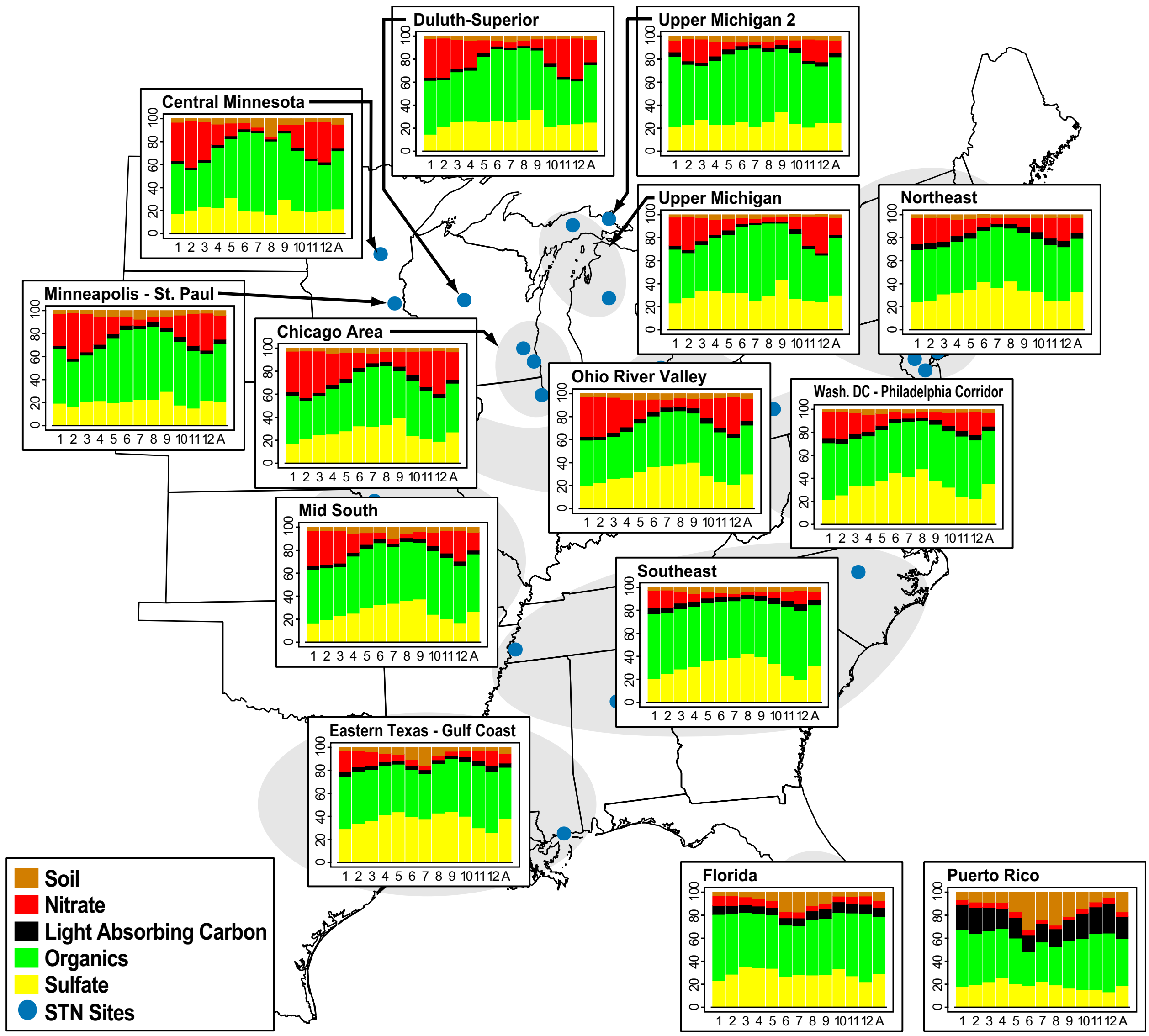

Figure 3.12. Map of stacked bar charts of monthly percent contribution to reconstructed fine mass (%) of fine aerosol species in the eastern U.S. regions of the STN network.

3.2 SPATIAL VARIABILITY OF AVERAGE MONTHLY PATTERNS IN PARTICULATE EXTINCTION COEFFICIENTS

In this section, the spatial variability in the seasonal patterns in particulate extinction and how they differ from aerosol mass concentration are explored for the IMPROVE network. Spatial and temporal patterns in the reconstructed particulate extinction are similar to those observed for aerosols since reconstructed particulate extinction is calculated from aerosol concentrations. However, because specific scattering of sulfates and nitrates is larger than other fine aerosols because of associated water, light-absorbing carbon has relatively high specific extinction, and coarse particle scattering contributes to total particulate extinction, the extinction budgets are somewhat different from fine aerosol budgets. Additionally, the temporal and spatial trends in relative humidity further modify the patterns observed in aerosol concentrations. Coarse mass (CM) is not measured by the STN so the seasonal patterns in the particulate extinction components are explored without an analysis of the total particulate extinction budget.

3.2.1 Fine Particle Ammonium Sulfate Extinction

In general, the seasonal patterns in ammonium sulfate extinction were similar to those in ammonium sulfate mass concentrations, the main difference being enhanced fractional contributions in extinction. Additionally, the seasonality, as indicated by the ratio of the maximum to the minimum monthly average, was increased in extinction in about one-third of the rural regions as compared to mass concentrations. The seasonality in relative humidity can act to either enhance or suppress the seasonality in ammonium sulfate concentrations. The rural regions where extinction had a slightly higher degree of seasonality (an increase of less than 1 in the max/min ratio) were primarily in the eastern United States. The only rural regions to show a large (greater than 1) change in the max/min ratio were all in the West and had larger max/min ratios in ammonium sulfate concentrations compared to extinction coefficients. The opposite was true in the urban regions where seasonality was increased in extinction for approximately 65% of both eastern and western regions. The only two regions to have a large change in the max/min ratio were the Wasatch Front in Utah and Boise, Idaho, both of which had greater seasonality in extinction coefficients.

For nearly all IMPROVE regions, both the maximum and minimum monthly contributions of ammonium sulfate to reconstructed particulate extinction were greater than the maximum and minimum contributions to reconstructed fine mass. The exceptions were Phoenix and the Virgin Islands, where the maximum contributions to concentrations were greater, and the Great Basin and Mogollon Plateau, where the minimum contributions were greater.

In most regions the maximum and minimum monthly values in ammonium sulfate mass concentrations and extinction coefficients occurred within a month of each other. The timing of maximum and minimum contributions to reconstructed particulate extinction and mass was also similar for most IMPROVE regions; comparison was possible for the STN regions. In several regions, the timing of the maxima or minima differed by more than 2 months when comparing mass concentration and extinction coefficients, with no real impact on the overall seasonal pattern. However, a few regions had markedly different seasonal patterns in ammonium sulfate extinction as compared to mass concentrations. Rural Alaska had a spring peak in ammonium sulfate mass concentrations but a summertime peak in extinction because of the lower springtime f(RH) values. Columbia River Gorge, Puget Sound, and Portland went from a summertime peak in concentrations to a fall peak in extinction due to the comparably higher f(RH) values that are at a minimum in summer in this region. In the Great Basin region, where f(RH) is also at a minimum in summer, the ammonium sulfate maximum shifted from summer in the mass concentration to spring in extinction. In the Virgin Islands, there is both a winter and spring peak in the fractional contribution of ammonium sulfate to reconstructed fine mass and reconstructed particulate extinction. The maximum percent contribution of ammonium sulfate to RCFM occurred during the winter peak, whereas it occurred during the spring peak for reconstructed particulate extinction.

The rural maximum monthly ammonium sulfate extinction coefficient ranged from 4.3 Mm^{-1} in the Great Basin to 121.2 Mm^{-1} in Appalachia, and rural monthly minimums ranged from 1.7 Mm^{-1} in the Great Basin to 31.1 Mm^{-1} in the Southeast. The urban regional monthly maximum ranged from 5.8 in northwest Nevada to 110.0 in the Washington, D.C.-Philadelphia corridor; the minimum ranged from 3.0 in Boise Idaho to 33.2 in the Ohio River valley. Maximum monthly ammonium sulfate contributions to reconstructed particulate extinction ranged from 20% in Phoenix to 82% in Appalachia, and the minimum contributions ranged from 5% in Phoenix to 55% in Hawaii.

3.2.2 Fine Particle Organic Carbon Extinction

The seasonal patterns in OMC extinction were the same as for OMC concentration because no humidity dependence for organics was considered. However, the seasonal patterns in percent contribution of OMC to extinction were influenced by the seasonality of the humidity-impacted terms of reconstructed particulate extinction. In contrast to ammonium sulfate, the contributions of OMC to reconstructed particulate extinction were generally reduced as compared to its contributions to reconstructed fine mass in the IMPROVE regions; comparison was possible for the STN regions. The only exception was southern Arizona where very low relative humidity suppressed the growth of sulfate and nitrate, which resulted in both the maximum and minimum monthly OMC contributions to extinction being higher by one to two percentage points than for the maximum and minimum contributions to mass. There were several regions where the timing of the minimum or maximum contribution differed by more than 2 months when comparing seasonal patterns in reconstructed particulate extinction to those in reconstructed mass. However, only Hells Canyon exhibited a noticeable change in the overall seasonal pattern, with a shift from a springtime to a winter minimum in percent OMC contribution.

The rural maximum monthly OMC extinction coefficients ranged from 2.4 Mm^{-1} in the Virgin Islands to 31.0 Mm^{-1} in the northern Rockies, and monthly minimums ranged from 0.7 Mm^{-1} in Alaska to 10.4 Mm^{-1} in the rural Ohio River valley. The urban maximum ranged from 13.1 Mm^{-1} in North Dakota to 102.5 Mm^{-1} in Missoula, and the minimum ranged from 0.9 Mm^{-1} in upper Michigan to 27.1 Mm^{-1} in Los Angeles. Maximum monthly contributions to reconstructed particulate extinction ranged from 11% in the Virgin Islands to 70% in the northern Rockies, and the minimum contributions ranged from 4% in the Virgin Islands to 27% in Phoenix.

3.3 Fine Particle Light-Absorbing Carbon Extinction

Similar to OMC, the seasonal patterns in LAC extinction were the same as for LAC mass concentration because no humidity dependence for LAC was considered. In all IMPROVE regions where comparison was possible, the maximum and minimum percent contributions of LAC were greater in reconstructed particulate extinction than in reconstructed mass. While there were several regions where the timing of the minimum or maximum contribution differed by more than 2 months when comparing seasonal patterns in contributions to reconstructed particulate extinction to those in reconstructed mass, none had a noticeable impact in the overall seasonal pattern in LAC contributions.

The rural maximum monthly LAC extinction coefficients ranged from 0.8 Mm^{-1} in Hawaii to 5.7 Mm^{-1} in the Ohio River valley, and monthly minimums ranged from 0.3 Mm^{-1} in Alaska to 3.8 Mm^{-1} in the Ohio River valley. The urban maximum ranged from 2.6 Mm^{-1} in Duluth-Superior to 21.9 Mm^{-1} in Phoenix, and the minimum ranged from 1.5 Mm^{-1} in upper Michigan to 8.6 Mm^{-1} in Los Angeles. Maximum monthly contributions to reconstructed particulate extinction ranged from 4% in Hawaii to 17% in Phoenix, and the minimum contributions ranged from 1% in the Virgin Islands to 10% in Phoenix.

3.3.1 Fine Particle Ammonium Nitrate Extinction

Like ammonium sulfate, the seasonal patterns in ammonium nitrate extinction were typically similar to those in ammonium nitrate mass concentrations, with the main difference being enhanced fractional contributions in extinction. For ammonium nitrate, the seasonality as indicated by the ratio of the maximum to the minimum monthly average was increased in extinction as compared to concentrations in approximately two-thirds of the rural regions and half of the urban regions. Many of the urban and rural western regions had significantly larger max/min ratios (greater than 1 increase in ratio) in extinction as compared to concentration; this was not true in the East. The only large changes in ratio, where the seasonality was higher for concentration, occurred in the eastern urban regions of central Minnesota and Minneapolis-St. Paul.

For nearly all IMPROVE regions, both the maximum and minimum monthly contributions of ammonium nitrate to reconstructed particulate extinction were greater than the maximum and minimum contributions to reconstructed fine mass. The exceptions were the Virgin Islands where the maximum contribution and Phoenix where the minimum contribution to reconstructed mass were greater.

In most regions, the maximum and minimum monthly values in ammonium nitrate mass concentrations and extinction coefficients occurred within a month of each other, as did the maximum and minimum contributions to reconstructed particulate extinction and mass. In several regions, the timing of the maxima or minima differed by more than 2 months when comparing mass concentration and extinction coefficients with no real impact on the overall seasonal pattern. However, in Death Valley the seasonal pattern in extinction coefficients with a winter peak and late fall minimum was quite different from the pattern in mass concentrations of a summer peak and late fall minimum.

The rural maximum monthly ammonium nitrate extinction coefficients ranged from 1.4 Mm^{-1} in the Great Basin to 43.2 Mm^{-1} in the Columbia River Gorge, and monthly minimums ranged from 0.5 Mm^{-1} in the Great Basin to 8.2 Mm^{-1} in southern California. The urban maximums ranged from 1.7 Mm^{-1} in urban west Texas to 16.9 Mm^{-1} in the Sacramento and San Joaquin valleys, and the minimums ranged from 0.1 Mm^{-1} in west Texas to 8.0 Mm^{-1} in Los Angeles. Maximum monthly contributions to reconstructed particulate extinction ranged from 9% in the Virgin Islands to 56% in the central Great Plains, and the minimum contributions ranged from 2% in Appalachia to 17% in southern California.

3.3.2 Fine Particle Soil Extinction

Soil, like OMC and LAC, is assumed to have no humidity dependence, and therefore the seasonal patterns in extinction coefficients are the same as those for mass concentrations. In all IMPROVE regions, the maximum and minimum percent contributions of soil were greater in reconstructed fine mass than in reconstructed particulate extinction. While there were several regions where the timing of the minimum or maximum contribution differed by more than 2 months when comparing seasonal patterns in reconstructed particulate extinction to those in reconstructed mass, none had a noticeable impact in the overall seasonal pattern in soil contributions.

The rural maximum monthly soil extinction coefficients ranged from 0.3 Mm^{-1} in Alaska to 4.5 Mm^{-1} in the Virgin Islands, and monthly minimums ranged from 0.1 Mm^{-1} in Alaska to 1.2 Mm^{-1} in southern Arizona. Urban maximums ranged from 0.5 Mm^{-1} in upper Michigan to 3.9 Mm^{-1} in Phoenix; minimums ranged from 0.1 Mm^{-1} in upper Michigan to 2.4 Mm^{-1} in Phoenix. Maximum monthly contributions to reconstructed particulate extinction ranged from 1% on the East Coast to 14% in the Virgin Islands, and the minimum contributions ranged from 0.3% in the Columbia River Gorge to 5% in southern Arizona.

3.3.3 Coarse Particle Mass Extinction

Since no humidity dependence for CM is assumed in the extinction model, the seasonal patterns in extinction coefficients and mass concentrations are comparable. While the seasonal patterns in CM concentrations are not discussed in this report since the prior discussion was focused on the reconstructed fine mass model, seasonal coarse mass values can be found in Appendix B. In general, the seasonal patterns in CM are very similar to those in soil, with the maximum and minimum extinction coefficients in CM and soil occurring within a month of each other in over half and over three-quarters of the regions, respectively. Thus, peak CM extinction coefficients are typically in spring or summer, and minimum values are typically in fall or winter.

However, several regions that had a spring maximum in soil extinction coefficients had a summer maximum in CM; these included Boundary Waters, Hells Canyon, the Northeast, the northern Great Plains, and southern California. One region, the East Coast, had the reverse situation of a springtime maximum in CM and a summertime maximum in soil. Additionally, several regions that had fall time minima in soil extinction had winter minima in CM; these were Hawaii, the East Coast, and the mid-South. The central Rockies had the reverse situation of a winter minimum in soil and a fall maximum in CM. Exceptions to the spring-summer maximum in CM extinction coefficients included several regions that had spring or summertime maximums in soil but had fall maxima in CM; these were Alaska, the California coast, the Sierra Nevada, Oregon and northern California, the Ohio River valley, Phoenix, and Washington, D.C. Washington, D.C., was also an exception to the fall-winter minimum in CM extinction coefficients with its minimum monthly extinction value occurring in June.

The ratios of maximum to minimum CM extinction coefficients were lower than the ratios in soil extinction in most regions, particularly in regions with a high degree of seasonality in soil concentrations. The maximum to minimum ratio in CM extinction coefficients ranged from 1.5 to 12, with a median value of 3, whereas soil ratios ranged from 1.5 to 20, with a median value of 5. Different seasonal patterns in CM and soil might indicate different sources for the two aerosol types.

The maximum monthly CM extinction coefficients ranged from 1.7 Mm^{-1} in Alaska to 15.3 Mm^{-1} in Phoenix, and monthly minimums ranged from 0.3 Mm^{-1} in Hells Canyon to 10.5 Mm^{-1} in Phoenix. Maximum monthly contributions to reconstructed particulate extinction ranged from 4% in Appalachia to 45% in the Virgin Islands, and the minimum contributions ranged from 1% in Death Valley to 14% in Hells Canyon.

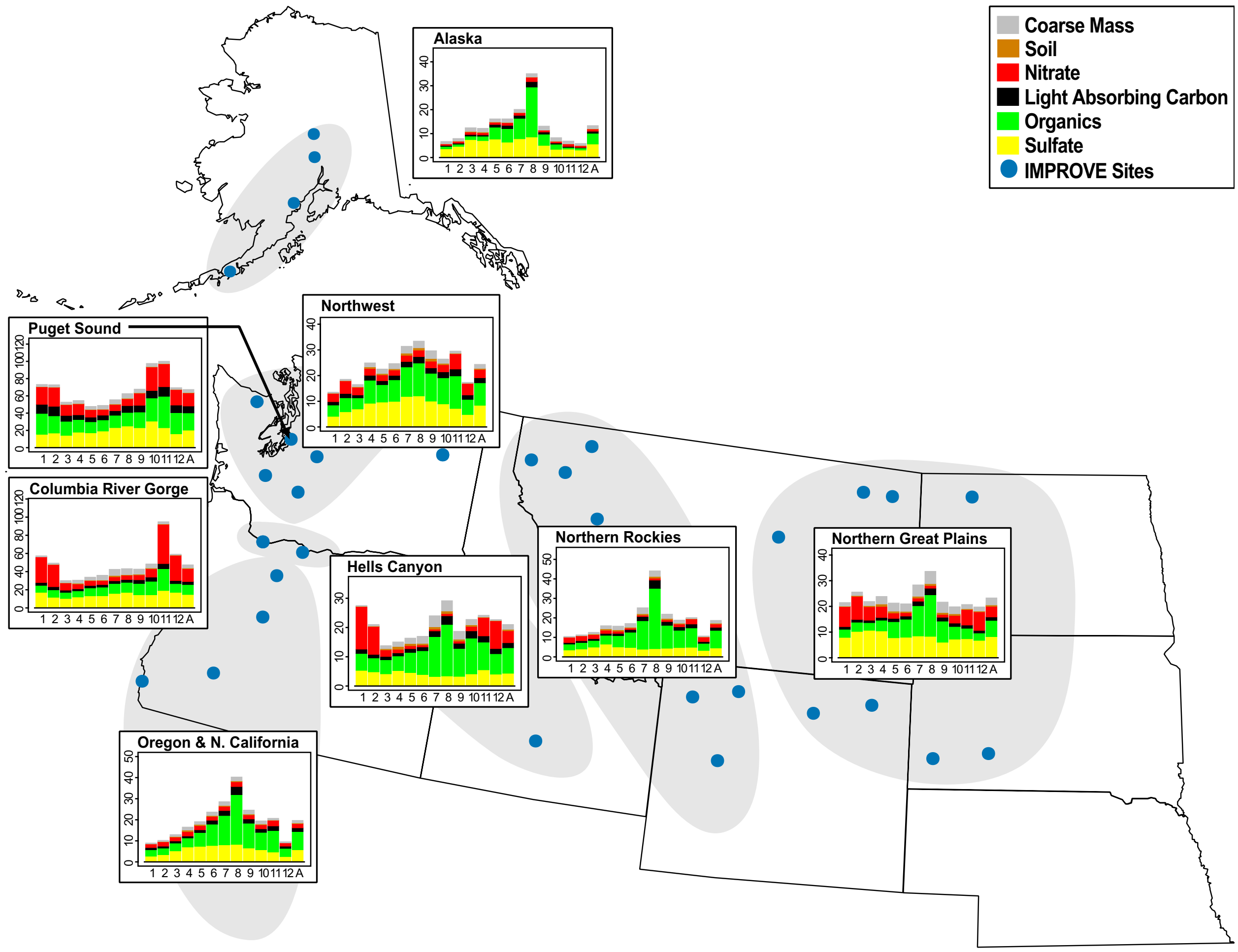

Figure 3.13. Map showing stacked bar charts of monthly distributions of particulate extinction coefficients for the northwestern U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

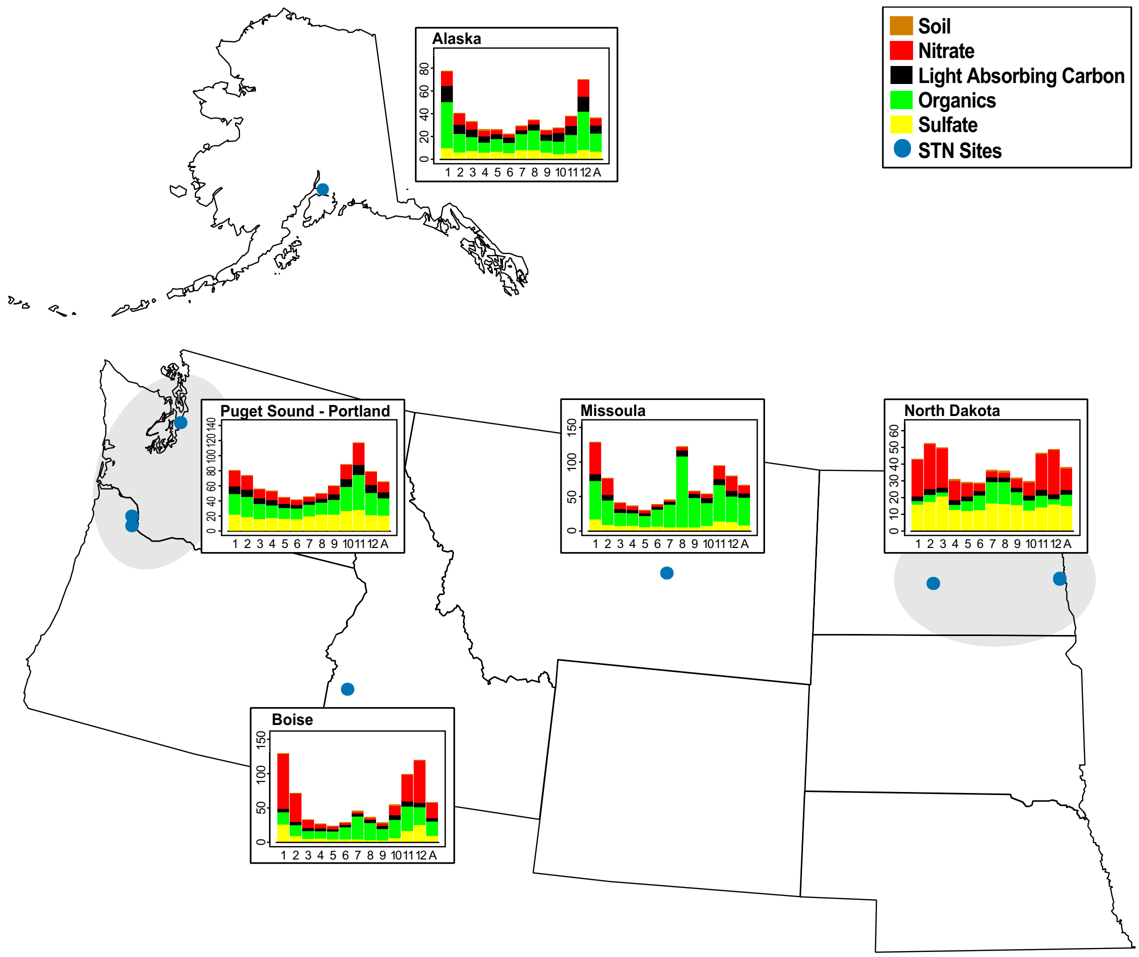

Figure 3.14. Map showing stacked bar charts of monthly distributions of fine particulate extinction coefficients (Mm^{-1}) for the northwestern U.S. regions of the STN network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, and soil are the order of presentation. Coarse mass measurements were not available for STN and so are not included.

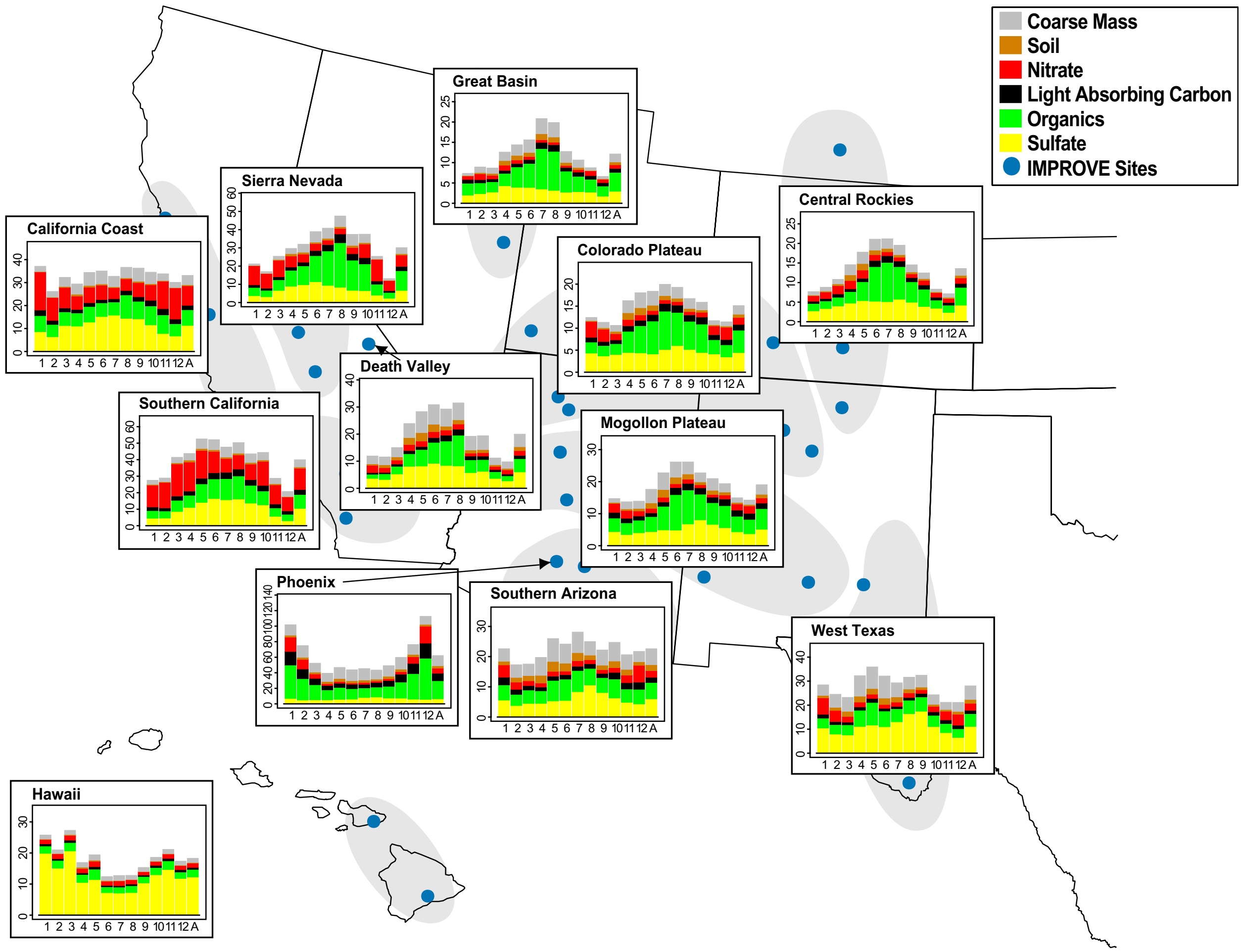

Figure 3.15. Map showing stacked bar charts of monthly distributions of particulate extinction coefficients (Mm^{-1}) for the southwestern U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

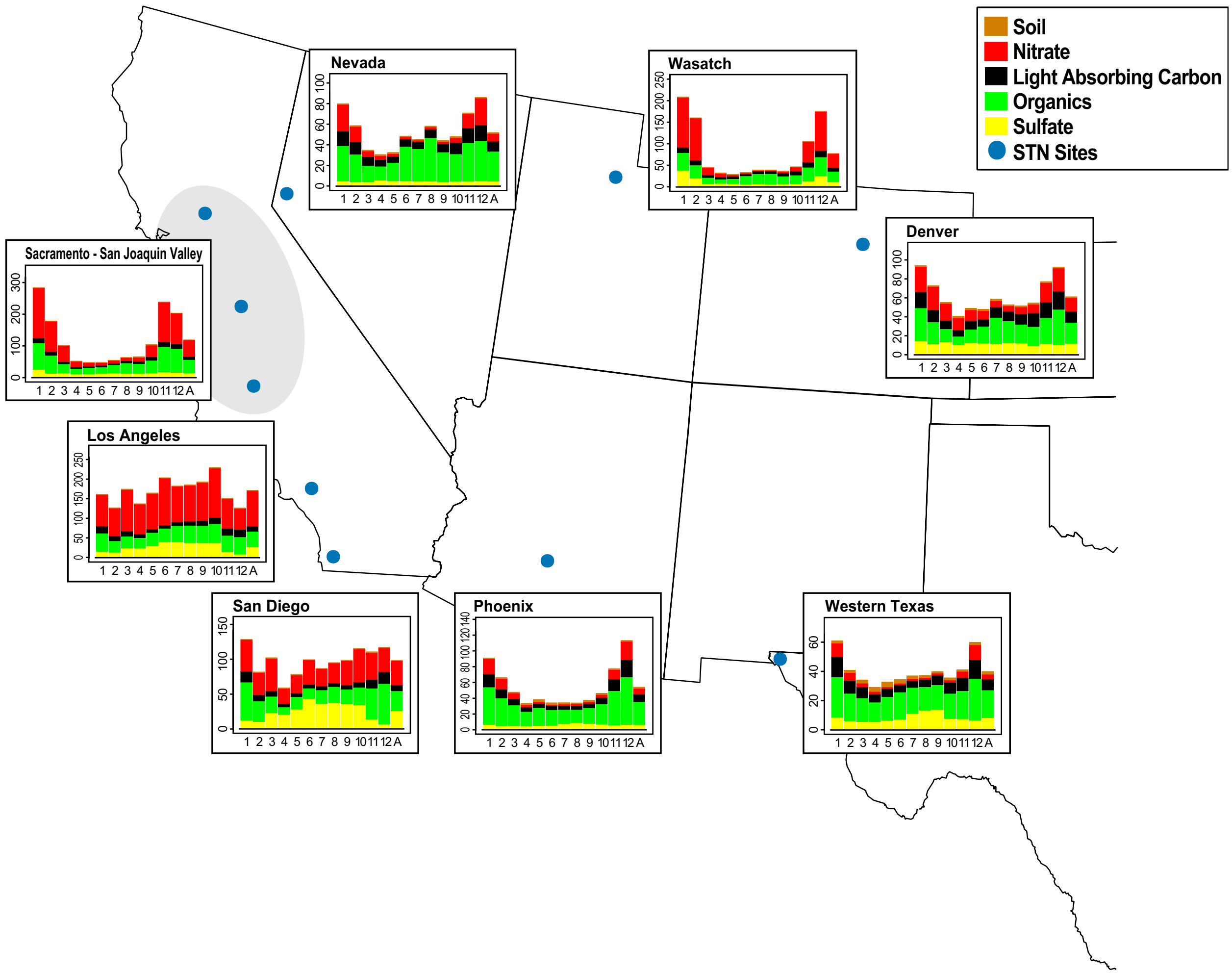

Figure 3.16. Map showing stacked bar charts of monthly distributions of fine particulate extinction coefficients (Mm^{-1}) for the southwestern U.S. regions of the STN network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, and soil are the order of presentation. Coarse mass measurements were not available for STN and so are not included.

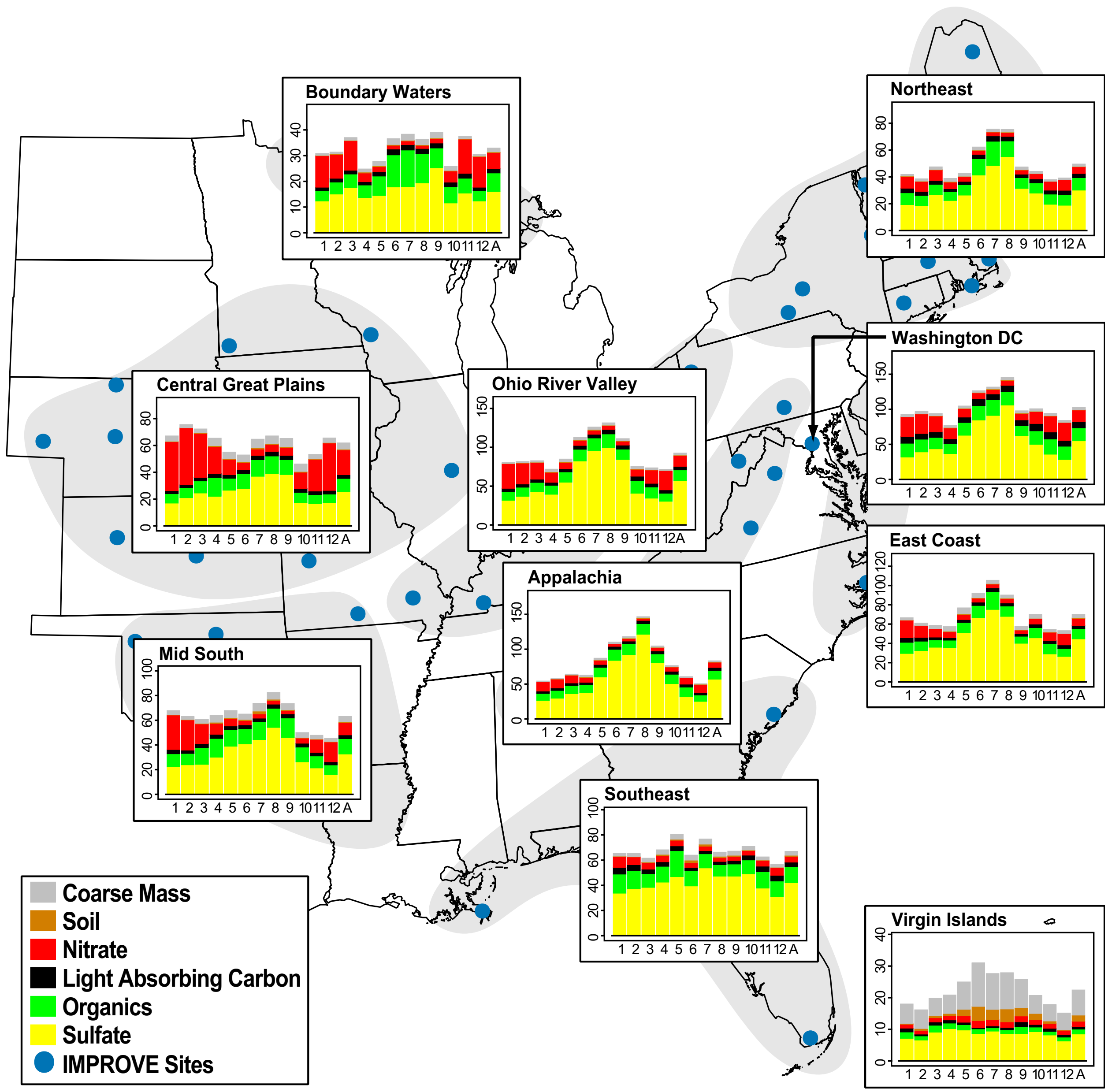

Figure 3.17. Map showing stacked bar charts of monthly distributions of particulate extinction coefficients (Mm^{-1}) for the eastern U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

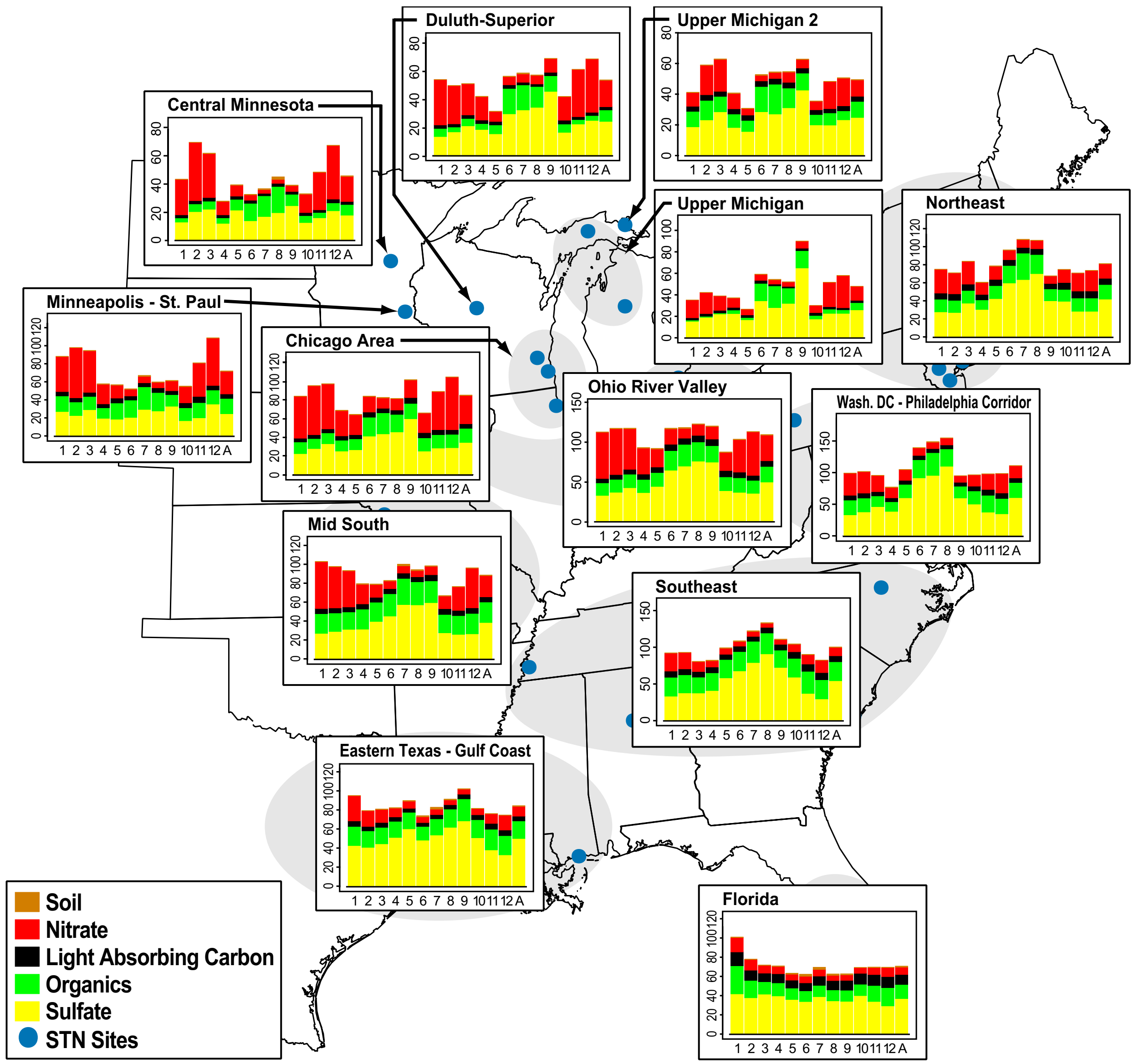

Figure 3.18. Map showing stacked bar charts of monthly distributions of fine particulate extinction coefficients (Mm^{-1}) for the eastern U.S. regions of the STN network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, and soil are the order of presentation. Coarse mass measurements were not available for STN and so are not included.

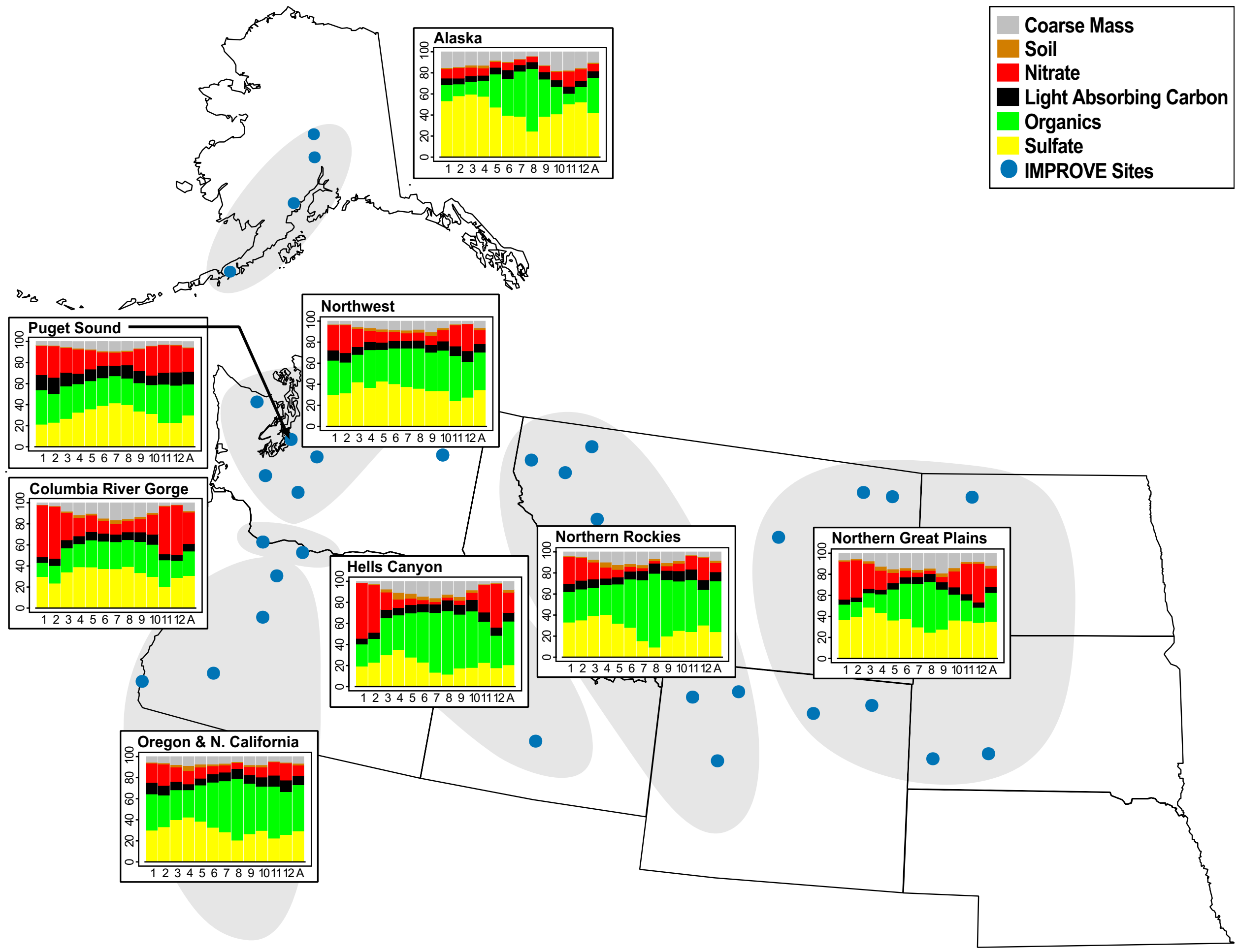

Figure 3.19. Map showing stacked bar charts of monthly percent contribution to reconstructed particulate extinction (%) for particulate extinction coefficients for the northwest U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

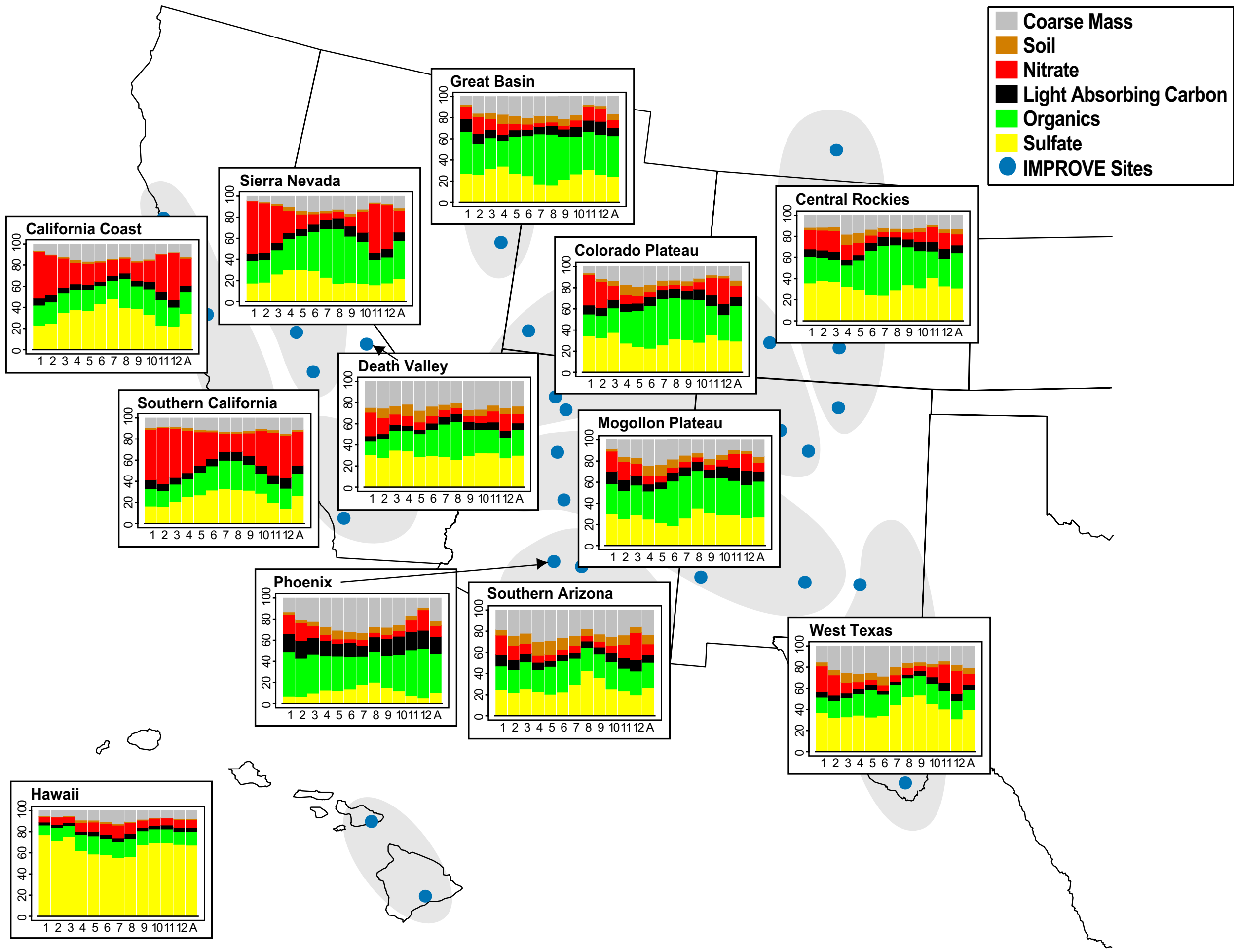

Figure 3.20. Map showing stacked bar charts of monthly percent contribution to reconstructed particulate extinction (%) for particulate extinction coefficients for the southwest U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

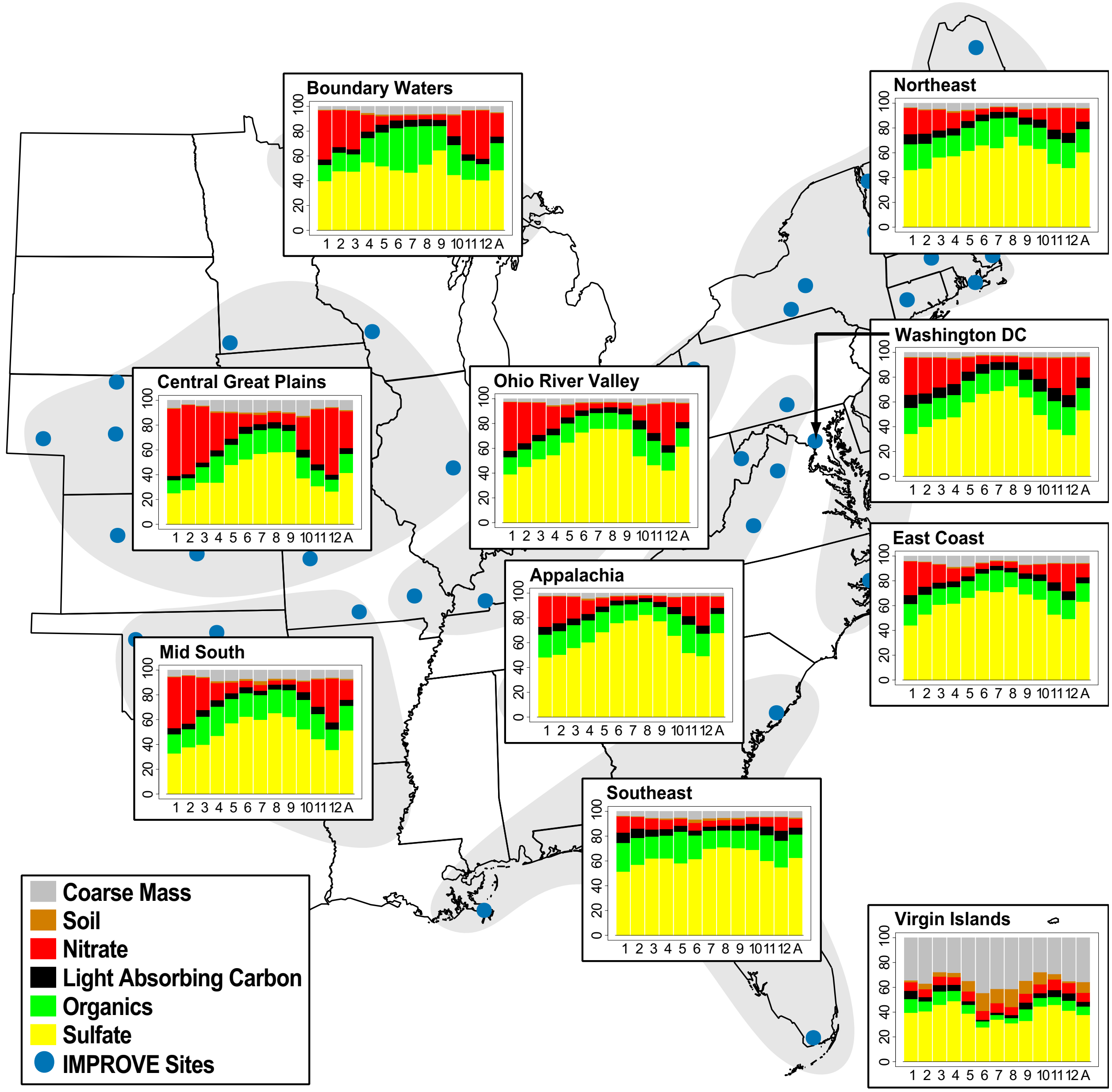

Figure 3.21. Map showing stacked bar charts of monthly percent contribution to reconstructed particulate extinction (%) for particulate extinction coefficients for the eastern U.S. regions of the IMPROVE network. Starting from the base of the chart, ammonium sulfate, organics, light-absorbing carbon, ammonium nitrate, soil, and coarse mass are the order of presentation.

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