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Key Points:
- Fine mineral dust and coarse aerosol mass varied significantly both seasonally and spatially at remote sites across the United States
- Elemental composition of fine mineral dust suggests influence from long-range transport from North Africa and Asia to the United States
- Increases in fine mineral dust concentrations have occurred in the Southwest in spring and Midwest in summer and fall

Supporting Information:
- Supporting Information S1

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Spatial and seasonal variability in fine mineral dust and coarse aerosol mass at remote sites across the United States

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Abstract Understanding the spatial and temporal variability in fine mineral dust (FD, mineral aerosols with diameters less than 2.5 μm) and coarse aerosol mass (CM, mass of aerosols with diameters between 2.5 and 10 μm) is important for accurately characterizing and perhaps mitigating their environmental and climate impacts. The spatial and seasonal variability of ambient FD and CM was characterized at rural and remote sites across the United States for 2011–2014 using concentration and elemental chemistry data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) aerosol monitoring network. FD concentrations were highest (and had ≥50% contributions to $PM_{2.5}$ mass) in the southwestern United States in spring and across the central and southeastern United States in summer (20–30% of $PM_{2.5}$ mass). CM was highest across the Southwest and southern Great Plains in spring and central United States in spring, summer, and fall (≥70% contributions to PM_{10} mass). Similar FD and CM seasonal variability was observed near source regions in the Southwest, but a seasonal decoupling was observed in most other regions, suggesting the contribution of nonlocal sources of FD or perhaps non-dust-related CM. The seasonal and spatial variability in FD elemental ratios (calcium, iron, and aluminum) was fairly uniform across the West; however, in the eastern United States a shift in summer elemental composition indicated contributions from nonlocal source regions (e.g., North Africa). Finally, long-term trend analyses (2000–2014) indicated increased FD concentrations during spring at sites across the Southwest and during summer and fall in the southeastern and central United States.

1. Introduction

Dust is a major component of global aerosols [*Prospero et al.*, 2002; *Tanaka and Chiba*, 2006; *Shao et al.*, 2011] and has far-reaching environmental and climate impacts. It affects health [e.g., *Grineski et al.*, 2011; *Morman and Plumlee*, 2013; *Goudie*, 2014] and visibility degradation [e.g., *Kavouras et al.*, 2009; *Hand et al.*, 2011, 2014; *Ashley et al.*, 2015], ecosystem dynamics [*Field et al.*, 2010], hydrological cycles [e.g., *Painter et al.*, 2007; *Sorooshian et al.*, 2013], heterogeneous chemistry [e.g., *Krueger et al.*, 2004; *Sullivan et al.*, 2007], transport of bioaerosols [e.g., *Kellogg and Griffin*, 2006; *Hallar et al.*, 2011], direct and indirect climate effects [e.g., *Arimoto*, 2001; *Huang et al.*, 2014], and biogeochemical cycles [e.g., *Jickells et al.*, 2005]. The study of dust sources, emissions, transport, and above listed impacts has been the focus of many measurement and modeling studies globally in an effort to understand the spatial and temporal variability of dust and to reduce uncertainties in the estimations of its environmental, health, and climate impacts.

Dust in the United States derives from a combination of natural and anthropogenic sources. *Ginoux et al.* [2012] characterized natural and anthropogenic (e.g., agricultural) dust sources as part of a global study using satellite-based Moderate-Resolution Imaging Spectroradiometer (MODIS) Deep Blue estimates of dust optical depth along with land use data sets. Dust sources in the High Plains east of the Rocky Mountains from Montana to Texas include some of the largest anthropogenic sources of dust in the United States due to agricultural land use, while both natural and anthropogenic sources exist west of the Rocky Mountains. This region includes sources from the arid Southwest (e.g., the Mojave, Great Basin, Sonoran, and Chihuahuan Deserts) and gives rise to some of the highest aeolian dust concentrations in the United States [*Prospero et al.*, 2002]. The Southwest has been the focus of many local and regional studies to characterize dust concentrations that peak in spring and early summer due to synoptic-scale meteorological patterns that transport dust across the region and continent [e.g., *Brazel and Nickling*, 1986; *Stout and Lee*, 2003; *Novlan*

et al., 2007; *Park et al*., 2007; *Lee et al*., 2009; *Rivera Rivera et al*., 2009; *Hahnenberger and Nicoll*, 2012; *Tong et al*., 2012; *Lei and Wang*, 2014; *Reynolds et al*., 2016]. Fewer studies have reported dust concentrations in the central and northern Great Plains regions [e.g., *Hand et al*., 2011, 2012a]. Long-range intercontinental transport of dust also regularly impacts the United States, such as from Asia in spring in the Northwest [e.g., *Fischer et al*., 2009] and North Africa in summer in the Southeast [e.g., *Prospero et al*., 2010].

Many of the dust characterization studies that have occurred in the Southwest and other regions of the United States have used diverse types of data, such as column optical depth from satellite [e.g., *Ginoux et al*., 2012], low-visibility meteorological records [e.g., *Okin and Reheis*, 2002; *Novlan et al*., 2007], ground-based ion deposition [e.g., *Brahney et al*., 2013; *Sorooshian et al*., 2013], and other ground-based assessments using different particle size ranges and dust definitions [e.g., *Reheis and Urban*, 2011; *Tong et al*., 2012; *Neff et al*., 2013; *Lei and Wang*, 2014; *Reynolds et al*., 2016]. In addition, many studies focus only on details of episodic events [e.g., *Lee et al*., 2009]. Reconciling results from the variety of data sets, approaches, and dust definitions can be challenging.

Fine mineral dust (FD) is defined here to include mineral particles with aerodynamic diameters less than 2.5 μm. An algorithm to estimate FD used by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network assumes common oxide forms of typical mineral species found in soil in order to reconstruct aerosol mass and visibility at remote sites across the United States [*Malm et al*., 1994; *Hand et al*., 2011]. Advantages of this type of characterization are that it is highly quantitative and spatially and temporally consistent. FD, by definition, corresponds to the size range of particles conducive to regional or long-range transport [*Lawrence and Neff*, 2009]. Many studies have focused on FD to investigate its role in regional visibility degradation [*Kavouras et al*., 2009; *Hand et al*., 2011, 2014], regional contribution to $PM_{2.5}$ mass [*Sorooshian et al*., 2011; *Hand et al*., 2012a], long-term trends [*Sorooshian et al*., 2011; *Hand et al*., 2016], and long-range transport of dust from intercontinental sources using FD elemental ratios specific to different source regions [e.g., *Perry et al*., 1997; *VanCuren and Cahill*, 2002; *VanCuren*, 2003; *VanCuren et al*., 2005; *Prospero et al*., 2010; *Creamean et al*., 2014].

Dust elemental composition can vary depending on individual source regions [e.g., *Reheis et al*., 2002; *White et al*., 2015; *Engelbrecht et al*., 2016] and processes that occur during emission and transport. Elemental composition can affect ecosystem health and plant nutrition [e.g., *Reynolds et al*., 2006], dust radiative forcing [e.g., *Sokolik and Toon*, 1996, 1999], ice nucleation efficiency [e.g., *Zimmermann et al*., 2008; *Sorooshian et al*., 2013], reactivity with gases in the atmosphere [e.g., *Sullivan et al*., 2007], and solubility, which can influence atmospheric processing and biogeochemical impacts [*Jickells et al*., 2005]. Some global dust modeling efforts incorporate mineralogy in order to more accurately predict the environmental and climate impacts of dust [e.g., *Journet et al*., 2014; *Perlwitz et al*., 2015a, 2015b]. A recent study by *Engelbrecht et al*. [2016] investigated the physicochemical and optical properties of dust as a function of mineralogy and derived relationships between single-scatter albedo and iron content and mineralogy for entrained dust. Several regional studies in the United States have used elemental composition to attribute FD to sources in Asia [*Kurtz et al*., 2001; *Jaffe et al*., 2003; *VanCuren*, 2003; *Debell et al*., 2004; *Vicars and Sickman*, 2011; *Creamean et al*., 2014], North Africa [*Perry et al*., 1997; *Prospero*, 1999a, 1999b; *Prospero et al*., 2001; *Holmes and Miller*, 2004], as well as specific dust emission regions such as White Sands, New Mexico [*White et al*., 2015], the Mojave Desert [*Muhs et al*., 2007], specific sources within the Mojave Desert [*Reheis and Kihl*, 1995], and specific landforms such as playas (dry lake beds) [*Reheis et al*., 2009; *Pratt et al*., 2010]. However, continental-scale spatial and seasonal variability of measured FD elemental composition across the United States has yet to be reported.

FD is presumed to be associated with the fine tail of coarse dust size distribution [*Malm et al*., 1994; *Hand and Malm*, 2007]. Coarse mineral dust is often assumed to compose coarse mass (calculated as the difference between PM_{10} and $PM_{2.5}$ gravimetric mass: $CM = PM_{10} - PM_{2.5}$, the mass of particles with diameters between 2.5 and 10 μm); however, previous studies have suggested that CM can contain substantial contributions (40–50%) of carbonaceous aerosols and inorganic salts such as calcium nitrate and sodium nitrate [e.g., *Lee et al*., 2008]. A yearlong CM speciation study at nine remote IMPROVE sites across the United States in 2004 [*Malm et al*., 2007] found that on average mineral aerosols were the largest contributor to CM, ranging from 76% in the Southwest to 34% in the Northwest. Carbonaceous aerosols were the second largest contributor, ranging from 40% in the Southeast to 59% in the Northwest. Nitrate was the third largest contributor. CM composition can therefore vary significantly both regionally and seasonally. The assumption

that CM is a proxy for dust is stronger when supported by additional evidence [e.g., *Tong et al*., 2012; *Lei and Wang*, 2014; *Huang et al*., 2015; *Lei et al*., 2016]. One such line of evidence is the comparison of FD and CM seasonal and spatial patterns that may inform as to similar aerosol type, source regions, or transport patterns.

We examine the spatial and seasonal variability in a consistent dust metric, FD, using ambient aerosol data from remote sites in the IMPROVE network across the United States from 2011 to 2014. We also examine CM to identify similarities in regional and seasonal variability in both FD and CM to address assumptions of CM-related dust. Seasonal and spatial contributions of FD and CM to $PM_{2.5}$ and PM_{10} mass, respectively, are also reported to demonstrate the importance of FD to $PM_{2.5}$ mass and CM to PM_{10} mass. We also report the degree of seasonal and spatial variability in FD elemental composition and identify regions potentially influenced by long-range transport of dust. Finally, long-term (2000–2014) seasonal trends in FD and its contribution to $PM_{2.5}$ mass across the United States were computed to identify regions and seasons corresponding to increased FD concentrations. This analysis of overall spatial and temporal variability of FD and CM, their contributions to $PM_{2.5}$ and PM_{10} mass, and FD elemental composition is necessary in order to understand and reduce uncertainties in estimating environmental and climate impacts of mineral aerosols.

2. Data and Methods

The IMPROVE network was initiated in 1988 [*Malm et al*., 1994] and currently operates approximately 160 remote and rural sites, such as national parks and wilderness areas, across the United States. The network collects 24 h samples every third day from midnight to midnight, and ambient concentrations are reported as mass per air volume at local temperature and pressure. Four independent modules are used to collect filters for gravimetric mass measurements ($PM_{2.5}$ and PM_{10}) and $PM_{2.5}$ speciated chemical analyses, including anions, carbonaceous aerosols, and elemental concentrations determined from X-ray fluorescence (XRF). Since 2011 a Panalytic XRF system has been used to determine elemental concentrations, and data are used as reported (no substitutions for values below detection limits). PM_{10} and $PM_{2.5}$ mass concentrations are determined through gravimetric weighing, and CM is calculated from the difference. Speciated analyses are not performed on the PM_{10} filters. Additional details regarding IMPROVE site location, sampling, analysis methodology, and detailed descriptions of network operations and data analysis have been previously reported [*Hand et al*., 2011, 2012a]. IMPROVE data advisories were followed for the data reduction and analysis in this study (see http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory.htm), and IMPROVE data are available for download (http://views.cira.colostate.edu/fed/).

FD concentrations were estimated by assuming oxide norms associated with predominant soil species (e.g., Al_2O_3, SiO_2, CaO, K_2O, FeO, Fe_2O_3, and TiO_2), with a correction factor for other compounds such as MgO, Na_2O, water, and carbonate (equation (1)). Potassium is not specifically included because of its nonsoil component from biomass smoke; it is accounted for by using Fe as a surrogate [*Malm et al*., 1994]. Equation (1) does not differentiate between types of mineral dust (agricultural, fugitive, natural, etc.).

$$\mathrm{FD} = 2.20[\mathrm{Al}] + 2.49[\mathrm{Si}] + 1.63[\mathrm{Ca}] + 2.42[\mathrm{Fe}] + 1.94[\mathrm{Ti}] \quad (1)$$

Aluminum [Al], silicon [Si], calcium [Ca], iron [Fe], and titanium [Ti] mass concentrations ($\mu g\,m^{-3}$) are determined through XRF analyses, as stated above. The factors used in equation (1) were confirmed through comparisons of resuspended soils and ambient aerosols in the western United States [*Cahill et al*., 1981; *Pitchford et al*., 1981]. Multilinear regression analysis by *Malm and Hand* [2007] suggested that FD may be underestimated by ~20% on average at IMPROVE sites across the United States, indicating that equation (1) misses some contribution from mineral dust components not included. FD fractional contributions to $PM_{2.5}$ were computed as the ratio of FD to reconstructed fine mass (accounting for other major $PM_{2.5}$ aerosol species such as sulfates, nitrates, carbonaceous aerosols, and sea salt [e.g., *Hand et al*., 2012a]), and CM fractions were calculated as the ratio of CM to gravimetric PM_{10} mass.

Monthly mean FD and CM concentrations were computed by requiring 50% completeness criteria for daily data. Seasonal means were computed from the monthly means for winter (December, January, and February), spring (March, April, and May), summer (June, July, and August), and fall (September, October, and November) and required two thirds of the data from each 3 month season to be valid, and 8 valid months of data were required for an annual mean. Data were aggregated from 2011 through 2014 to include only XRF data from the Panalytic system, resulting in 160 "complete" IMPROVE sites. Data were interpolated

using an ordinary kriging algorithm [*Isaaks and Mohan Srivastava*, 1989]. Regional concentrations were computed by aggregating individual site data into 28 rural geographic regions based empirically on site location and magnitudes and seasonal distribution [*Hand et al*., 2012a]. Three regions included only one site (Columbia River Gorge, Death Valley, and Virgin Islands). Elevation was not considered in regional definitions.

Based on reanalysis of archived filters from 1995 to 2010 at three sites, *Hyslop et al*. [2015] discovered spurious temporal trends in some of the species used to estimate FD, such as Al, Si, and Ti, due to changes in analytical methods. Based on this analysis, seasonal-specific trends were computed using Fe due to its stability over time and it being a key constituent of dust. Before 2011 Fe concentrations below minimum detection limits (MDL) were replaced by 0.5 × MDL; after and including 2011, no substitutions were made. Trends were computed for the years 2000–2014 using a linear Theil regression [*Theil*, 1950] following the methods used by *Hand et al*. [2012b, 2014]. Selection of the beginning time period was based on the network expansion in 2000. Trends were calculated from seasonal means and required valid data in 10 of the 15 years. Normalized trends (% yr^{-1}) were computed by dividing the slope from the regression by the median of the relevant seasonal mean concentrations. Significance levels (p) were computed using Kendall tau statistics. Trend isopleths were generated similarly to mass concentration isopleths. Fe emissions in the western United States are dominated by mineral dust sources, but combustion sources could contribute in the East and Northwest [e.g., *R. Wang et al*., 2015]. Spatial and seasonal trends in Fe were similar to those in Ca as well as FD, even with the issues reported by *Hyslop et al*. [2015]. Trends in fraction contribution of FD to reconstructed $PM_{2.5}$ mass similarly were computed. CM trends are not reported since it is the difference of two measurements and potentially affected by changes to the PM_{10} inlet.

3. Results

3.1. FD Seasonal and Spatial Patterns

Isopleths of 2011–2014 seasonal mean FD concentrations ($\mu g\,m^{-3}$) are shown for winter, spring, summer, and fall in Figures 1a–1d, respectively, using a similar scale. The highest FD concentrations occurred in spring in the southwestern United States, especially in Arizona, New Mexico, and southwestern Texas (~2.5 $\mu g\,m^{-3}$) and were lower in surrounding states (Figure 1b). The lobe of lower dust concentrations reaching southward toward the border between Arizona and New Mexico follows the Mogollon Rim and likely reflects elevation differences and changes in land surface characteristics in the region. Elevated spring dust levels across the Southwest are common due to abundant local or regional sources and synoptic-scale weather patterns [e.g., *Brazel and Nickling*, 1986; *Lee et al*., 1994, 2009, 2012; *Novlan et al*., 2007; *Kavouras et al*., 2009; *Rivera Rivera et al*., 2009, 2010; *Baddock et al*., 2011, 2016; *Hahnenberger and Nicoll*, 2012; *Hand et al*., 2016; *Reynolds et al*., 2016].

The second highest fine dust season in the United States occurred in summer (Figure 1c), with higher concentrations in the Southwest (the peak dust season in the Sonoran Desert), associated with the North American monsoon, which initiates frequent but largely mesoscale dust events through strong winds associated with thunderstorms [*Lei et al*., 2016]. The elevated dust pattern extends from the southern Great Plains into the central Great Plains and mid-South regions (1.5–2.0 $\mu g\,m^{-3}$). Southern states along the Gulf of Mexico, southern Florida, and the Virgin Islands also experienced elevated concentrations in summer (1–2.5 $\mu g\,m^{-3}$), likely associated with transport from North Africa [e.g., *Perry et al*., 1997]. The lowest seasonal mean concentrations ($<0.5\ \mu g\,m^{-3}$) corresponded to winter and fall (Figures 1a and 1d, respectively), although slightly elevated levels occurred in the Virgin Islands, central and northern Great Plains, and at a handful of sites in southern Arizona and western Texas. Winter dust storms are not uncommon in the Southwest [*Lee et al*., 1994, 2009; *Rivera Rivera et al*., 2010].

Seasonal mean contributions of FD to $PM_{2.5}$ mass are shown in Figures 2a–2d, for winter, spring, summer, and fall, respectively. FD is a major component of $PM_{2.5}$ mass (≥50%) during springtime at most sites across the Southwest and contributed 30% at western sites reaching northward nearly to Canada (Figure 2b). Winter and fall contributions in the Southwest were lower (Figure 1a) but reached 35–40% at several sites in Nevada and California and along the U.S.-Mexico border. Contributions of FD to $PM_{2.5}$ mass in the mid-South and central United States were around 25–30% during summer and closer to 20% in the fall, stretching northward into the northern Great Plains. Contributions of FD to $PM_{2.5}$ in the East were much lower than in

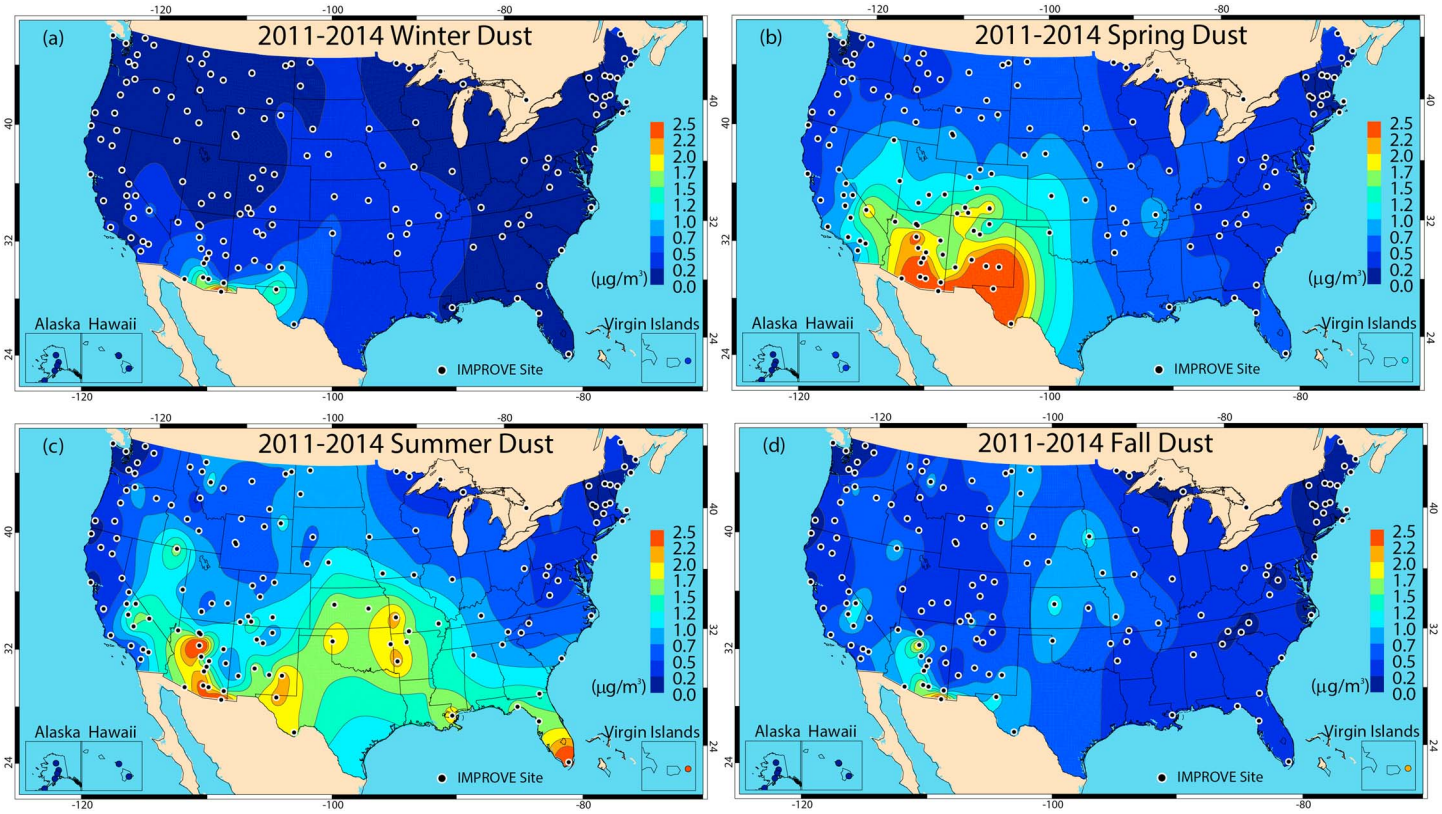

Figure 1. IMPROVE 2011–2014 seasonal mean fine ($PM_{2.5}$) dust concentrations ($\mu g\,m^{-3}$) for (a) winter, (b) spring, (c) summer, and (d) fall.

the West (<10% year round), with the exception of summer in southern Florida (~40%) and the Virgin Islands (~50%), regions influenced by the transport of North African dust [*Perry et al*., 1997; *Prospero*, 1999a, 1999b].

3.2. CM Seasonal and Spatial Patterns

CM seasonal mean concentrations are shown in Figures 3a–3d for winter, spring, summer, and fall, respectively. Spatial and seasonal patterns in CM tend to be similar to those of FD, although with some important differences. CM concentrations were highest (~15 $\mu g\,m^{-3}$) during spring in the Southwest, but the spatial patterns extend northeastward from the U.S.-Mexico border into the southern Great Plains and central United States (Figure 3b) rather than northwestward into Colorado, Nevada, and beyond as observed for FD. This pattern implies northeastward advection and is consistent with dust transport from the Chihuahuan Desert and southernmost Great Plains [e.g., *Novlan et al*., 2007; *Park et al*., 2007*; Rivera Rivera et al*., 2009]. The differences in the spatial patterns of FD and CM may be due to different sources of CM [e.g., *Malm et al*., 2007] or different CM transport pathways as a result of shorter CM atmospheric lifetimes due to size, elevation-limited transport, or deposition to different land surface characteristics (more vegetated). In fact, site elevation and CM concentrations were strongly ($p < 0.01$) inversely correlated for 59 sites in Arizona, New Mexico, Colorado, Utah, Texas, Nevada, and California for all seasons (−0.42, −0.39, −0.60, and −0.60 for winter, spring, summer, and fall, respectively). Linear regression analysis suggested a seasonal mean decrease of $2.4 \pm 1.0\,\mu g\,m^{-3}$ CM with 1 km elevation gain. Summer concentrations were high (10–15 $\mu g\,m^{-3}$) in southern California, southern Arizona, and eastern New Mexico, perhaps related to agricultural activity and convective storms related to the North American monsoon [*Adams and Comrie*, 1997; *Gill et al*., 2009; *Lee et al*., 2012]. Higher concentrations also stretched from the southern into the central and northern Great Plains (Figure 3c). Some East Coast sites also experienced higher CM (8–10 $\mu g\,m^{-3}$), including sites in southern Florida and the Virgin Islands. CM concentrations were elevated during fall across the Great Plains and into the central United States (~15 $\mu g\,m^{-3}$; Figure 3d), likely related to fugitive dust from agricultural activities. Winter CM concentrations were low throughout the United States (~1–3 $\mu g\,m^{-3}$), with the exception of sites in the southern Great Plains and

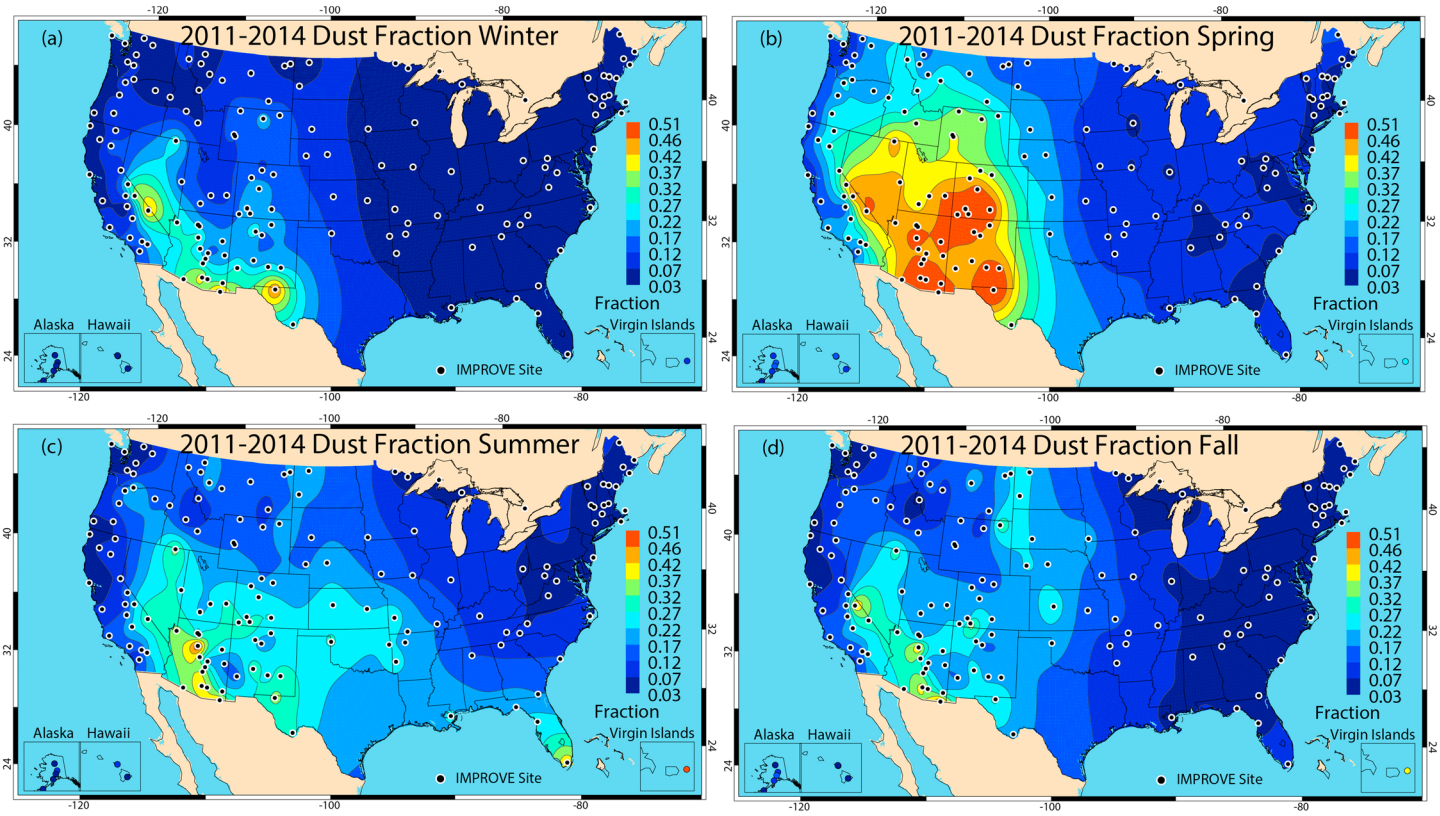

Figure 2. IMPROVE 2011–2014 seasonal mean fine dust fraction of reconstructed $PM_{2.5}$ mass (a) winter, (b) spring, (c) summer, and (d) fall.

central United States (Figure 3a) where winter is an important dust season and dust is transported primarily east and northeast.

Seasonal mean contributions of CM to PM_{10} mass are shown in Figures 4a–4d, for winter, spring, summer, and fall, respectively. CM was a major contributor to PM_{10} mass (≥70%) across the central and western United States, especially during spring in the Southwest (Figure 4b), year round in central and southern California and along the U.S.-Mexico border, and during the fall in the Great Plains, stretching from Texas to North Dakota (Figure 4d). Low contributions (<30%) extended across the East, especially in winter (Figure 4a) but were also low in the Northwest and Northeast year round.

3.3. FD and CM Regional Seasonality

Some of the differences in the spatial and seasonal patterns of FD and CM are worth exploring in more detail in order to examine possible influences of long-range transport on FD, regional variations in sources and composition of FD and CM, and/or perhaps non-dust-related CM. Regional, monthly mean FD and CM were each normalized to their respective annual means to investigate and compare their seasonal variability. Coupled seasonal variability suggests similar source regions and transport lifetimes, while decoupled seasonality suggests long-range or regional transport of FD or non-dust-related CM. Meteorological influences such as prevailing transport patterns and precipitation also likely affect these patterns. Regional, monthly mean normalized FD and CM in the southwestern, northwestern, and eastern United States are summarized in Figures 5–7, respectively. Regions deeper in the Southwest (e.g., southern Arizona, Mogollon Plateau, and West Texas) had similar seasonal patterns in FD and CM with strong spring maxima (Figure 5), associated with synoptic-scale winds that loft dust from local and regional sources coupled with seasonal variations in soil moisture, land cover, and land management practices. The strength of the CM spring maxima decreased at regions on the northern and western edge of the Southwest (i.e., Colorado Plateau, Central Rockies, Great Basin, and Sierra Nevada), consistent with the spatial gradients observed in Figures 1b and 3b. This decoupling intensified for most of the regions in the northwestern United States (Figure 6) with a spring

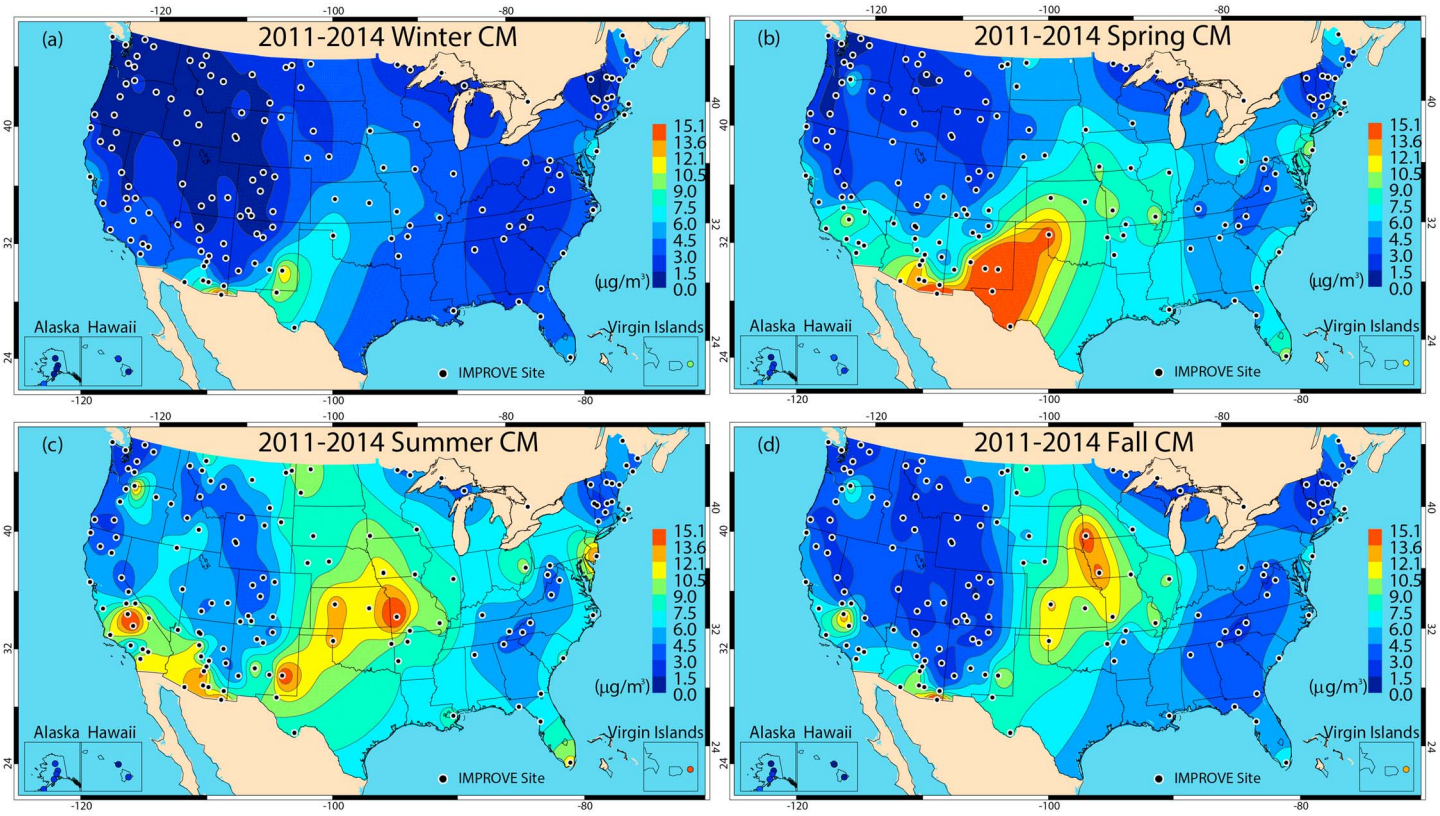

Figure 3. IMPROVE 2011–2014 seasonal mean coarse mass (CM = $PM_{10} - PM_{2.5}$) (µg m^{-3}) (a) winter, (b) spring, (c) summer, and (d) fall.

FD maximum and a shift to summer/fall CM maxima, perhaps associated with CM from local agricultural activity [e.g., *Singh et al.*, 2012] or carbonaceous aerosols [e.g., *Malm et al.*, 2007]. Unlike at the southwestern regions, spring maxima in FD at northwestern regions generally did not include March, suggesting different local source regions and seasonal emission or transport patterns, such as possible influence from Asian transport, which has been well documented [e.g., *VanCuren and Cahill*, 2002; *Jaffe et al.*, 2003; *Zhao et al.*, 2008; *Fischer et al.*, 2009; *Creamean et al.*, 2014]. The Hawaii and Alaska regions both had strong spring FD maxima but flat and decoupled CM seasonality, suggesting possible Asian influence that, unlike regions in the Northwest, also included March [*Kurtz et al.*, 2001; *Cahill*, 2003].

Interestingly, the spring Asian FD influence may have extended as far east as the Boundary Waters and Northeast regions, where a spring decoupling in FD and CM was also observed (Figure 7). Asian dust impacts in the northeast previously have been observed [*Debell et al.*, 2004; *Cottle et al.*, 2013], as well as dust transport from the Southwest [*Park et al.*, 2007]. All regions in the East experienced a seasonal decoupling of FD and CM, with the mid-South, Southeast, Appalachia, and Ohio River Valley regions having strong FD maxima in summer, likely associated with North African dust transport. The strongest seasonality in the United States occurred in the summer in the mid-South and Southeast regions, where summer FD was 3 times higher than the annual mean. Seasonality in CM was much flatter and varied per region, but CM was generally highest in spring and summer in the East. Similar seasonality in FD and CM was observed at the Virgin Islands.

3.4. FD Elemental Composition

Elemental ratios (e.g., Al/Ca and Fe/Ca) have been used to trace or apportion long-range sources of dust, such as transport from Asia or Africa to the United States, with the assumption that given source regions have identifying elemental ratios [e.g., *Perry et al.*, 1997; *VanCuren et al.*, 2005; *Lawrence and Neff*, 2009; *Creamean et al.*, 2014]. Due to the episodic and regional impact of long-range transport, previous studies typically focused on specific regions and seasons. We investigated continent-wide, seasonal patterns in elemental composition of FD. We focused specifically on Ca, Fe, and Al. Si and Ti contributed relatively large and

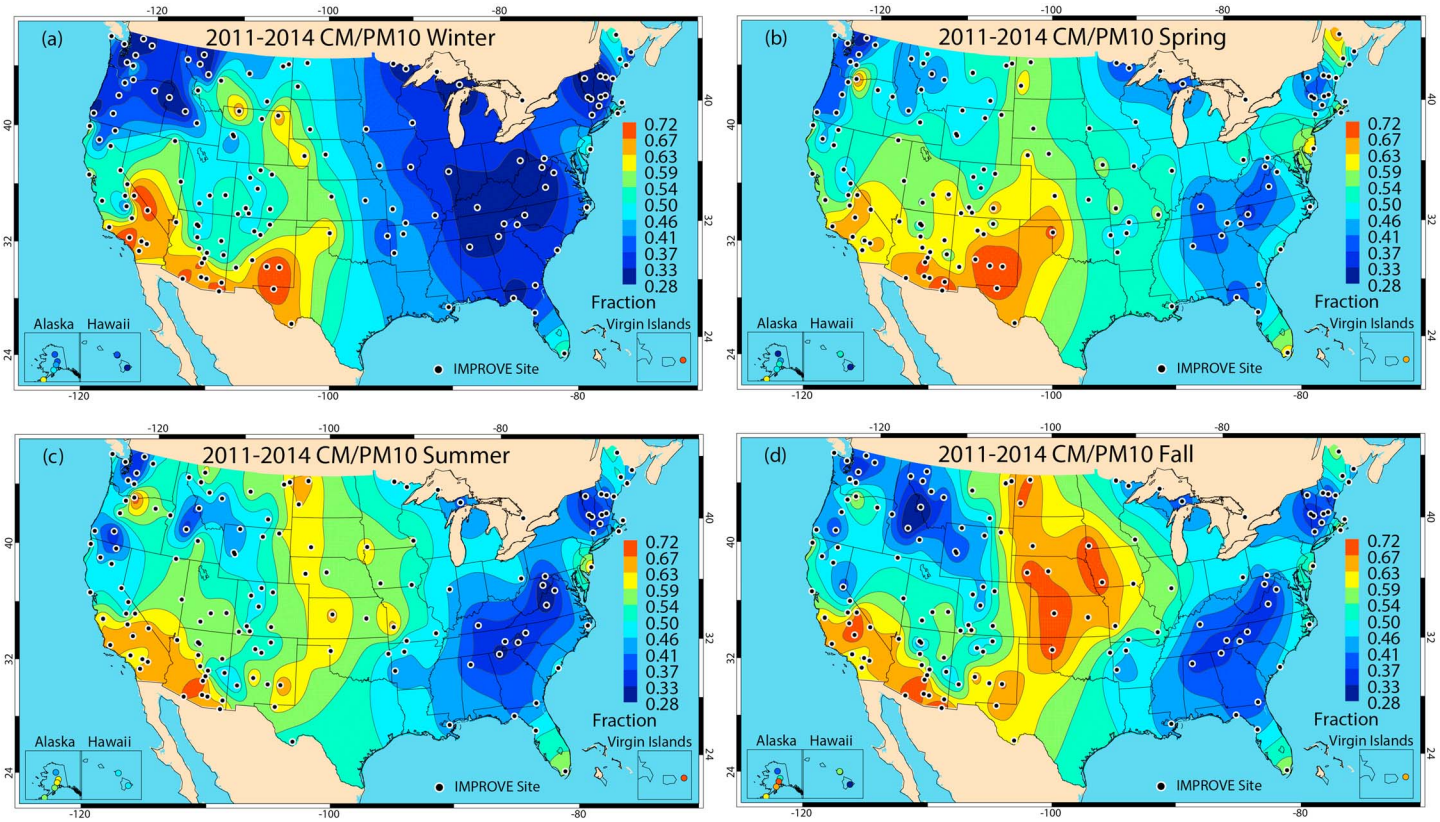

Figure 4. IMPROVE 2011–2014 seasonal mean CM fraction of measured PM_{10} mass (CM/ PM_{10}) (a) winter, (b) spring, (c) summer, and (d) fall.

small fractions, respectively, to FD and therefore were less informative on a fractional basis. Ca, Fe, and Al concentrations were monthly averaged, converted to mass fractions, regionally aggregated, and then plotted on ternary diagrams to visualize the data. Regional aggregation of sites was qualitatively based on similar spatial patterns in annual mean isopleths of mass fractions (see Figures S1a–S1c in the supporting information). Twelve regions were used, fewer than those shown in Figures 5–7 due to the more uniform spatial variability in elemental ratios. The range in seasonal variability for each region was calculated as the standard deviation in the annual mean mass fraction (for each species) and then quadratically summed. Seasonal mean ternary diagrams of Ca-Fe-Al for winter, spring, summer, and fall are shown in Figures 8a–8d, respectively. The circles are colored according to region (see inset map in Figure 8), and the size of the circle reflects the degree of seasonality, with the largest corresponding to the highest standard deviation in the annual mean.

The influence of North African transport on elemental composition in the East can be inferred from the shift in elemental composition in eastern regions (i.e., Appalachia and Southeast) especially in summer (Figure 8c), the season associated with transport of dust from North Africa. In summer the eastern regions shifted noticeably toward higher Al and lower Ca fractions and became aligned with the position of the Virgin Islands region on the ternary diagram. North African dust is reportedly relatively rich in Al compared to most North American dusts [*Gatz and Prospero*, 1996; *Bozlaker et al*., 2013; *Shelley et al*., 2015]. The summer, regional mean mass fractions for the Virgin Islands, Appalachia, and Southeast regions for Al, Fe, and Ca fractions were 0.51, 0.32, and 0.17, respectively. The West Texas region also shifted toward a higher Al fraction in summer; influence of North African dust on sites in West Texas previously has been reported [*Perry et al*., 1997; *Hand et al*., 2004]. However, the West Texas region also had a higher Ca fraction, consistent with calcium carbonate-enriched Chihuahuan Desert soils and dusts [e.g., *Monger et al*., 2009] and local sources of calcium sulfate that can also be entrained as dust [*White et al*., 2015]. During fall the position of the Virgin Islands region on the ternary diagram remained fixed but the Appalachia and Southeast regions shifted back toward lower Al fraction (Figure 8c), likely due to the seasonal transition in the transport pathways of North African

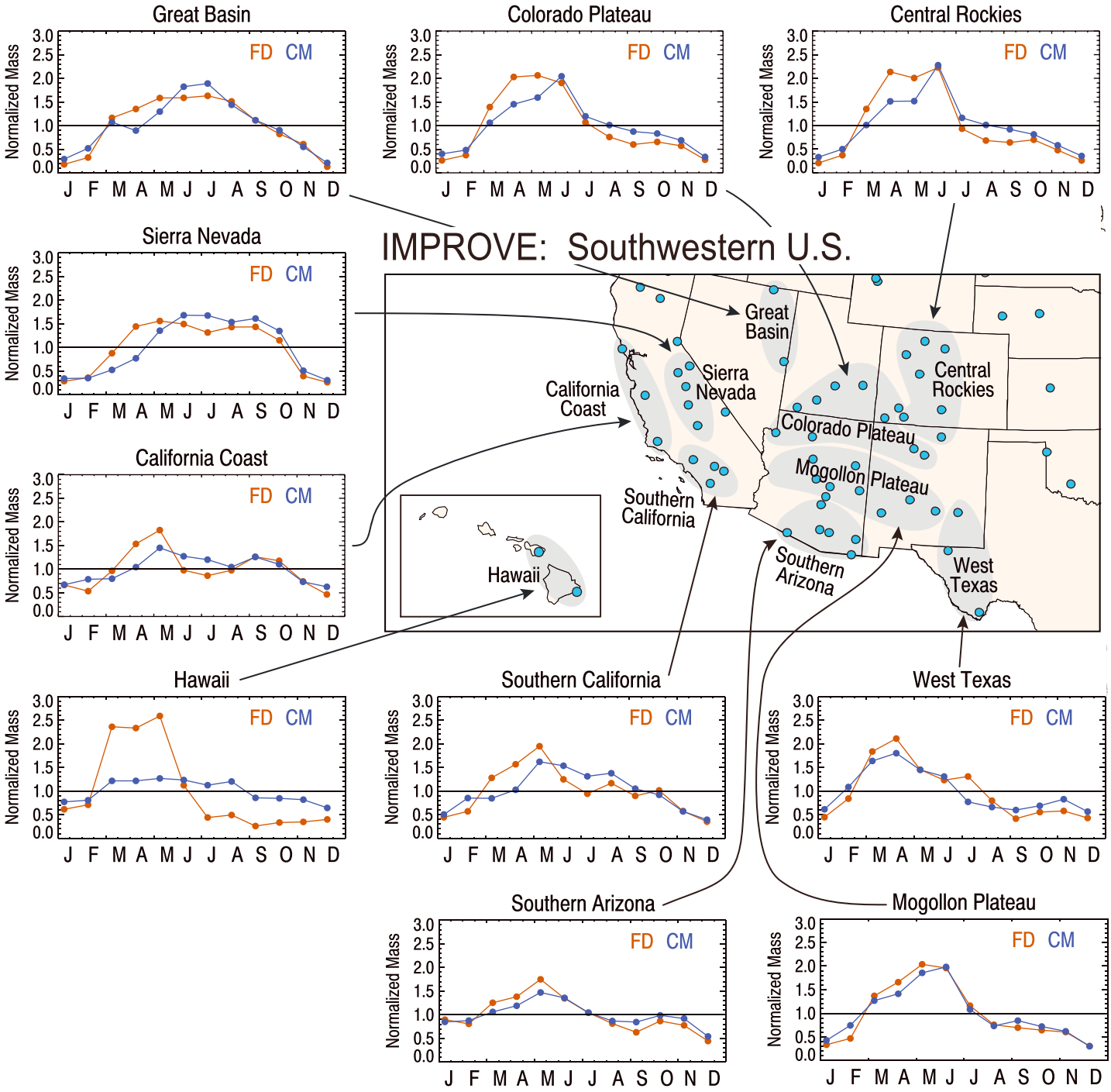

Figure 5. IMPROVE 2011–2014 regional, monthly mean normalized (by annual mean) fine dust (FD) and coarse mass (CM) for the southwestern United States. Sites are shown as blue dots and regional groupings in gray on the inset map.

dust [*Gläser et al.*, 2015]. The position of the Virgin Islands on the winter ternary diagram (Figure 8a) shifted toward higher Ca and lower Al and Fe fractions. Overall, the eastern regions corresponded to the highest seasonal variability in elemental ratios, consistent with the large degree of seasonal variability of FD observed in Figure 7.

The possible shift in elemental composition associated with the long-range transport of dust from Asia was more subtle relative to the shift corresponding to African dust influence. During spring, the season with the

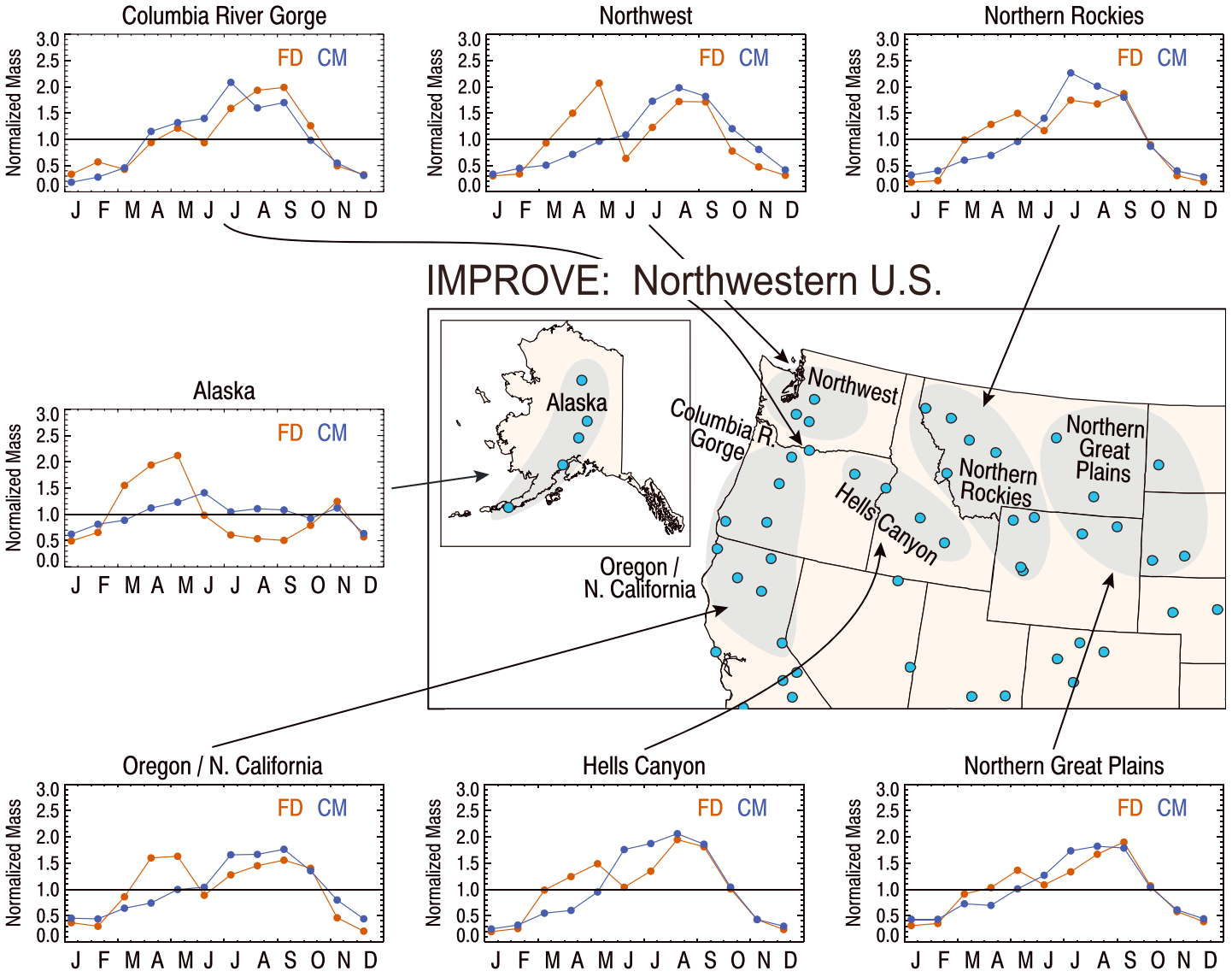

Figure 6. IMPROVE 2011–2014 regional, monthly mean normalized (by annual mean) fine dust (FD) and coarse mass (CM) for the northwestern United States. Sites are shown as blue dots and regional groupings in gray on the inset map.

reportedly highest Asian dust influence, Alaska, Hawaii, and the Northwest regions clustered near similar values (Figure 8b), especially relative to other seasons. The spring mass fraction average for these three regions for Al, Fe, and Ca were 0.42, 0.30, and 0.29, respectively. The Northeast and Appalachia regions were also clustered nearby, although with slightly lower Al fractions, suggesting some possible Asian influence. The West region was near these regions in spring but, along with the Northern Plains region, remained fairly fixed in this position for all seasons. The West and Northern Plains regions had the lowest degree of seasonal variability of all regions (see Figures 8a, 8c, and 8d).

The Midwest and the West Coast regions experienced relatively higher seasonality in elemental ratios compared to the West region. Ca fractions were highest during fall in the Midwest (Figure 8d); recall from Figures 1c and 3c that FD and CM concentrations, respectively, increased in this region during this season. The Ca fraction was highest for the West Coast region during summer (Figure 8c), perhaps due to marine influence. There was a noticeable seasonal shift in Fe fractions for some regions, which could influence dust optical properties and ecological impacts. The lowest Fe fractions ($<$0.25) occurred in winter (Figure 8a), and the highest fractions (0.25–0.50) occurred in summer and fall (Figures 8b and 8c), with the exception of low Fe

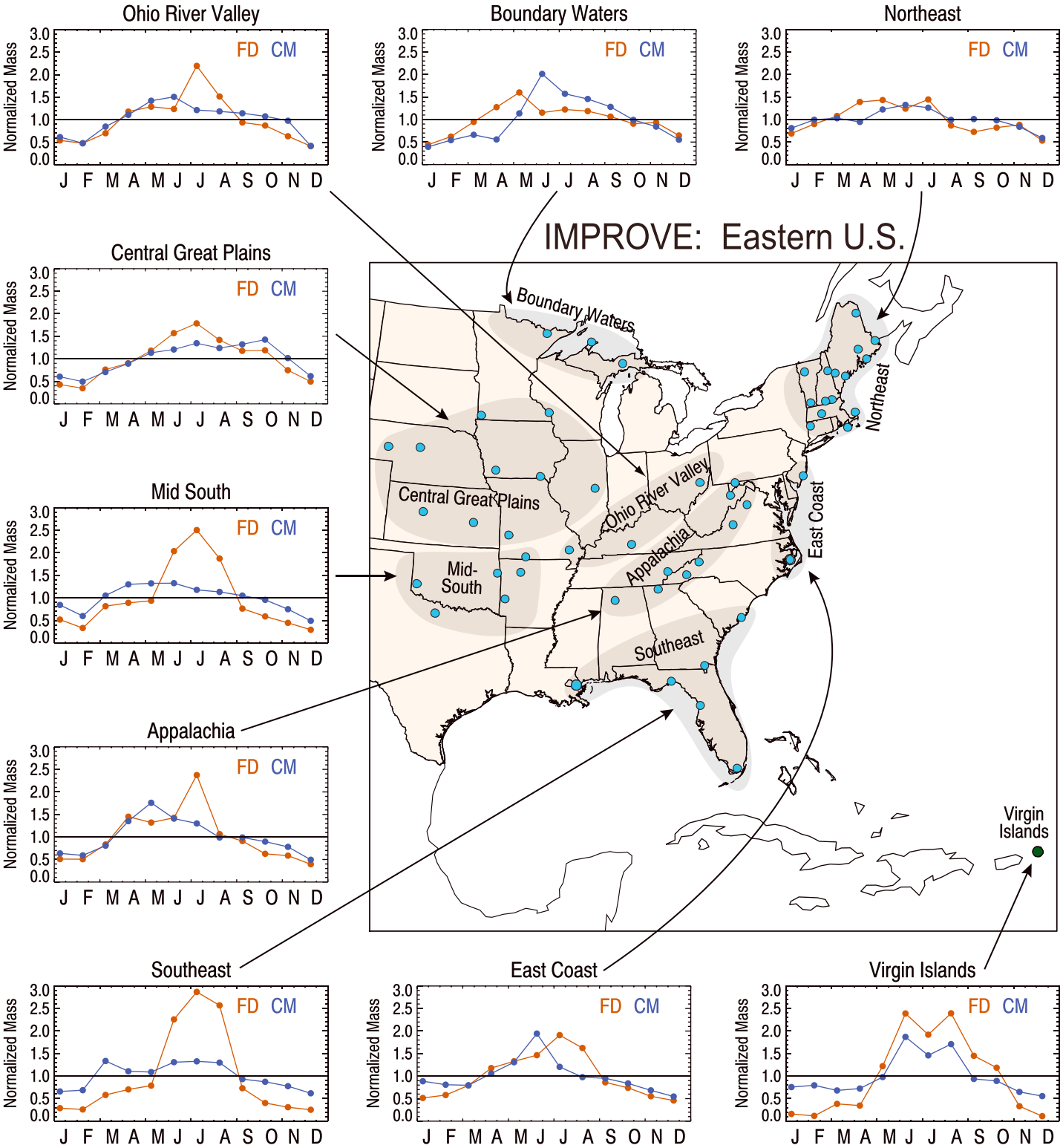

Figure 7. IMPROVE 2011–2014 regional, monthly mean normalized (by annual mean) fine dust (FD) and coarse mass (CM) for the eastern United States. Sites are shown as blue dots and regional groupings in gray on the inset map.

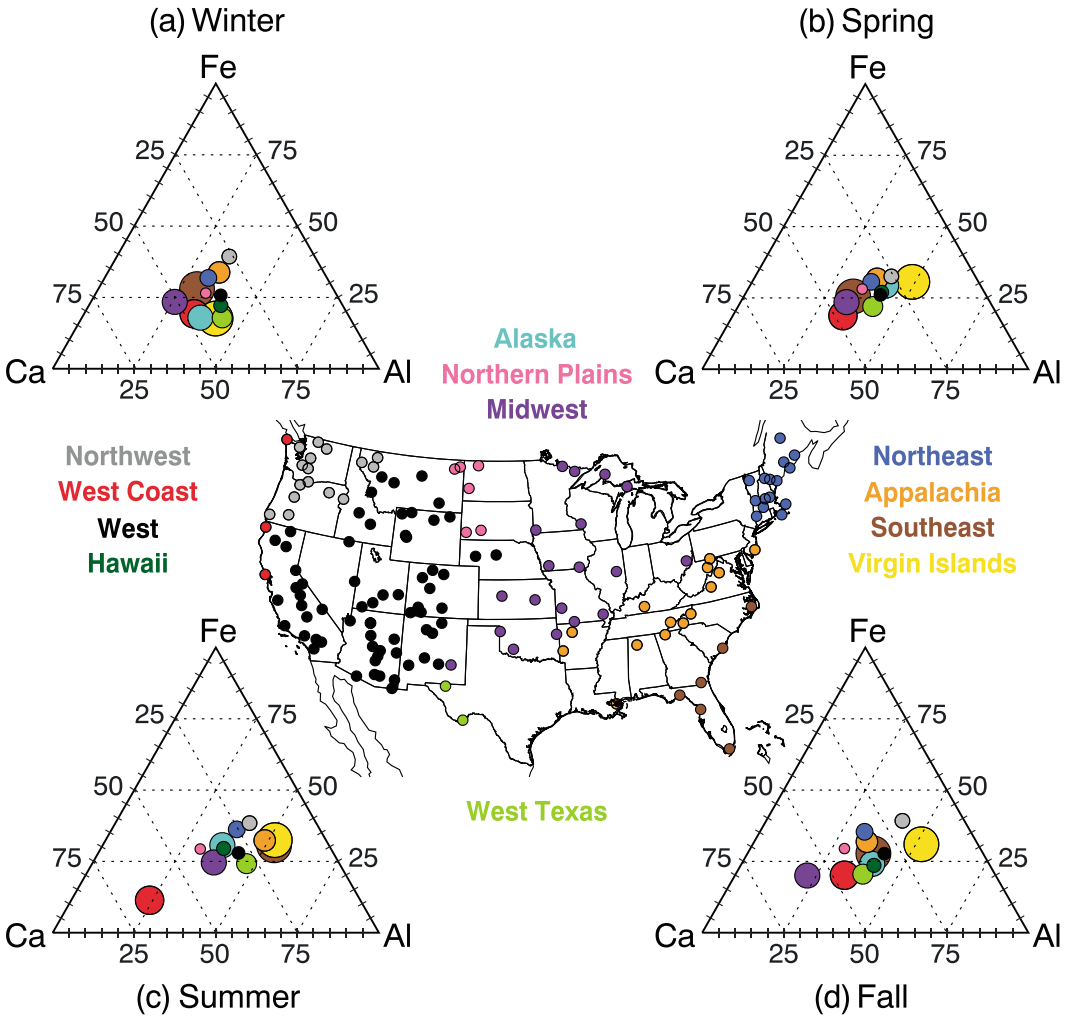

Figure 8. IMPROVE 2011–2014 regional, seasonal mean $PM_{2.5}$ ternary diagram (Ca-Fe-Al) for (a) winter, (b) spring, (c), summer, and (d) fall. The circles correspond to IMPROVE continental sites, and the color of the circle corresponds to the region. The circle size corresponds to the relative degree of seasonal variability (large circles representing highest seasonal variability). The Hawaii, Alaska, and Virgin Islands sites and regions are shown in Figures 5–7, respectively.

fractions at the West Coast region. Of all the regions and seasons the highest Fe fractions occurred in the Northwest region.

3.5. Seasonal Mean Trends in FD

Temporal trends in seasonal mean fine Fe (as a proxy for FD; recall section 2) from 2000 to 2014 are shown in Figures 9a–9d for winter, spring, summer, and fall, respectively. Trends (% yr^{-1}) are displayed as isopleths with triangles denoting locations of IMPROVE sites. Upward pointing triangles correspond to increased Fe and vice versa. Filled triangles reflect statistical significance of $p < 0.10$. As stated earlier, trends in Fe were corroborated with trends in Ca and in FD.

Seasonal mean trends were often driven by trends in individual months. For example, increased Fe concentrations in the spring in the Southwest were driven by regionally extensive, strong positive trends in March but less so in April or May. The increased trends in March reflected an earlier onset of the spring FD season in the Southwest by 1–2 weeks since 1995 [*Hand et al.*, 2016] and were associated with large-scale climate variability (e.g., Pacific Decadal Oscillation and El Niño–Southern Oscillation—ENSO) and associated drought. *Brahney et al.* [2013] and *Clow et al.* [2016] also reported increased wet deposition of mineral aerosols in the West during spring. Fine Fe concentrations in the Southwest during spring have increased significantly at

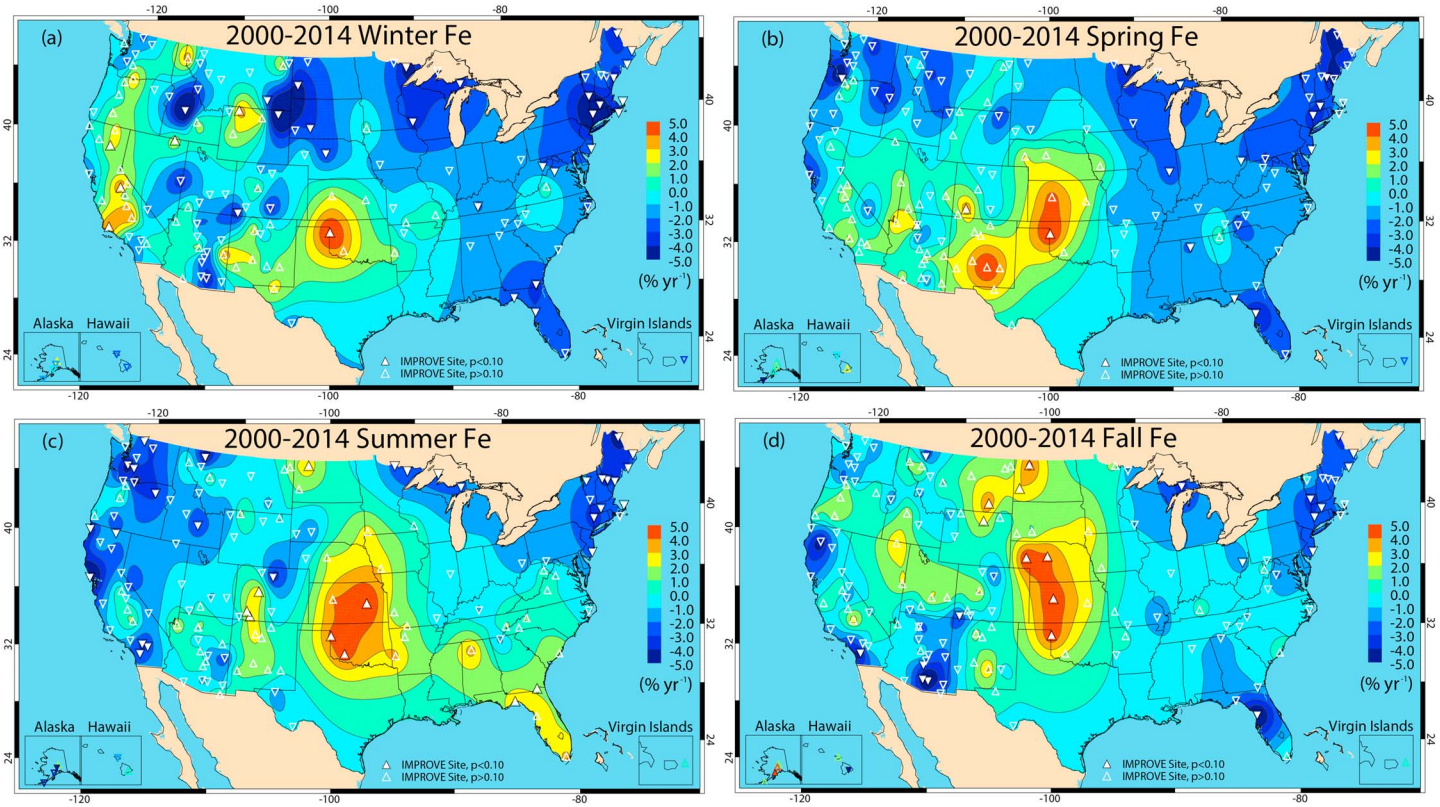

Figure 9. IMPROVE 2000–2014 seasonal mean trends in $PM_{2.5}$ iron (Fe) (% yr^{-1}) for (a) winter, (b) spring, (c) summer, and (d) fall.

many sites in New Mexico, and insignificant positive trends occurred at sites across the West and central United States (Figure 9b). Increased Fe concentrations in summer (Figure 9c), especially July, have occurred at many southeastern sites as well as at sites in the central United States in June. Sites in the central and northern Great Plains have also experienced increased Fe loadings in fall months, concurrent with drought in the region [*Brahney et al.*, 2013; *Cook et al.*, 2013; *Hoerling et al.*, 2014; *S.-Y. Wang et al.*, 2015]. Winter Fe has significantly increased at one site in Oklahoma and sites in California and Wyoming (Figure 9a). Fine Fe concentrations in most of the eastern United States have decreased during all seasons (with the exception of the Southeast in summer), and Fe has generally decreased during all seasons in the Northwest. These regions may have greater influence to Fe from combustions sources [*R. Wang et al.*, 2015].

Seasonal mean trends in FD $PM_{2.5}$ mass fractions are presented in Figures 10a–10d for winter, spring, summer, and fall, respectively. These results indicate changes in the FD contributions to the $PM_{2.5}$ mass budget and should be interpreted in the context of trends in other aerosol species. While other major aerosol species such as sulfate and organic carbon have decreased across the region [e.g., *Hand et al.*, 2012b, 2013, 2014], both the absolute concentration and relative contribution of FD to $PM_{2.5}$ mass have increased in the West in spring (Figure 10b). Contributions of FD to $PM_{2.5}$ mass have increased significantly at sites across the central United States in summer and fall (Figures 10c and 10d, respectively) and in the West and Southwest across all seasons but especially in winter and spring (Figures 10a and 10b, respectively). The increase in FD contribution in the East during summer (Figure 10c) reflects the decrease in sulfate and organic carbon mass which compose most of the $PM_{2.5}$ mass.

4. Discussion and Summary

The Southwest is one of the most studied areas in the United States with respect to ambient mineral dust because the highest concentrations in the United States occur there during spring months. During 2011–2014 FD contributed 50% or more to $PM_{2.5}$ in spring across the region, and CM contributed over 70% to PM_{10} during the same months. FD and CM had similar seasonal variability (spring maxima) at

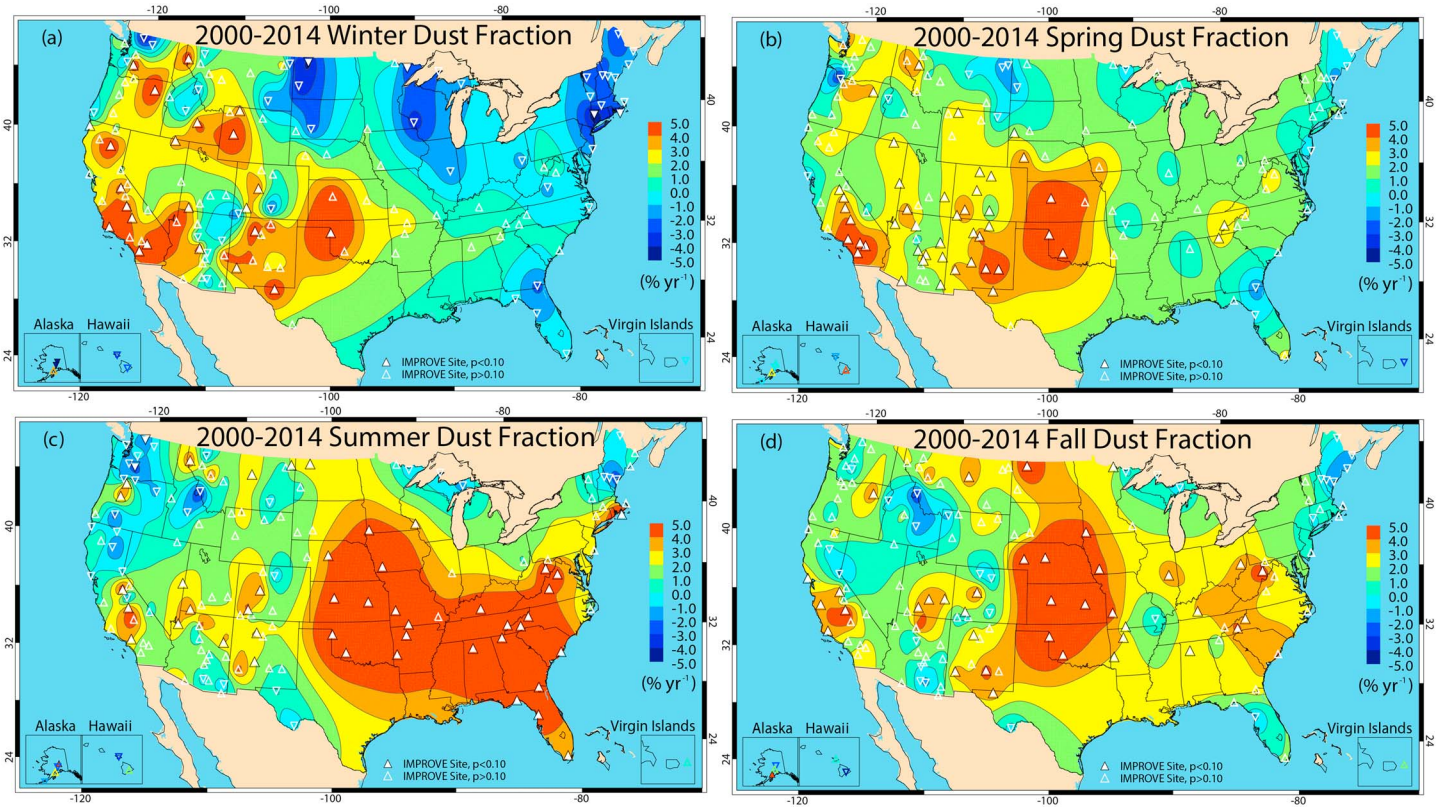

Figure 10. IMPROVE 2000–2014 seasonal mean trends in the contribution of fine dust to reconstructed $PM_{2.5}$ mass (% yr^{-1}) for (a) winter, (b) spring, (c) summer, and (d) fall.

sites in the Southwest, suggesting that similar dust sources influence the region due to the synergy of synoptic-scale meteorological patterns in spring and seasonal land use/land cover, precipitation, and soil moisture cycles. However, FD and CM seasonality decoupled at western sites farther north, with spring FD maxima and summer/fall CM maxima. CM concentrations were inversely correlated with elevation at western sites, suggesting that size-, elevation-, and land use/cover-limited CM generation, deposition, and transport was also evident in the strong gradients in the spatial distribution of CM. Differences in FD and CM seasonality may also indicate long-range versus local transport. In the northwestern United States, long-range transport may have influenced the decoupling of FD and CM seasonality, with FD spring maxima and CM summer/fall maxima (perhaps related to local sources or nondust CM). The strong decoupling of FD and CM was also observed in spring at Hawaiian and Alaskan sites and, along with shifts in elemental composition, suggested influence from Asian transport. Northwest regional mean elemental composition tended to cluster near the Hawaii and Alaska regions during spring, and FD seasonality also suggested long-range transport to these regions. These seasonal shifts in composition were unlike the rest of the western United States, where highly uniform seasonal and spatial elemental ratios were observed. Variable spring seasonal mean trends in the West were driven by significant increased regional concentrations in March, less so in April and May [*Hand et al.*, 2016]. FD contributions to the $PM_{2.5}$ mass budget have increased across the West during all seasons but especially in winter and spring. Increased dust concentrations in this region have important implications, including contributions to overall health effects [e.g., *Morman and Plumlee*, 2013], dust-borne diseases such as valley fever [*Centers for Disease Control and Prevention (CDC)*, 2013], transport of bioaerosols [*Hallar et al.*, 2011], the hydrologic cycle of the Intermountain West including snow melt behavior [e.g., *Painter et al.*, 2007] and precipitation formation [*Sorooshian et al.*, 2013], highway and aviation safety [*Ashley et al.*, 2015], and visibility degradation. In fact, the West has experienced less improvement in visibility on the haziest days compared to the East [*Hand et al.*, 2014].

The influence of North African dust to the eastern United States during 2011–2014 was suggested by the shift in elemental composition in summer especially for sites in the Southeast, Appalachia, and Virgin Islands

regions. In addition, FD was up to 3 times higher in summer relative to other months at most sites in the East. FD and CM seasonality was strongly decoupled at eastern sites, with the exception of the Virgin Islands site. FD contributed 20–30% of $PM_{2.5}$ mass in summer across the Southeast and up to 30–40% in southern Florida, while CM was typically 40% of PM_{10} in the Southeast (50–60% in southern Florida). Trend analysis suggested that FD concentrations have also increased at many of these sites, as has the contribution of FD to $PM_{2.5}$ mass.

In the central United States, FD and CM concentrations were elevated in summer and fall across the region. FD contributed up to 30% of $PM_{2.5}$ mass, and CM contributed over 70% in fall and stretched from the central United States into the northern Great Plains. FD maxima occurred in summer, while CM was elevated from summer through fall. Sites in the central United States corresponded to some of the highest Ca fractions of any region, especially in fall. High Ca fractions may be linked to calcareous geological formations and soils [e.g., *Gustavson and Holliday*, 1999; *Halfen and Johnson*, 2013] and/or agricultural activity that also results in high nitrate levels [e.g., *Pitchford et al*., 2009]. *Sullivan et al*. [2007] reported that nitrate was more associated with calcite-rich dust, and *Lee et al*. [2008] reported that a fraction (~10%) of the total nitrate was associated with coarse calcium nitrate at an agricultural site in Illinois (Bondville), even given the elevated levels of ammonium. High Ca fractions could affect dust reactivity and solubility in the atmosphere and have implications for its lifetime and heterogeneous chemistry [e.g., *Sullivan et al*., 2007]. Transport of dust and dust-related CM from this region also has important implications for topsoil removal and regional air quality and visibility. In fact, in October 2012 dust emitted from the central United States was transported and reportedly influenced air quality and visibility as far southeast as Great Smoky Mountains National Park in Tennessee (earthobservatory.nasa.gov/IOTD/view.php?id = 79459; J. Renfro, personal communication, 2016). Positive trends in the absolute and relative FD contributions to $PM_{2.5}$ mass identified in high-dust seasons in this region will likely exacerbate these issues.

The average spatial and seasonal patterns reported here are first steps in understanding the large-scale variability of FD and CM across the United States. As regulated sources of secondary aerosols continue to decline [e.g., *Hand et al*., 2012b, 2013, 2014], the importance of FD and CM to the mass budget will continue to increase, especially given that the regions and seasons with the highest concentrations have also experienced the largest increases. This relative increase in FD has important implications for air quality, visibility, heterogeneous chemistry, ecology, hydrology, health and safety, land management, and climate change. Mitigating the impacts will require identifying source regions and characteristics and causal mechanisms for dust episodes. Efforts such as the newly formed National Wind Erosion Network [*Webb et al*., 2016] should help identify and improve our understanding of dust sources and emissions in the United States, as will new satellite technology and retrieval algorithms (e.g., GOES-R, MODIS Deep Blue 2 Collection 6 [*Baddock et al*., 2016]) for identifying dust sources and transport. Causal mechanisms for increased dust, such as influences of climate change and large-scale climate variability on drought and soil properties, and anthropogenic land disturbances should be studied in order for land and air quality managers to develop effective strategies for managing future dust impacts.

Acknowledgments

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