Seasonal composition of remote and urban fine particulate matter in the United States

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[1] Speciated aerosol composition data from the rural Interagency Monitoring for Protected Visual Environments (IMPROVE) network and the Environmental Protection Agency's urban/suburban Chemical Speciation Network (CSN) were combined to evaluate and contrast the $PM_{2.5}$ composition and its seasonal patterns at urban and rural locations throughout the United States. We examined the 2005–2008 monthly and annual mean mass concentrations of $PM_{2.5}$ ammonium sulfate (AS), ammonium nitrate (AN), particulate organic matter (POM), light-absorbing carbon (LAC), mineral soil, and sea salt from 168 rural and 176 urban sites. Urban and rural AS concentrations and seasonality were similar, and both were substantially higher in the eastern United States. Urban POM and LAC concentrations were higher than rural concentrations and were associated with very different seasonality depending on location. The highest urban and rural POM and LAC concentrations occurred in the southeastern and northwestern United States. Wintertime peaks in AN were common for both urban and rural sites, but urban concentrations were several times higher, and both were highest in California and the Midwest. Fine soil concentrations were highest in the Southwest, and similar regional patterns and seasonality in urban and rural concentrations suggested impacts from long-range transport. Contributions from sea salt to the $PM_{2.5}$ budget were non-negligible only at coastal sites. This analysis revealed spatial and seasonal variability in urban and rural aerosol concentrations on a continental scale and provided insights into their sources, processes, and lifetimes.

1. Introduction

[2] The importance of the role of aerosols in visibility degradation, climate change, and health effects is well known. Because of their ability to scatter and absorb solar radiation, aerosols degrade visibility in both remote and urban locations [e.g., *Malm et al.*, 1994; *Quinn et al.*, 2001, 2004; *Brewer and Adloch*, 2005; *Tsai*, 2005; *Malm and Hand*, 2007; *Singh et al.*, 2008] and can have a direct effect on climate [e.g., *Intergovernmental Panel on Climate Change (IPCC)*, 2007; *Ramanathan and Carmichael*, 2008; *Kopp and Mauzerall*, 2010]. Indirect effects on climate include their role in cloud processing and lifetime [e.g., *Boucher*, 1995; *Williams et al.*, 2001; *Lohmann and Feichter*, 2005]. Aerosols are also a health concern and linked to respiratory and cardiovascular disease [e.g., *Pope and Dockery*, 2006]. The magnitude of the effects of aerosols in all of these areas depends on their size and chemical composition. For example, hygroscopic aerosols, such as inorganic salts, absorb water and scatter more sunlight than aerosols that are weakly or nonhygroscopic, such as carbonaceous aerosols or mineral dust, resulting in significantly higher visibility degradation in high relative humidity environments [e.g., *Day and Malm*, 2001; *Malm et al.*, 2005]. Hygroscopic species are also more efficient cloud condensation nuclei [e.g., *Petters et al.*, 2009], while light-absorbing aerosols can affect cloud lifetimes through the semidirect effect [e.g., *Hansen et al.*, 1997; *Lohmann and Feichter*, 2001]. Highly reflective aerosols, such as sulfates and nitrates, result in a direct cooling effect compared to aerosols with low single scattering albedo that absorb solar radiation, such as light-absorbing carbon, some organic carbon species, and some components of mineral soil [e.g., *Kiehl and Briegleb*, 1993; *Haywood and Ramaswamy*, 1998; *IPCC*, 2007]. Health effects may also differ because of different aerosol composition [*U.S. Environmental Protection*

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Agency (*EPA*), 2009a]. *Mauderly and Chow* [2008] discuss the adverse health effects associated specifically with organic aerosols. Uncertainties surrounding the role of aerosols in climate, visibility, and health studies can be significant because chemical composition data may not be widely available in space and time.

[3] Reducing uncertainties associated with aerosol effects requires observations of aerosol speciation from long-term, spatially extensive, ground-based networks. The Interagency Monitoring of Protected Visual Environments (IMPROVE) program is a cooperative effort designed to establish current visibility and aerosol conditions in mandatory Class I areas (visibility-protected federal areas), to identify chemical species and emission sources responsible for existing anthropogenic and natural visibility impairment, to document long-term trends for assessing progress toward the national visibility goal, and, with the enactment of the Regional Haze Rule (RHR) (U.S. Federal Register 64, 1999), to provide regional haze monitoring representing all Class I areas where practical [*Malm et al.*, 1994, 2004]. The IMPROVE network began monitoring in remote and rural areas in 1987 with approximately 30 sites. Currently the network operates 170 sites in mostly remote and rural locations around the United States, including a few urban sites. Speciated urban aerosol concentrations can be determined from the EPA's $PM_{2.5}$ speciation program, established in 1997 as a complement to the $PM_{2.5}$ Federal Reference Method (FRM) mass network [*Frank*, 2006; National Primary and Secondary Ambient Air Quality Standards for Particulate Matter, U.S. Federal Register 50, Appendix L, 2009]. The Speciated Trends Network (STN) and other urban monitoring sites are collectively known as the U.S. EPA's Chemical Speciation Network (CSN) and were deployed in the fall of 2000 [*EPA*, 2004a]. The objectives of EPA's CSN are to track progress of emission reduction strategies through the characterization of trends, evaluation of air quality modeling and source apportionment activities, support of regulatory efforts such as the RHR, and support of health effects and exposure studies. The CSN operates approximately 50 long-term trend sites, with another ~150 sites operated by state, local, and tribal agencies, primarily in urban/suburban settings.

[4] Spatial and temporal patterns of the absolute concentration of the species that comprise fine particulate matter, as well as the $PM_{2.5}$ mass budget, can vary significantly depending on species and location. Previous results from the IMPROVE network demonstrated that rural and remote aerosol species vary seasonally and spatially across the United States [*Malm et al.*, 1994, 2004, 2007]. For example, *Malm et al.* [2004] reported 2001 monthly mean speciated aerosol concentrations from IMPROVE monitors across the United States and demonstrated that ammonium sulfate (AS) concentrations were highest in the eastern United States and dominated fine mass in the summer. Particulate organic matter (POM) concentrations were highest in spring, summer, or fall months, depending on sources and location, and ammonium nitrate (AN) concentrations were highest during winter months, because of favorable formation mechanisms. Fine soil contributed up to 40% to fine mass during spring at sites in the western United States. The seasonal patterns in secondary aerosols such as AS, AN, and secondary POM are driven by meteorological parameters, whereas the seasonality

in primary aerosols, such as primary POM or LAC, is primarily determined by source emissions that vary significantly depending on location and species. For example, *Malm et al.* [2004] demonstrated that POM had the opposite seasonality at IMPROVE urban sites such as Phoenix, Arizona, and Puget Sound, Washington, compared to nearby rural sites, most likely because of different sources in urban locations or meteorological effects such as winter inversions. Other studies have also showed that urban carbonaceous aerosols may demonstrate different seasonality relative to rural carbonaceous aerosols at nearby sites [*Malm et al.*, 2004; *Tanner et al.*, 2004; *Chow et al.*, 2006; *Ding et al.*, 2008; *Dabek-Zlotorzynska et al.*, 2011; *Holden et al.*, 2011; *Malm et al.*, 2011; *Rattigan et al.*, 2011].

[5] Other national monitoring networks in the United States, including the Clean Air Status and Trends Network (CASTNET) and the National Atmospheric Deposition Program (NADP) provide spatial and temporal information on aerosols. CASTNET collects weekly integrated open-air samples for the analysis of atmospheric sulfur and nitrogen species. Comparisons of IMPROVE and CASTNET data at collocated sites have been the focus of several studies [e.g., *Ames and Malm*, 2001; *Malm et al.*, 2002; *Sickels and Shadwick*, 2008; *Lavery et al.*, 2009] and generally suggest good agreement for sulfate ion concentrations, less so for nitrate ion concentrations [*Lavery et al.*, 2009]. Using weekly precipitation samples collected by the NADP, *Lehmann and Gay* [2011] report spatial and temporal trends for sulfate and nitrate concentrations in precipitation that suggest similar spatial patterns to particulate concentrations in the atmosphere. Integration of data from spatially extensive networks provides insights into the differences in the seasonal and spatial variability in aerosol concentrations on regional and continental scales and can be used to infer sources, transport, and urban excess. In addition, these data sets are useful for evaluation of regional and global models and satellite retrievals [e.g., *Park et al.*, 2004, 2006; *Heald et al.*, 2006; *Chin et al.*, 2007; *Liu et al.*, 2008; *Drury et al.*, 2010].

[6] The purpose of this work is to compare and contrast absolute $PM_{2.5}$ aerosol concentrations and the $PM_{2.5}$ budget in rural and urban areas across the United States using data from both the IMPROVE network and the CSN. We present 2005–2008 monthly and annual mean $PM_{2.5}$ AS, AN, POM, LAC, mineral soil, and sea salt concentrations from both networks. This work builds on the previous study by *Malm et al.* [2004] by examining data from more recent years from the IMPROVE monitoring network. Following the introduction, we provide a description of the monitoring networks and the methodology for estimating aerosol mass concentrations (section 2). Spatial variability in monthly mean concentrations across the United States is provided in section 3, followed by a discussion and summary in section 4.

2. Methodology

2.1. Monitoring Networks

[7] The IMPROVE network collects 24 h samples every third day from midnight to midnight local time. A list of sites and metadata is provided by *Hand et al.* [2011]. The IMPROVE sampler consists of four independent modules (A, B, C, and D). Each module incorporates a separate inlet, filter pack, and pump assembly. Modules A, B, and C are

equipped with a 2.5 μm cyclone that allows for sampling of particles with aerodynamic diameters less than 2.5 μm, while module D is fitted with a PM$_{10}$ inlet to collect particles with aerodynamic diameters less than 10 μm. Each module contains a filter substrate specific to the analysis planned [*Malm et al.*, 2004].

[8] Module A is equipped with a Teflon® filter that is analyzed for PM$_{2.5}$ gravimetric fine mass and elemental analysis. Elemental analysis is performed by X-ray florescence (XRF). Anion (sulfate, nitrate, nitrite, and chloride) concentrations are determined from ion chromatography using a nylon filter in Module B preceded with a sodium carbonate coated denuder that is changed annually. Module C utilizes quartz fiber filters that are analyzed by thermal optical reflectance (TOR) for particulate organic carbon (OC) and LAC [*Chow et al.*, 1993], also known as elemental carbon (EC). Finally, Teflon filters utilized by module D are used to determine PM$_{10}$ aerosol mass concentrations gravimetrically. OC concentrations reported by IMPROVE are corrected for a positive additive artifact [*Watson et al.*, 2009; *Dillner et al.*, 2009; *Chow et al.*, 2010] but not a multiplicative negative artifact [*Malm et al.*, 2011]. Corrections will be discussed further in section 2.2. Additional details regarding IMPROVE sampling, including artifact corrections (both for OC and anion concentrations), are provided by *Hand et al.* [2011]. Concentrations are reported at ambient conditions. All IMPROVE data, metadata, and detailed descriptions of the network operations and data analysis and visualization results are available for download from http://views.cira.colostate.edu/fed/.

[9] The CSN collects 24 h samples every third or sixth day, on the same sampling schedule as IMPROVE. A list of sites is provided by *Hand et al.* [2011]. Historically, CSN utilized several types of samplers, including the Thermo Andersen RAAS, Met One SASS, and the URG MASS. The specific sampler employed at a given site was chosen by the state, local, or tribal agency; however, the Met One had been the predominant sampler used [*EPA*, 2004a]. All samplers utilize a PM$_{2.5}$ inlet and three channels containing Teflon, nylon, and quartz filters. PM$_{2.5}$ gravimetric mass and elemental compositions are analyzed from the Teflon filter, anions and cations from the nylon filter preceded with a magnesium oxide or sodium carbonate coated denuder that is changed every ~3 months [*EPA*, 2000], and carbon from the quartz filter. The carbon analysis was historically performed using thermal optical transmittance (TOT), using a method based on the National Institute of Occupational Safety and Health (NIOSH) protocol. Because IMPROVE samplers and TOR analysis produce different OC and LAC concentrations compared to the CSN samplers and TOT analysis [e.g., *Watson et al.*, 2009; *Malm et al.*, 2011; *Rattigan et al.*, 2011], the CSN has transitioned to TOR analysis for consistency with the IMPROVE network [*EPA*, 2004a, 2009b; *Rattigan et al.*, 2011]. In addition to the transition from TOT to TOR, the EPA decided to replace the carbon channel sampling and analysis methods with a URG-3000N as part of the effort toward consistency with the IMPROVE sampler. The conversion began in May 2007 with 56 sites, followed by another 63 sites in April 2009 and 78 additional sites in October 2009 [*EPA*, 2009b]. Data are reported at ambient conditions. The CSN cold ships their filters, while the IMPROVE network does not. CSN

data can be downloaded from http://views.cira.colostate.edu/fed/ or http://www.epa.gov/ttn/airs/airsaqs/.

2.2. Estimates of Aerosol Mass Concentrations

[10] PM$_{2.5}$ mass is composed of a complex mixture of aerosol species, including sulfates, nitrates, carbonaceous aerosols, mineral soil, and sea salt. Determining the specific molecular forms of these species requires additional measurements than those regularly performed by the IMPROVE or CSN networks; therefore, assumptions are made regarding their particular molecular forms in order to compute the PM$_{2.5}$ mass budget. Previous researchers investigated the validity of these assumptions and found them to be reasonable approximations of aerosol composition [e.g., *Malm et al.*, 1994; *Lowenthal and Kumar*, 2006; *Malm and Hand*, 2007; *Pitchford et al.*, 2007; *Malm et al.*, 2011; *Simon et al.*, 2011]. Similar to past studies, we assumed the PM$_{2.5}$ fine mass was composed of AS, AN, POM, LAC, soil and sea salt. Reconstructed fine mass (RCFM) is the sum of these species. Table 1 summarizes the assumptions used in computing these species. We assumed sulfate is in the form of fully neutralized AS as an upper bound of mass associated with dry sulfate, although the degree of sulfate neutralization can vary spatially and temporally [e.g., *Gebhart et al.*, 1994; *Liu et al.*, 1996; *Day et al.*, 1997; *Lowenthal et al.*, 2000; *Lefer and Talbot*, 2001; *Quinn et al.*, 2002; *Chu*, 2004; *Hogrefe et al.*, 2004; *Schwab et al.*, 2004; *Tanner et al.*, 2004; *Brewer and Adloch*, 2005; *Zhang et al.*, 2005; *Simon et al.*, 2011]. Without additional measurements of ammonium ion concentration, the degree of neutralization is unknown. Nitrate was assumed to be in the form of AN, although *Lee et al.* [2008] showed that in many locations nitrate is associated with the coarse mode from reactions of gas phase nitric acid with sea salt or calcium carbonate. In these situations the nitrate measured in the fine mode is actually the tail of coarse mode nitrate. POM was computed assuming an average molecular weight per carbon weight for OC of 1.8 based on the work of *Malm and Hand* [2007], although it is also spatially and temporally variable, and typical values could range from 1.2 to 2.6 [*Turpin and Lim*, 2001; *El-Zanan et al.*, 2005; *Malm and Hand*, 2007; *Malm et al.*, 2011; *Simon et al.*, 2011]. Although the POM multiplier is most likely lower in urban regions [*Turpin and Lim*, 2001; *Malm et al.*, 2011], we applied the same value to data from both networks. LAC is a primary aerosol produced by combustion sources. In urban areas LAC sources are associated with diesel engines or residential heating, while biomass smoke is a predominant source in rural areas. Biomass smoke emissions are sources of both graphitic carbon and brown carbon, which refers to light-absorbing organic aerosols that absorb light in the UV spectrum [e.g., *Hecobian et al.*, 2010]. LAC refers to carbonaceous aerosols that absorb light, such as EC, graphitic carbon (soot), or brown carbon, as determined by its optical and thermal properties. The thermal/optical method used here has been known to characterize brown carbon as EC [*Hand et al.*, 2005]. We use "LAC" instead of "EC" based on the recommendation of *Bond and Bergstrom* [2006] and to avoid possible improper classification. Soil mass concentrations were estimated by a general method that sums the oxides of elements that are typically associated with soil (Al$_2$O$_3$, SiO$_2$, CaO, K$_2$O, FeO, Fe$_2$O$_3$, TiO$_2$), with a correction for other

Table 1. Form of Molecular Species Assumed

PM$_{2.5}$ Aerosol Species	Calculated	Assumptions
Ammonium sulfate AS = (NH$_4$)$_2$(SO$_4$)	1.375[SO$_4^{2-}$]	Sulfate is assumed to be fully neutralized. Same assumption for both IMPROVE[a] and CSN[a] data.
Ammonium nitrate AN = NH$_4$NO$_3$	1.29[NO$_3^-$]	Nitrate is assumed to be ammonium nitrate. Same assumption for both IMPROVE and CSN data.
Particulate organic matter (POM)	1.8[OC]	Derived from organic carbon (OC) assuming an average organic molecule is 55% carbon. See text regarding IMPROVE and CSN carbon data.
Light-absorbing carbon (LAC)	LAC	See text regarding IMPROVE and CSN carbon data.
Soil	2.2[Al] + 2.49[Si] + 1.63[Ca] + 2.42[Fe] + 1.94[Ti]	Based on the IMPROVE soil formula [*Malm et al.*, 1994]. Same assumption for both IMPROVE and CSN data.
Sea salt (SS)	1.8[Cl$^-$] or 1.8[Cl]	Sea salt is 55% chloride by weight. IMPROVE sea salt is computed from chloride ion data, while CSN is computed from chlorine concentrations (Cl$^-$ not available).
Reconstructed fine mass (RCFM)	[AS] + [AN] + [POM] + [LAC] + [Soil] + [SS]	—

[a]IMPROVE, Interagency Monitoring for Protected Visual Environments network; CSN, Chemical Speciation Network.

compounds such as MgO, Na$_2$O, H$_2$O and carbonates [*Malm et al.*, 1994]. Fine soil concentrations are most likely associated with the tail of the coarse mode characterized with the PM$_{2.5}$ measurement and could include a fly ash component. Sea salt concentrations were computed using a factor of 1.8 multiplied by the chloride ion concentration (IMPROVE) and chlorine concentration from XRF (CSN) because sodium ion data were not available from both networks (sea salt is 55% Cl$^-$ by weight as defined by the composition of seawater by *Seinfeld* and Pandis [1998], *Pitchford et al.* [2007], and *White* [2008]). Sea salt estimates are likely an underestimation due to the depletion of chloride during the reaction of gaseous nitric acid with sea salt, producing sodium nitrate particles and the release of gaseous HCl.

[11] Carbonaceous aerosol measurements are sensitive to sampling artifacts and thermal/optical analyses that result in different OC and LAC concentrations depending on the protocol [e.g., *Chow et al.*, 2004; *Rattigan et al.*, 2011]. We combined data from three protocols: (1) IMPROVE data collected using a sampler with high face velocity, analyzed using TOR, and corrected for a positive artifact; (2) pretransition CSN data collected with samplers having relatively low face velocities, analyzed with a modified NIOSH TOT, and uncorrected for artifacts; (3) post-transition CSN data collected and analyzed similar to the IMPROVE protocol but uncorrected for positive artifacts. The impacts of these different sampling protocols on carbonaceous aerosol concentrations have been examined by others [e.g., *Chow et al.*, 2004, 2010; *Watson et al.*, 2009; *Malm et al.*, 2011; *Rattigan et al.*, 2011]. By examining collocated IMPROVE and pretransition CSN data from 2005 to 2006, *Malm et al.* [2011] developed a methodology to reconcile differences in order to combine the data sets in an integrated analysis.

[12] We adopted the methods of *Malm et al.* [2011] to adjust the CSN pretransition carbon data obtained prior to the 2007–2009 transition to the IMPROVE protocol. Adjustments were applied in order to integrate the data as well as to provide a consistent CSN data set pretransition

and post-transition. The adjustments included correcting the CSN data for a positive artifact, reducing the CSN OC concentrations to reflect the IMPROVE negative multiplicative artifact, and increasing LAC to account for differences in TOT versus TOR analyses. For CSN post-transition data we applied a positive artifact correction of 0.3 μg m^{-3} to approximate the median of OC concentrations from field and backup filters collected at select CSN sites. Analyses of these data showed little spatial or seasonal variation supporting the use of a constant positive artifact correction. Our value is consistent with the work of *Rattigan et al.* [2011] who reported OC backup filter concentrations at urban and rural sites in New York that ranged from 0.15 to 0.45 μg m^{-3} depending on location and season, with less seasonality for data collected at the urban location.

[13] As discussed in section 2.3, adjustments to CSN data resulted in close agreement between IMPROVE and CSN data collected from 2005 to 2008 at collocated sites. In addition, there were no discontinuities in carbon concentrations over time between adjusted CSN pretransition data and post-transition data, and no significant differences in the urban/rural comparisons before and after the transition. The negative multiplicative OC artifact applied to the CSN pretransition data does introduce an additional bias relative to ambient concentrations. However, this uncertainty is less than that in the organic multiplier used to compute POM that likely varies from 1.3 to 1.9 depending on location and season [*Malm and Hand*, 2007; *Malm et al.*, 2011; *Simon et al.*, 2011].

2.3. IMPROVE and CSN Collocated Data Comparisons

[14] The IMPROVE and CSN networks operate collocated samplers in several urban/suburban sites. Data from collocated sites that met the completeness criteria (see section 2.4) were compared in order to identify relative biases between IMPROVE and CSN speciated aerosol concentrations. We used 2005–2008 monthly mean data from collocated sites at
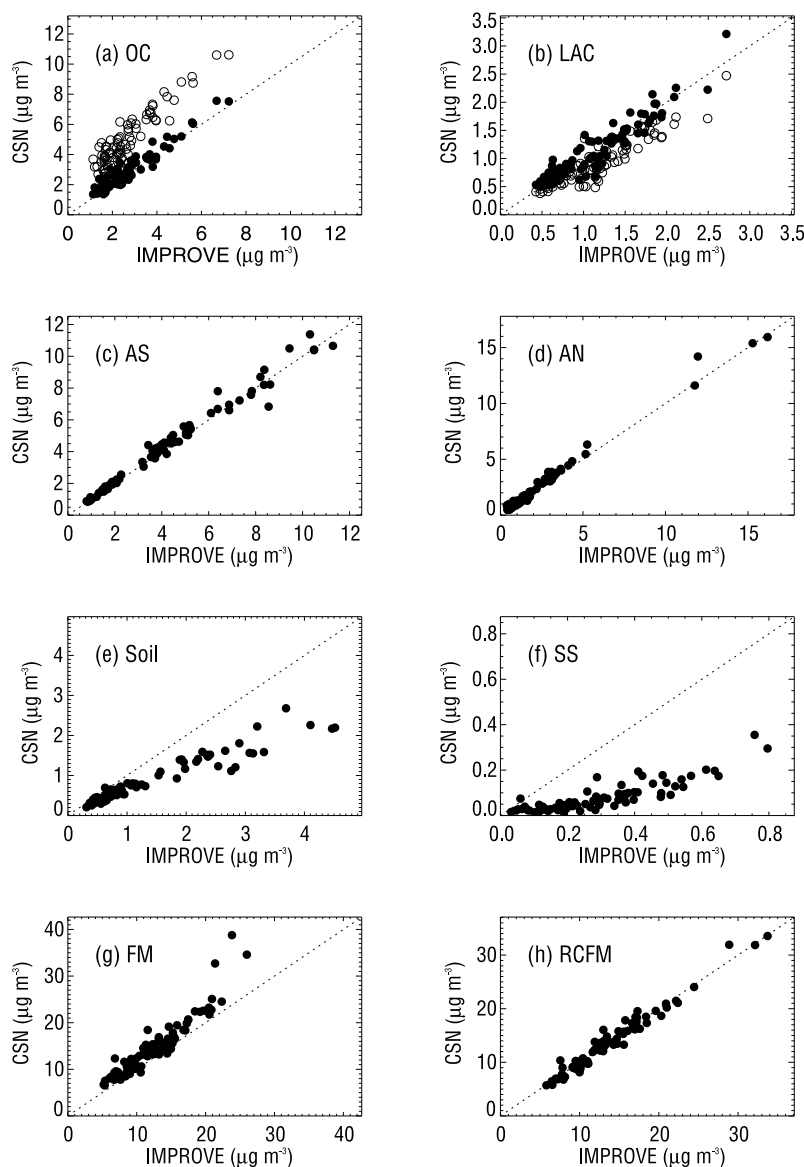
Figure 1. Comparisons of 2005–2008 monthly mean aerosol mass concentrations (μg m^{-3}) for seven collocated Interagency Monitoring for Protected Visual Environments (IMPROVE) and Chemical Speciation Network (CSN) sites (see text) for (a) adjusted organic carbon (OC) as filled circles and unadjusted OC as open circles, (b) adjusted light-absorbing carbon (LAC) as filled circles and unadjusted LAC as open circles, (c) ammonium sulfate (AS), (d) ammonium nitrate (AN), (e) soil, (f) sea salt (SS), (g) PM$_{2.5}$ gravimetric fine mass (FM), and (h) PM$_{2.5}$ reconstructed fine mass (RCFM = AS+AN+POM+LAC+Soil+SS).

Baltimore, Maryland, Birmingham, Alabama, Fresno, California, New York City, New York, Phoenix, Arizona, Puget Sound, Washington, and Washington, DC. We compared AS, AN, OC, LAC, soil, sea salt, PM$_{2.5}$ gravimetric fine mass (FM), and RCFM.

[15] Comparisons of monthly mean IMPROVE and CSN species mass concentrations for all collocated sites are presented in Figure 1. A summary of results is provided in Table 2. As reported by *Hyslop and White* [2008], precisions for IMPROVE data were 4%, 10% and 33%, for sulfate, nitrate and chloride ions, respectively, 17% and 22% for OC and LAC, respectively, and 6% for PM$_{2.5}$ gravimetric mass, based on data from collocated IMPROVE samplers.

Comparisons between collocated IMPROVE and CSN data suggest that with the exception of soil and sea salt, errors for other species were less than 20% and within measurement uncertainty. CSN concentrations were somewhat higher than IMPROVE concentrations for most species (positive relative biases correspond to higher CSN concentrations), but data from the two networks were fairly highly correlated. The relative biases in AS and AN were 4.2% and 15.2%, respectively, with a correlation of 0.99 for both species. Some of the discrepancy between ion data from the two networks could be due to the fact that while IMPROVE applies artifact corrections to ion data, CSN does not. Differences could also be due in part to CSN cold-shipping their

Table 2. Comparisons Between Seven Collocated IMPROVE and CSN Sites for Monthly Mean Data From 2005 to 2008

Statistic	OC$_{unadj}$[a]	LAC$_{unadj}$[b]	OC$_{adj}$[c]	LAC$_{adj}$[d]	AS[e]	AN[f]	Soil	Sea Salt[g]	Fine Mass	RCFM
Average IMPROVE (μg m^{-3})	2.7	1.2	2.6	1.2	4.0	2.2	1.3	0.3	12.5	13.8
Average CSN (μg m^{-3})	5.0	1.0	2.8	1.2	4.1	2.5	0.8	0.07	14.7	13.8
Bias[h] (%)	96.5	−18.6	8.7	1.1	4.2	15.2	−32.1	−72.8	18.6	−0.15
Error[i] (%)	87.3	20.5	9.3	11.7	5.3	10.7	32.2	76.5	16.4	4.2
r	0.92	0.92	0.96	0.93	0.99	0.99	0.96	0.87	0.94	0.98
IMP/CSN[j]	0.53	1.3	0.93	1.0	0.97	0.91	1.6	3.7	0.9	1.0
Number of data points (N)	82	82	84	84	84	84	84	84	84	84

[a]OC$_{unadj}$ refers to comparisons between IMPROVE organic carbon and unadjusted CSN organic carbon.

[b]LAC$_{unadj}$ refers to comparisons between IMPROVE light-absorbing carbon and unadjusted CSN light-absorbing carbon.

[c]OC$_{adj}$ refers to comparisons between IMPROVE organic carbon and adjusted CSN organic carbon.

[d]LAC$_{adj}$ refers to comparisons between IMPROVE light-absorbing carbon and adjusted CSN light-absorbing carbon.

[e]AS = 1.375[sulfate ion].

[f]AN = 1.29[nitrate ion].

[g]Sea salt = 1.8[chloride ion] for IMPROVE and 1.8[chlorine] for CSN.

[h]Bias = $\frac{1}{N}\sum_i^N \frac{\bar{X}_i - \bar{Y}_i}{\bar{Y}_i}$; $\bar{X}_i$ and $\bar{Y}_i$ are the monthly mean data for CSN and IMPROVE concentrations, respectively. The number of data points is given by N.

[i]Error = $\text{median}\left(\left|\frac{\bar{X}_i - \bar{Y}_i}{\bar{Y}_i}\right|\right)$.

[j]IMP/CSN refers to the ratio of the average IMPROVE to average CSN concentrations.

filters while IMPROVE does not. *Gego et al.* [2005] compared average CSN and IMPROVE sulfate (derived from sulfur data) and nitrate data for three nearby sites (not collocated) in the eastern United States during 1 July 2001 to 31 July 2002. While the data from their comparisons showed a similar degree of high correlation, CSN sulfate concentrations were somewhat lower than IMPROVE concentrations (IMPROVE/CSN = 1.05) and CSN nitrate concentrations were larger (IMPROVE/CSN = 0.78). However, *Rattigan et al.* [2011] report very similar agreements to those reported here for sulfate concentrations measured at collocated sites in New York.

[16] The collocated comparisons of carbon data suggested that the adjustments derived by *Malm et al.* [2011] and applied to the CSN carbon data were appropriate and effective. The errors and relative biases between unadjusted CSN carbon and IMPROVE carbon data were much higher than for the adjusted data. Unadjusted data refer to CSN data prior the switch to the URG-3000N sampler. Errors and relative biases in unadjusted OC were 87.3% and 96.5%, respectively, and improved to 9.3% and 8.7%, respectively, for adjusted OC. Errors and relative biases for unadjusted LAC were 20.5% and −18.6%, respectively, and 11.7% and 1.1% for adjusted LAC, respectively. The ratio of mean IMPROVE to CSN data of 0.53 and 1.3 for OC and LAC, respectively, improved to 0.93 and 1.0, respectively, after the adjustment. Additional comparisons of unadjusted and adjusted carbon data are separated by site and year and reported in Appendix A of *Hand et al.* [2011].

[17] IMPROVE soil and sea salt concentrations were much higher than collocated CSN soil and sea salt concentrations, with relative biases of −32.1% and −72.8% for soil and sea salt, respectively. *Rattigan et al.* [2011] reported CSN calcium concentrations that were 14% lower than IMPROVE calcium concentrations at collocated sites using a Met One sampler and suggested that the size selection of the Met One sampler removed more coarse particles, a possible explanation as to some of the discrepancy between the two estimates. Also recall that IMPROVE sea salt concentrations were computed using chloride ion concentrations, whereas CSN sea salt concentrations were computed using chlorine concentrations. IMPROVE chlorine concentrations

were often below detection limit and significantly lower than both the IMPROVE chloride and CSN chlorine concentrations. The physical cause for the relative biases remains unidentified and so no adjustments were applied. Additional comparisons of individual elemental species used to compute soil are reported in Appendix A of *Hand et al.* [2011]. The relative biases in soil and sea salt mass concentrations are sufficiently large that comparisons between those species should be treated as semiquantitative.

[18] The relative bias in RCFM was low (−0.15%) because of close agreement in the concentration of major species, especially the adjusted CSN carbon concentrations, and compensating biases. However, CSN FM concentrations were higher than IMPROVE FM concentrations on average, with a relative bias of 18.6%. OC concentrations measured by CSN samplers prior to 2007 were higher than those obtained with IMPROVE samplers because of sampling artifacts and different analytical techniques, which would also affect FM measurements. Negative artifacts associated with the sampling systems may also affect FM measurements on Teflon filters and contribute to the relative bias in FM concentrations between the two networks. While CSN carbon data have been adjusted for sampling artifacts to agree with IMPROVE carbon data, FM data have not. Comparisons of CSN FM to RCFM data may be affected by this discrepancy, which could be an issue now that CSN has completed the transition to the URG-3000N sampling system for its carbon monitoring but maintains its FM monitoring system [*Rattigan et al.*, 2011; *Malm et al.*, 2011]. Some of these issues might be accounted for using an approach such as SANDWICH (sulfate, adjusted nitrate, derived water, inferred carbonaceous material balance [*Frank*, 2006]), a method that adjusts for retained particle-bound water, infers carbonaceous mass and accounts for loss of nitrates from a Teflon filter.

2.4. Spatial and Temporal Aggregations

[19] Monthly and annual mean data from a 4 year time period (2005–2008) are examined in this paper. We applied completeness criteria to ensure that the data were representative of the entire time period. Fifty percent completeness of the data (2 years of valid monthly mean data) for a given site

was required to be included in the analysis. Half of the total observations in a given month had to be valid for a monthly mean. In addition, 67% of each 3 month season was required for an annual mean (a total of 8 months, but representative across each season, was required for an annual mean). Seasons correspond to winter (December, January, February), spring (March, April, May), summer (June, July, August), and fall (September, October, November). These criteria were applied for each species separately. Values below the minimum detection limit (MDL) were handled according to how they were reported by each network, i.e., we made no substitutions for values below MDLs. For the IMPROVE network, ion and carbon data were reported below their MDLs. XRF data were reported as zero if they were below MDLs. Data from the CSN were handled similarly. Applying the completeness criteria resulted in 168 IMPROVE sites and 176 CSN sites being used in the analyses. Monthly mean data presented here are available from *Hand et al.* [2011, Appendix B and D].

[20] Investigating regional aerosol concentrations required grouping sites into geographic regions. The IMPROVE regions were empirically defined based on site location and magnitudes and seasonal distribution of aerosol concentrations for major species [*Hand et al.*, 2011]. Elevation was not explicitly taken into account in these groupings. We used 28 rural IMPROVE regions, two of which included only one site (Death Valley and Virgin Islands). The IMPROVE site density is somewhat higher in the western United States, with 62% of the sites used in this analyses located west of $-100°$ longitude.

[21] We empirically defined 31 regions for the CSN sites based on seasonal distribution of aerosol concentrations and site location. For comparison purposes we grouped sites in regions similar to those defined for the IMPROVE network. Of the 31 regions, eight had only one site per region. Unlike the IMPROVE network, the CSN site density is higher in the eastern United States (80% of CSN sites used in this analysis are located east of $-100°$). When a specific region is referred to in this paper, it refers to an IMPROVE or CSN region, not necessarily a commonly used geographical region. For example, the IMPROVE "Northwest" region refers to a specific group of sites in the northwestern United States, but may differ slightly from the geographic region typically considered as "northwestern United States."

3. Regional and Seasonal Monthly Mean Mass Concentrations

[22] In this section we summarize the spatial patterns in the 2005–2008 monthly and annual mean absolute concentrations and relative contribution of a species' mass to RCFM for AS, AN, POM, LAC, soil, and sea salt. The evaluation of both the absolute and relative concentrations (also referred to as the PM$_{2.5}$ budget) highlights the importance of the behavior of species' mass concentrations relative to each other. For example, the relative contribution from a given species might vary seasonally although its absolute concentrations are steady (or vice versa), solely based on the behavior of other species. We recognize that monthly mean data could be skewed by episodic events, such as the impacts of long-range transport of dust or periods with active fires; we note these events in the discussion.

The monthly mean IMPROVE and CSN regional data are presented as stacked bar charts corresponding to defined regions. Monthly mean concentrations and mass fractions are depicted with the first letter of the month, followed by an "A" for annual mean. Bar charts are grouped into figures corresponding to three sections of the country: northwestern, southwestern, and eastern United States. Notice that the scales on the figures differ for each region in order to present the range in concentrations clearly. We begin each section with an overview to highlight significant findings, followed by more detailed discussion of absolute concentrations, relative concentrations, and seasonality. The range in seasonality for a given region is defined as the ratio of the maximum monthly mean concentration to minimum monthly mean concentration and is a measure of the temporal variability in aerosol concentration over the year.

3.1. Ammonium Sulfate

3.1.1. Overview

[23] Significant results for AS are the following: (1) urban and rural absolute and relative AS concentrations and seasonal trends were generally similar across the United States, although urban concentrations were somewhat higher; (2) AS concentrations were highest in the central eastern United States, with concentrations that were 5–10 times higher than in the West; (3) AS dominated the rural and urban PM$_{2.5}$ mass budget in the East, contributing roughly 40% year-round and up to 60% in summer; contributions were roughly 20% in the West; (4) AS concentrations were generally highest during summer and lowest during winter months, with the range in seasonality near a factor of 2. The exception in seasonality occurred in the northwestern United States, where the maximum AS concentrations occurred during spring.

3.1.2. Absolute Concentration

[24] The central eastern United States corresponded to the highest rural and urban AS concentrations in the United States, with monthly mean concentrations that were double or greater than concentrations in the West. The maximum regional monthly mean concentrations of 9.94 μg m^{-3} and 10.82 μg m^{-3} occurred at the rural Appalachia region in August and urban Washington DC/Philadelphia region in July, respectively. Figures 2a and 2b demonstrate the similar regional patterns of AS at rural and urban regions, respectively, in the East. High sulfur dioxide emissions from coal-fired power plants, combined with meteorological conditions favorable to sulfate formation and regional transport, led to regional-scale impacts that influenced both rural and urban regions alike, although urban concentrations were somewhat higher. Other studies have demonstrated similar AS concentrations for urban and rural sites in the East, with concentrations often dominated by regional transport of AS [e.g., *Chen et al.*, 2002; *Rao et al.*, 2003; *Hansen et al.*, 2003; *Liu et al.*, 2003; *Tanner et al.*, 2004; *Dutkiewicz et al.*, 2004; *Malm et al.*, 2004; *Qin et al.*, 2006; *Lall and Thurston*, 2006; *Fairlie et al.*, 2009; *Chen et al.*, 2010]. Although concentrations were much lower elsewhere in the United States (see Figures 3a and 3b), similar spatial patterns and magnitudes in urban and rural AS concentrations were common.

[25] The Southern California region was one exception. Summertime concentrations in the urban Los Angeles and
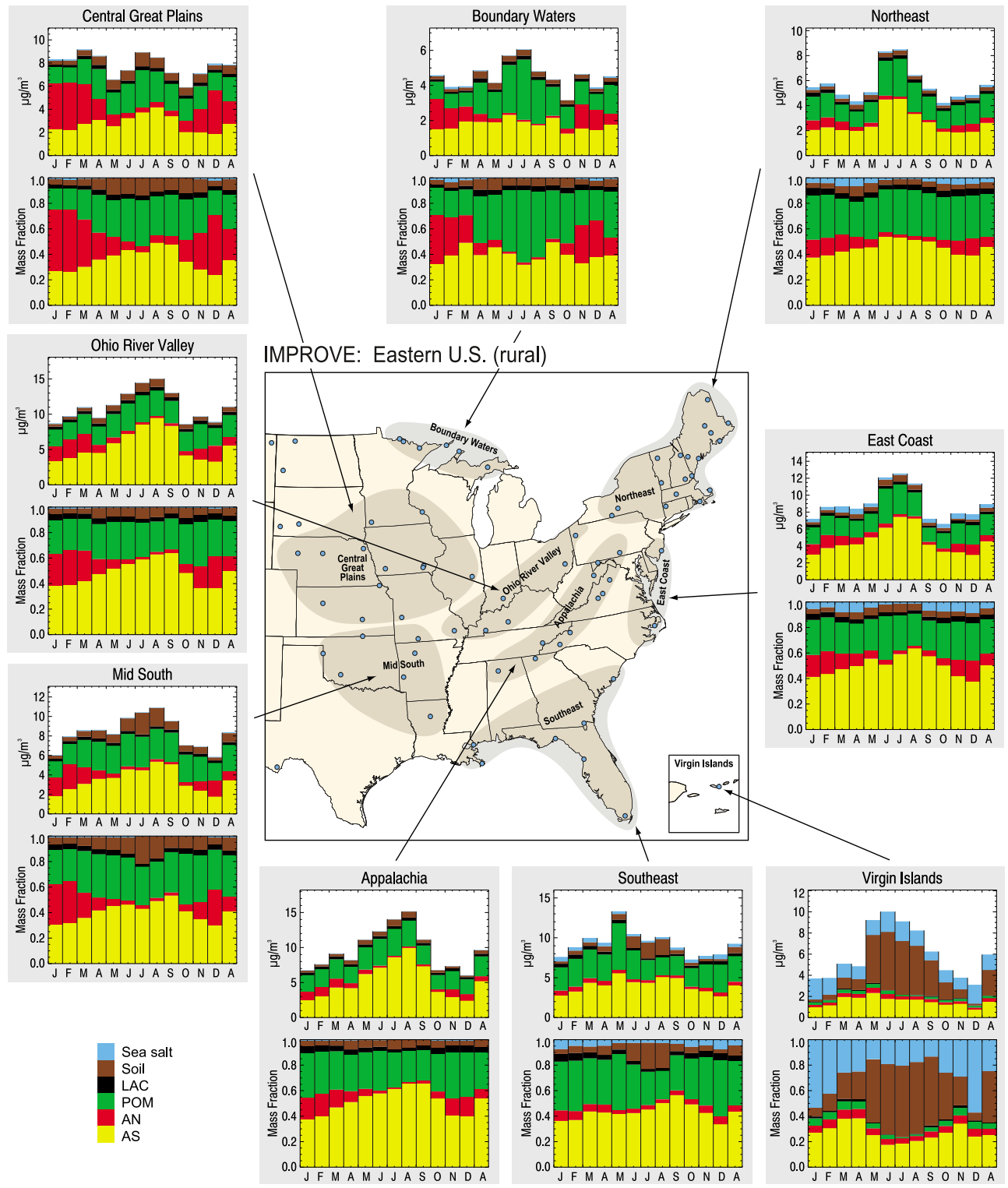
Figure 2a. IMPROVE 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions for the eastern United States, including the Virgin Islands region. The letters on the x axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.
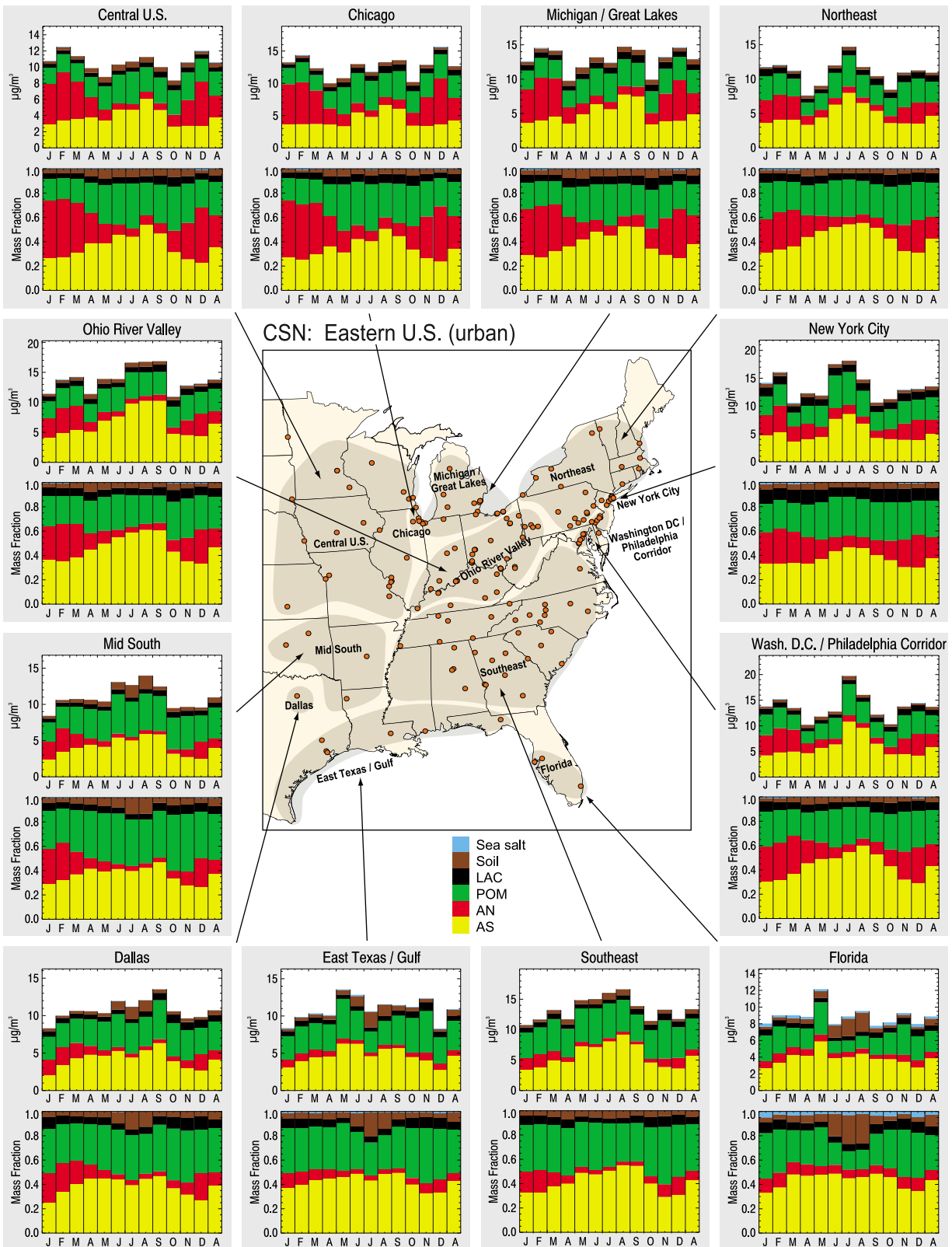
Figure 2b. CSN 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions for the eastern United States. The letters on the x axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.
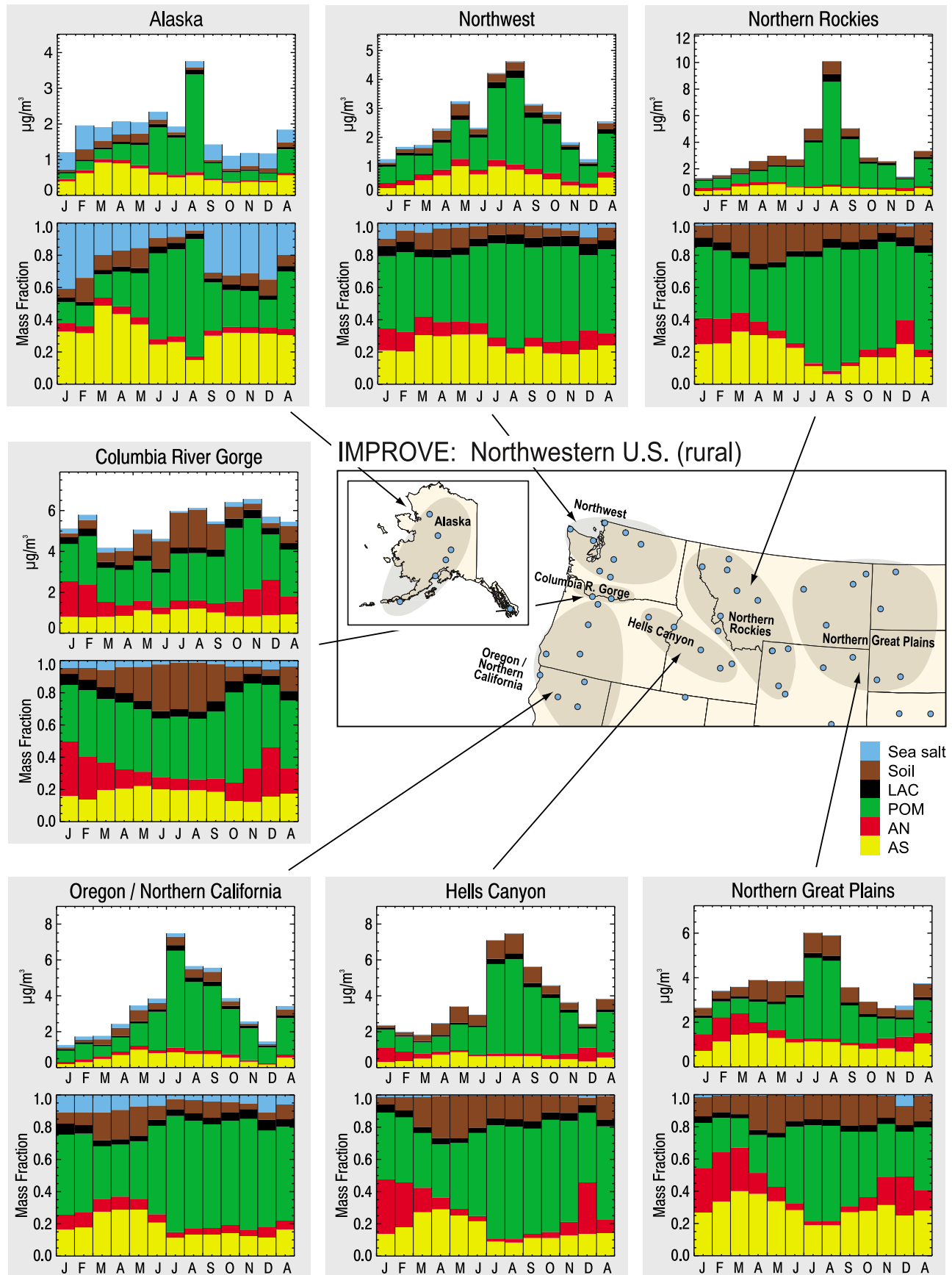
Figure 3a

San Diego regions ($\sim$5 μg m^{-3}) were more than double the concentrations in nearby rural regions (compare Figures 4a and 4b, respectively), perhaps because of their vicinity to shipping ports. However, the urban concentrations in southern California were unusual for the Southwest; most other urban and rural regions corresponded to concentrations of $\sim$2 μg m^{-3} in summer. The urban and rural regional and seasonal similarities of AS in the Southwest suggest regional impacts from sources such as power plants [*Malm et al.*, 2004], although long-range transport of sulfate off the Pacific coast (e.g., shipping emissions [*Xu et al.*, 2006]), Asia [*Park et al.*, 2004], Mexico, and the eastern United States can be important at some regions [e.g., *Gebhart et al.*, 2006]. In addition, stagnation events also can lead to regional impacts [*Chu*, 2004; *Tai et al.*, 2010]. Although AS concentrations are relatively low in the northwestern and southwestern United States, it is an important contributor to visibility degradation at remote regions in the West [e.g., *Malm et al.*, 1996; *Day and Malm*, 2001; *Hand et al.*, 2004] and is the main contributor to visibility degradation in the East [*Hand et al.*, 2011].

3.1.3. Relative Concentration

[26] AS contributions to RCFM were highest at rural and urban regions in the East and contributed roughly 40% year-round and up to 60% to RCFM in summer (see Figures 2a and 2b, respectively). The highest urban mass fraction of 61% occurred in the Ohio River Valley region in August. The role of AS in the PM$_{2.5}$ budget decreased farther West, where contributions of 20% were typical at regions in the northwestern (Figures 3a and 3b) and southwestern United States (Figures 4a and 4b), respectively. The highest regional rural mass fraction of 76% occurred in the Hawaii region in March.

3.1.4. Seasonality

[27] Summer maxima in urban and rural AS concentrations and relative contributions occurred at every area of the country except the northwestern United States, where nearly all of the rural and urban regions exhibited spring maxima in absolute and relative AS concentrations (20–30% of RCFM in spring; see Figures 3a and 3b). This is an intriguing diversion from the usual conceptual model that AS concentrations are higher in summer because of increased photochemical reactions and stagnation events [e.g., *Tai et al.*, 2010; *Chu*, 2004]. The sources of spring sulfate in the northwestern United States are not yet clearly understood but may be due to off-coast transport mentioned earlier. The range in seasonality for absolute and relative concentrations was around a factor of 2 for most urban and rural regions in the United States, with the exception of the regions along the West Coast and in the Southwest, where a higher range in seasonality (ratios of 5–6) occurred (see Figures 4a and 4b). The largest range in seasonality was observed in the urban Alaska region, where maximum concentrations in winter were 15 times higher than the minimum summer concentrations, in contrast to the rural Alaska region, where the

ratio of the maximum to minimum monthly mean AS concentration was 2.6.

3.2. Ammonium Nitrate

3.2.1. Overview

[28] Significant findings corresponding to AN are the following: (1) urban AN concentrations were 4–5 times higher than some nearby rural regions; (2) urban and rural AN concentrations were highest in California and the Midwest; (3) AN contributions to the PM$_{2.5}$ budget reached 50% at urban and rural regions in the Midwest and West and decreased to 10–15% at regions far from major sources; (4) AN concentrations peaked during winter at nearly every urban and rural region and demonstrated a high degree of seasonality.

3.2.2. Absolute Concentration

[29] Concentrations of AN were highest in the Midwest and California for both urban and rural regions. The rural maximum monthly mean AN mass concentration was 4.08 μg m^{-3} at the Central Great Plains region in February (Figure 2a), compared to higher concentrations at nearby urban regions, such as the December monthly mean concentration of 7.11 μg m^{-3} in Chicago (Figure 2b). Concentrations dropped considerably (1–2 μg m^{-3}) at regions farther northeast and in the southeastern United States. Large sources of ammonia from agriculture in the central United States combined with meteorological conditions in winter (low temperature and high relative humidity) provide favorable chemical and thermodynamic conditions for the formation of particulate AN through the equilibrium reaction of gaseous ammonia and nitric acid. Although both ammonia and nitric acid are required for the formation of AN, it is the abundance of ammonia in the Midwest that leads to high concentrations in this region compared to other locations in the United States [*Pitchford et al.*, 2009]. Conditions favorable to the particulate phase of this reversible reaction lead to longer lifetimes and regional extent of AN. In fact, regional-scale nitrate events are common in the Midwest [e.g., *Fischer and Talbot*, 2005; *Lee and Hopke*, 2006; *Pitchford et al.*, 2009; *Chen et al.*, 2010]. In urban regions high nitrogen oxide emissions can lead to significantly higher AN concentrations compared to rural regions, such as those in the Michigan/Great Lakes and Chicago regions (Figure 2b). In the eastern United States, *Frank* [2006] reported high wintertime nitrate concentrations for several urban sites. *Katzman et al.* [2010] found nitrate was responsible for exceedances in PM concentrations in the Great Lakes region during winter months in 2005.

[30] Urban AN concentrations in Utah and California were considerably higher than urban concentrations the Midwest (see Figure 4b). The maximum monthly mean urban concentration (14.09 μg m^{-3}) occurred in November at the Sacramento/San Joaquin Valley region, an area with both available ammonia and nitrogen oxide emissions. *Chow et al.* [2006] described the interaction between emissions

Figure 3a. IMPROVE 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions for the northwestern United States, including the Alaska region. The letters on the x axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.
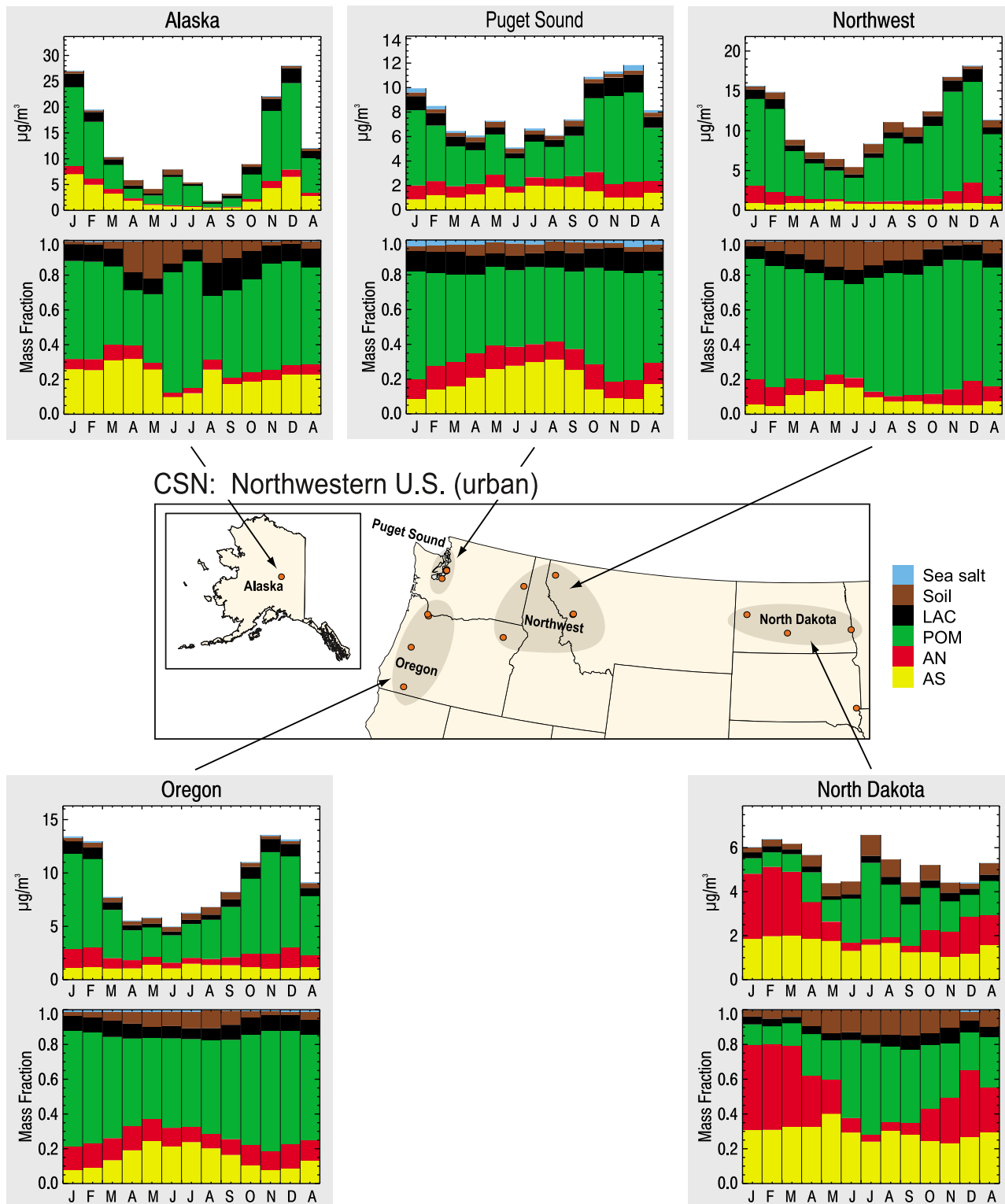
Figure 3b. CSN 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions for the northwestern United States, including the Alaska region. The letters on the x axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.
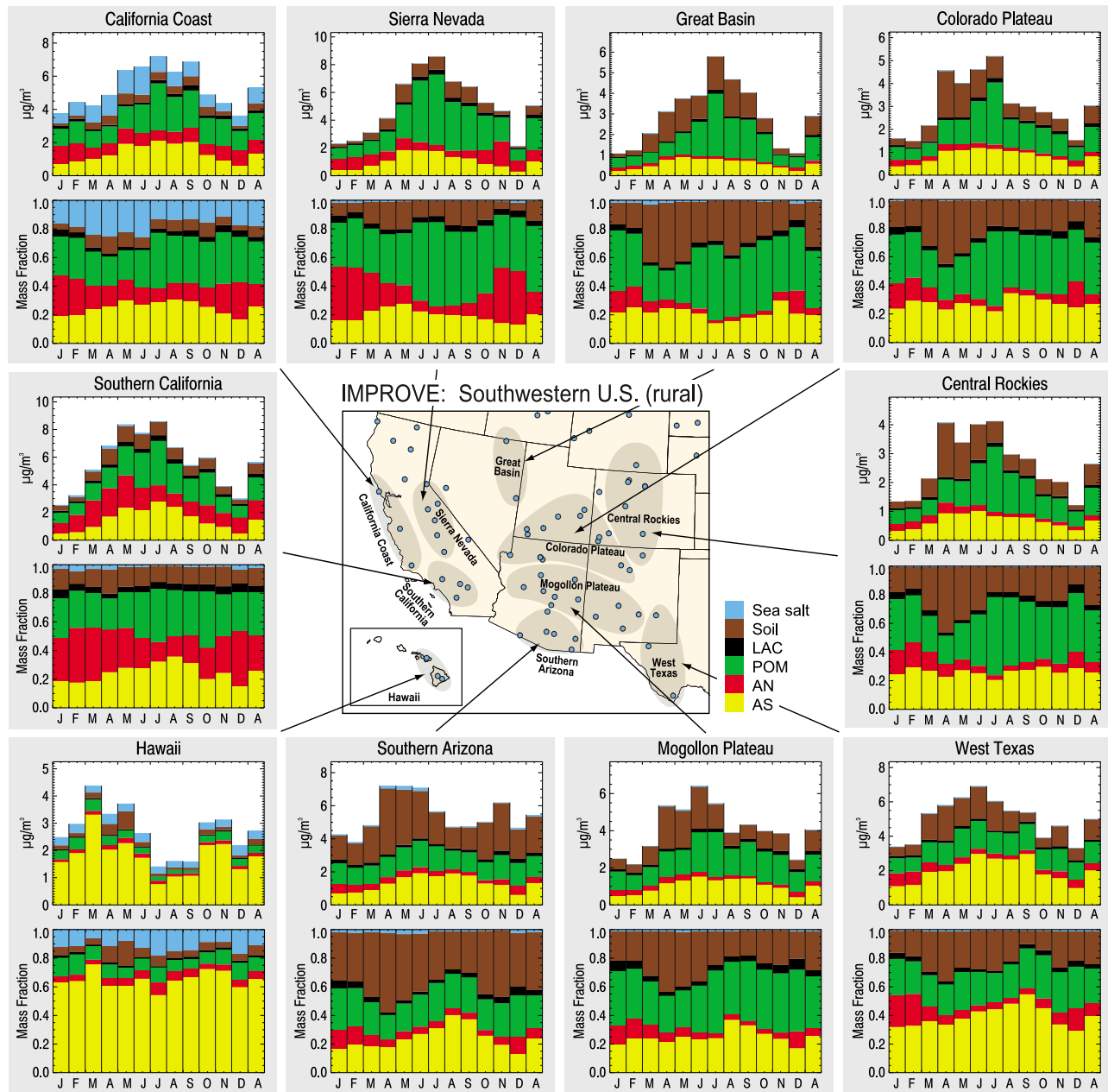
Figure 4a. IMPROVE 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions on bottom for the southwestern United States, including the Hawaii region. The letters on the *x* axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.

and meteorology that led to the formation and transport of regional nitrate events in winter in the San Joaquin Valley, California. These interactions resulted in urban AN concentrations that were 4–5 times higher than rural concentrations, ranging from approximately 1–2 μg m^{-3} and 5–10 μg m^{-3} for most rural and urban regions in California, respectively (see Figures 4a and 4b, respectively). *Rao et al.* [2003] also computed much higher urban nitrate concentrations compared to rural values in Salt Lake City, Utah, and

Fresno, California, for a 1 year period in 2001–2002. Results from 2003 data in similar locations were consistent [*EPA*, 2004b]. Some of the higher urban AN concentrations could be due to differences in elevation. *Chow et al.* [2006] reported that AN concentrations decreased rapidly as a function of altitude for sites in California during the Central California Regional PM$_{10}$/PM$_{2.5}$ Air Quality Study (CRPAQS) from 1999 to 2001 because of ventilation and transport of pollutants.
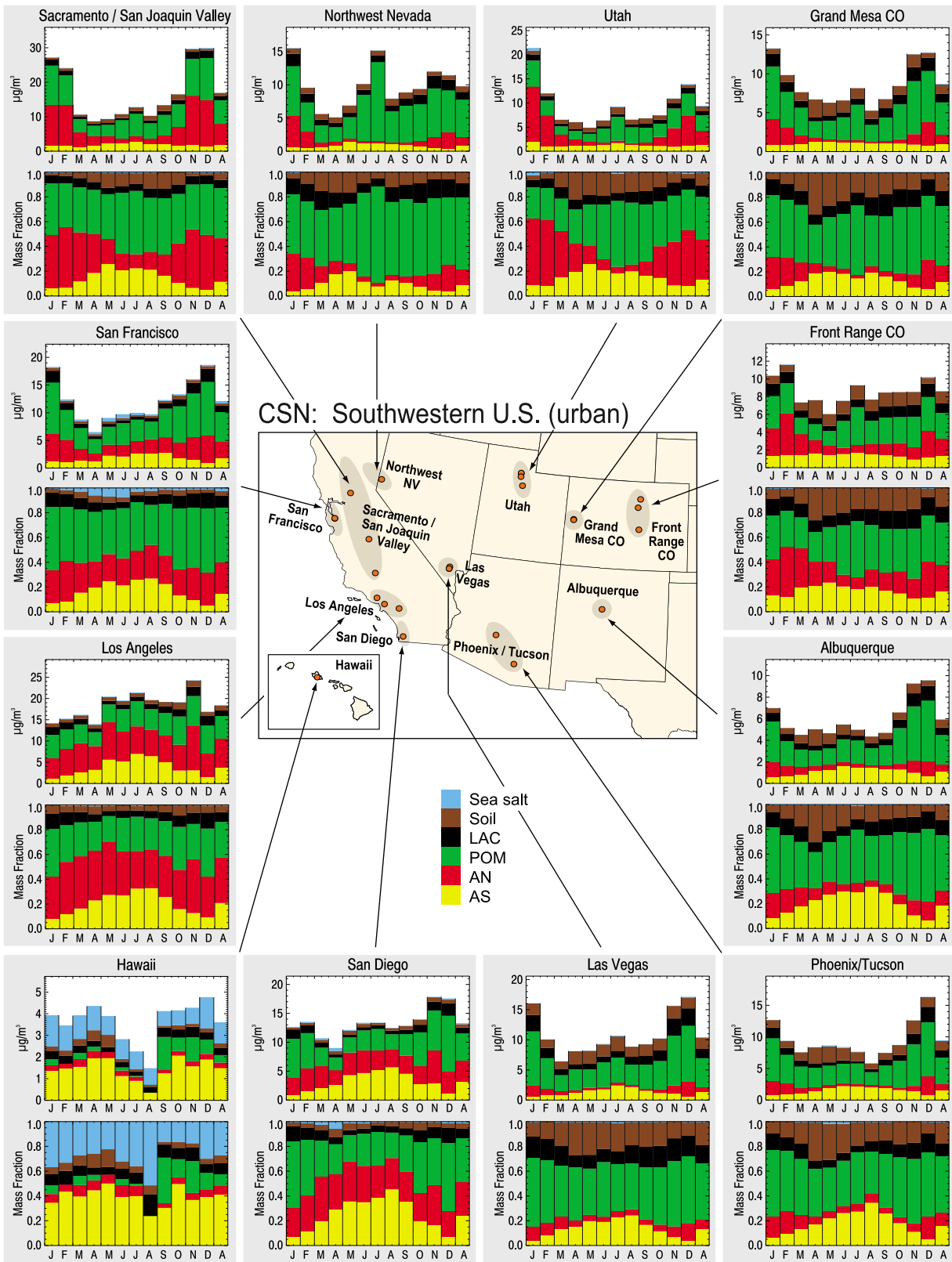
Figure 4b. CSN 2005–2008 regional monthly mean PM$_{2.5}$ mass concentrations (μg m^{-3}) surrounded by PM$_{2.5}$ reconstructed fine mass fractions on bottom for the southwestern United States, including the Hawaii region. The letters on the *x* axis correspond to the month, and "A" corresponds to "annual" mean. Ammonium sulfate (AS) in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light-absorbing carbon (LAC) in black, soil in brown, and sea salt in blue. The shaded area corresponds to the regions that comprise the sites used in the analysis, shown as dots.

3.2.3. Relative Concentration

[31] Contributions of AN reached up to 50% of RCFM at the rural Central Great Plains regions (Figure 2a) and the urban Central United States, Chicago, and Michigan/Great Lakes regions (Figure 2b). However, a steep gradient in the relative concentration was observed farther east and south, where both urban and rural contributions of 10–20% were common. Contributions were likewise low (10–20%) at most of the regions in the northwestern United States, with the exception of the rural Columbia River Gorge, Hells Canyon, and Northern Great Plains regions (Figure 3a). The urban North Dakota region demonstrated patterns in AN contributions that were unusual relative to other urban regions in that area (see Figure 3b) and more in line with contributions observed at urban regions in the Midwest. AN played an increasingly important role in the $PM_{2.5}$ budget, moving west into California and along the West Coast. The maximum urban mass fraction of 53% occurred in the Utah region in January. Contributions of 20–40% were typical at urban regions such as the Los Angeles, San Diego, San Francisco, and Sacramento/San Joaquin Valley regions (Figure 4b). The sources leading to relatively high contributions appeared to also influence nearby rural regions such as the California Coast, Sierra Nevada, and southern California regions, all of which had increased contributions relative to other rural regions in the southwestern United States (Figure 4a).

3.2.4. Seasonality

[32] Wintertime peaks in AN concentrations occurred for nearly every urban and rural region in the United States. This seasonal phenomenon is due to the favorable formation of particulate AN under cold temperature and high relative humidity conditions. In addition, sulfate concentrations were low in winter, allowing available ammonia to react with nitric acid beyond what was required to neutralize sulfate. Because of these favorable conditions, contributions of AN to RCFM dominated in winter at many locations. A unique feature of AN compared to other species is its wide range in seasonality, which varied considerably across the United States because of the sensitivity of AN to available precursors and conditions that drive its chemical and thermodynamic reactions. These sensitivities were reflected in the seasonal and geographic patterns in AN concentrations across the United States. For example, urban regions in the West, such as in Utah, Colorado, North Dakota, and Nevada, experienced a much greater range in seasonality compared to nearby rural regions, with maximum winter monthly mean concentrations that were 10–15 times the minimum summer concentrations and over 30 times higher in the Utah region. In contrast, the seasonal ratios in nearby rural regions, where AN concentrations were much lower, ranged between a factor of 2 and 3, probably because, although the seasonal temperature range was similar, emissions of ammonia and nitrogen oxides were lower, resulting in lower levels of AN year-round. The range in seasonality in the central United States was similar for both urban and rural regions, with maximum mean concentrations that were 10 times the minimum concentrations (the rural Boundary Waters region had the highest ratio at 20). The wide range between maximum and minimum mean concentrations demonstrated the effects of seasonal temperature variations on AN concentrations in

locations with abundant emissions of precursors, such as in the Midwest. In comparison, the range in seasonality for regions along the East Coast was closer to a factor of 5.

3.3. Particulate Organic Matter

3.3.1. Overview

[33] Significant findings for POM include the following: (1) urban concentrations were 2–5 times higher than rural concentrations; (2) urban and rural concentrations were highest in the southeastern and northwestern United States; (3) urban and rural POM contributions to RCFM ranged from 20% to 30% in the East to over 50% in the West; (4) POM concentrations demonstrated low seasonality in the eastern United States with summer maxima occurring for both urban and rural regions. In contrast, in the western United States POM was associated with highly seasonal concentrations associated with urban winter maxima and rural summer maxima.

3.3.2. Absolute Concentration

[34] Urban POM concentrations were higher than rural concentrations in general, and both were highest in the southeastern and northwestern United States. The maximum rural regional monthly mean POM concentration was 7.72 μg m^{-3} in the Northern Rockies region in August (Figure 3a), compared to the highest urban POM concentration (16.74 μg m^{-3}) in the Alaska region in December (Figure 3b). Urban POM concentrations in the northwestern United States were 2–3 times higher than rural regions (typically around 10 μg m^{-3}, compared to rural regions with concentrations ∼3 μg m^{-3} and greater; see Figures 3a and 3b). *Rao et al.* [2003] and *EPA* [2004b] also found that total carbon in urban areas was much higher than rural areas on an annual mean basis for 2001–2002 and 2003, respectively. Relatively high rural POM concentrations in the northwestern United States were most likely due to biomass burning emissions. Others have shown that wildfire activity is as a major contributor to rural POM concentrations, especially in the western and northwestern United States in summer [e.g., *Spracklen et al.*, 2007; *Jaffe et al.*, 2008]. The impact of smoke appeared to lessen at rural regions in the Southwest (1–2 μg m^{-3}), although rural regions to the north of the Southwest area (e.g., Great Basin, Colorado Plateau, Sierra Nevada, and Central Rocky Mountain regions) experienced impacts from the wildfire activity similar to regions in the northwestern United States during summer (Figure 4a). Smoke from biomass burning has been shown to have a large impact on visibility degradation in the West, such as occurred in the Sierra Nevada during the summer of 2002 [*McMeeking et al.*, 2006]. As in the northwestern United States, southwestern urban POM concentrations were significantly higher (2–3 times) than rural POM concentrations (Figure 4b). More will be said on urban POM sources in the West in the section describing seasonality.

[35] POM concentrations in the southeastern United States were relatively high in both rural and urban regions (see Figures 2a and 2b, respectively), although urban POM concentrations were nearly double the concentrations for some nearby rural regions. POM concentrations were similar to AS concentrations during nonsummer months (2–3 μg m^{-3} for rural and 3–4 μg m^{-3} for urban). Similar regional patterns in urban and rural POM concentrations suggested

sources leading to regional influences that were most likely associated with biomass smoke [*Zhang et al.*, 2010] and biogenic emissions, as many radiocarbon studies have shown that half or more of the carbon concentrations measured at eastern urban and rural sites were associated with modern carbon [e.g., *Tanner et al.*, 2004; *Bench et al.*, 2007; *Ke et al.*, 2007, *Schichtel et al.*, 2008; *Blanchard et al.*, 2011], and secondary organic aerosols could account for more than 30% of the fossil and contemporary carbon [*Schichtel et al.*, 2008]. An analysis of remote sensing data by *Goldstein et al.* [2009] suggested that the spatial and temporal distribution of aerosol optical depth over the southeastern United States is consistent with natural biogenic volatile organic compound emissions. Episodic impacts from large biomass burning events can significantly impact the POM concentrations in the East, such as during the spring of 2007 when several hundred fires that burned in Georgia and Florida contributed significantly to carbon concentrations in both rural and urban sites in the southeastern U. S. regions [*Christopher et al.*, 2009]. While in general urban POM concentrations were higher than rural concentrations, recall that the same carbon multiplier (1.8) was used to compute POM for both networks; therefore the discrepancy in concentrations may be overestimated as lower carbon multipliers are likely more appropriate for urban POM [*Turpin and Lim*, 2001; *Malm et al.*, 2011].

3.3.3. Relative Concentration

[36] POM was the dominant contributor to RCFM in both rural and urban regions in the northwestern United States (40–60% and 60–75% for rural and urban regions; see Figures 3a and 3b, respectively). The highest POM relative contribution occurred at the Northern Rockies region in August (76% of RCFM), most likely because of biomass burning emissions. *Holden et al.* [2011] also reported high (49%) primary smoke relative contributions to contemporary carbon at Rocky Mountain National Park in summer of 2005. The maximum urban POM mass fraction occurred in the Northwest Nevada region in July (78%; see Figure 4b). This region was somewhat unusual for urban regions in the Southwest, where POM contributions were typically 40% or less; it is possible that the Northwest Nevada region experienced some impact from biomass burning emissions given its proximity to fire locations. Rural and urban POM contributions were 20–40% in the Southwest and in the East and followed AS in importance to the PM$_{2.5}$ budget in summer; POM and AS contributions were similar during nonsummer months in the East (Figures 2a and 2b, respectively).

3.3.4. Seasonality

[37] One of the more interesting characteristics of POM was its seasonality. Urban and rural POM concentrations peaked during the same season at some regions of the United States, while at other areas, they peaked during opposite seasons. These differences provided some indication of sources. For example, in the eastern United States the rural and urban POM concentrations demonstrated similar seasonality, with summer maxima and winter/spring minima for both, consistent with biogenic emissions mentioned earlier. In general urban and rural POM contributions were fairly seasonally flat in the East, with seasonal ranges near a factor of 2. The seasonal behavior of urban and rural POM in the West was markedly different than in the East and indicated

very different sources and atmospheric processing of POM. The summer maxima in rural POM concentrations was consistent with impacts from biomass smoke and biogenic emissions, as mentioned earlier. However, the winter urban maxima observed at most regions were probably due to additional local sources (recall that urban POM concentrations were 2–3 times higher than rural concentrations) or due in part to meteorological conditions. An evaluation of CSN data by *Chu* [2004] suggested wintertime organics were associated with primary sources. *Chow et al.* [2006] reported that the differences in POM seasonality at rural and urban sites in the San Joaquin Valley in California were most likely due to winter meteorology and residential wood combustion. Differences in urban and rural seasonality were observed in both the northwestern and southwestern United States, although urban regions in the southwestern United States were typically associated with spring, rather than summer, minima (see Figure 4b). Only one urban region in the southwestern United States corresponded to summer maxima (northwest Nevada) and was most likely influenced by biomass burning because of its location. Rural regions experienced a much larger seasonal range in monthly mean POM concentrations in general, with maximum summer concentrations that were typically 5–10 times greater than minimum winter and spring concentrations. In contrast, urban winter maxima were only 3–4 times higher than summer minima.

3.4. Light-Absorbing Carbon

3.4.1. Overview

[38] The important findings for LAC include the following: (1) urban LAC concentrations were significantly higher than rural values, with the differences being the greatest for any species; (2) LAC concentrations were quite low relative to other species; (3) contributions to RCFM were on the order of a few percent for rural regions, and slightly higher at urban regions; (4) the seasonality in rural and urban LAC was similar in the East but with maxima occurring in different seasons than POM, while in the West urban and rural seasonality in LAC followed that of POM.

3.4.2. Absolute Concentration

[39] The differences between urban and rural LAC concentrations were the largest of any species, although the absolute concentrations were relatively low. Rural LAC concentrations of 0.1–0.3 μg m^{-3} were common around the United States (see Figures 2a, 3a and 4a). The highest regional monthly mean LAC mass concentration was 0.56 μg m^{-3} in the rural Northern Rockies region in August, compared to the highest urban concentration of 2.91 μg m^{-3} in the Alaska region in December. Urban LAC concentrations were considerably higher in general (1–2 μg m^{-3}). The maximum urban concentration in Alaska was nearly a factor of 30 times higher than the rural Alaska summer peak in LAC of $\sim$0.1 μg m^{-3}. This difference suggested different sources of LAC, with urban sources generally more likely associated with diesel emissions and residential heating in winter and rural sources associated with biomass burning emissions. Meteorological conditions, such as lower mixing heights, wind speeds, and temperature inversions, can also contribute. While urban LAC concentrations were significantly higher at many regions, low concentrations at nearby

rural regions suggested that the extent of its geographical influence was low.

3.4.3. Relative Concentration

[40] It is not surprising that the relatively low LAC concentrations contributed only a few percent to RCFM. Rural contributions of 3–5% to RCFM were common around the United States (see Figures 2a, 3a and 4a), with the highest rural value of 8% in the Mogollon Plateau region in December. Urban LAC contributions were somewhat higher (5–15%; see Figures 2b, 3b and 4b), with the highest urban contribution at 19% in the Alaska region in August. While the contribution of LAC to RCFM is relatively low at most regions in the United States, its contribution to visibility impairment is non-negligible (~10% at rural regions and 20% or higher at urban regions) because of its high extinction efficiency relative to other species [*Hand et al.*, 2011].

3.4.4. Seasonality

[41] Patterns in the seasonality of LAC suggested that it differed from POM in important ways. For example, like POM, both rural and urban LAC concentrations had similar seasonality in the East, but unlike the summer maxima observed for POM, LAC maxima occurred in fall and winter, because of sources such as residential heating that were important at both urban and rural regions (see Figures 2a and 2b). However, in contrast, in the West the LAC seasonality followed that of POM, with summer maxima for most rural regions and winter maxima for urban regions, suggesting biomass burning sources for rural regions and local sources like residential heating and transportation in urban regions. The range in seasonality for LAC was fairly low and similar for both urban and rural regions. Maximum concentrations were roughly double minimum concentrations in the East, 4–5 times higher in the West, and even higher for some rural regions that experienced episodic impacts of biomass burning.

3.5. PM$_{2.5}$ Soil

3.5.1. Overview

[42] The significant findings for PM$_{2.5}$ soil concentrations include the following: (1) soil concentrations were highest in the southwestern United States for both rural and urban regions, although relative biases made comparisons between the two networks difficult; (2) contributions of soil to RCFM reached 40% at urban and rural regions in the southwestern United States in spring and were lower elsewhere; (3) concentrations and contributions of soil to the PM$_{2.5}$ budget were highest in spring and summer, depending on region.

3.5.2. Absolute Concentration

[43] The dominant characteristic of fine soil is its importance to the rural PM$_{2.5}$ budget in the southwestern United States, with concentrations that were 2–4 times higher than elsewhere in the country. Rural soil concentrations typically ranged from 1 to 2 μg m^{-3} but reached up to 4 μg m^{-3} at some regions in the Southwest (e.g., southern Arizona; see Figure 4a). Urban soil concentrations were also higher in the southwestern United States, with the maximum urban soil concentration of 2.55 μg m^{-3} at Phoenix/Tucson in April (Figure 4b). Recall from section 2.4 that a relative bias existed between collocated IMPROVE and CSN soil concentrations (−32.1%), with considerably higher IMPROVE concentrations for reasons undetermined. Comparisons of urban and rural absolute magnitudes should be approached

with caution, but comparisons of regional and seasonal patterns are still meaningful. Sources of soil in the Southwest were most likely associated with local and upwind transport [*Kavouras et al.*, 2009], from long-range sources such as Mexico [*Rivera Rivera et al.*, 2009] and Asia [*Kavouras et al.*, 2009]. Rural and urban soil concentrations in the northwestern United States were typically less than 1 μg m^{-3} (see Figures 3a and 3b, respectively).

[44] The spatial trends in soil concentrations in the eastern United States displayed a very interesting pattern that reflected the impact of long-range transport from North Africa. The maximum soil concentration at all rural regions was 5.54 μg m^{-3} in the Virgin Islands in June. High soil concentrations during summer at the Virgin Islands site have been associated with long-range dust transport from North Africa [*Perry et al.*, 1997; *Prospero*, 1999; *Hand et al.*, 2004]. The impact of this transport reached regions in the southern United States such as the rural regions of the Southeast, Midsouth, and Appalachia and the urban regions of the Midsouth, Florida, Dallas, and East Texas/Gulf, where concentrations reached 2 μg m^{-3}. This spatial gradient was fairly steep, with relatively low soil concentrations characteristic of most other eastern rural (0.2–0.5 μg m^{-3}) and urban regions (0.3–0.8 μg m^{-3}; see Figures 2a and 2b, respectively).

3.5.3. Relative Concentration

[45] Contributions of soil to RCFM reached 40% at southwestern U.S. regions such as the Southern Arizona and Death Valley regions (Figure 4a). Urban contributions of soil to RCFM in the Southwest were higher than other areas of the United States but lower (~30%) than many rural regions in the same vicinity (Figure 4b). The maximum urban contribution of 33% occurred at the Grand Mesa CO region in April. Contributions of soil to RCFM were lower elsewhere in the country (<20%), although the impact from transport of North African dust in the eastern United States did impact the PM$_{2.5}$ budget at several southern regions where contributions reached up to 20%, with the rural maximum of 56% at the Virgin Islands site. In contrast, other urban and rural eastern regions experienced contributions that were less than 10% (see Figures 2a and 2b).

3.5.4. Seasonality

[46] The seasonal patterns in soil concentrations were driven mainly by emission sources and transport patterns because of its primary origin. Spring maxima occurred at urban and rural regions along the West Coast and in Colorado, Arizona, and New Mexico (see Figures 4a and 4b), compared to summer maxima for regions in between, such as the rural Great Basin, Hells Canyon, the Northern Rockies, and the Columbia River Gorge (see Figures 3a), and the urban regions of Utah, Las Vegas, Oregon, and the Northwest (Figure 3b). The urban exception was the fall maxima associated with regions in southern California. Several studies have suggested that contributions of Asian dust to fine soil concentrations in the United States can be significant episodically during spring months, affecting aerosol concentrations and mineralogy across the United States [e.g., *Husar et al.*, 2001; *Prospero et al.*, 2002; *VanCuren and Cahill*, 2002; *Jaffe et al.*, 2003; *VanCuren*, 2003; *Wells et al.*, 2007]. However, studies by *Kavouras et al.* [2007, 2009] using 2001–2003 IMPROVE data in the western United States suggested that local or upwind

transport of dust sources dominates Asian dust for the worst dust days in the northwestern United States. In the southwestern United States, bimodal seasonal peaks in soil concentrations in the rural southern Arizona region coincided with environmental conditions that may be responsible for higher soil concentrations during spring and winter from sources such as Mexico [*Rivera Rivera et al.*, 2009]. Long-range transport was very influential on seasonality in the eastern United States, as was already discussed. Summer maxima were common for urban and rural southeastern sites and transitioned to spring maxima in the northeastern United States. While the seasonality in urban and rural regions was generally similar across the United States, the range in seasonality was much larger for rural regions, especially in the West, with maximum mean concentrations that were 10 times greater or more than minimum concentrations, compared to 3 times at western urban regions.

3.6. PM$_{2.5}$ Sea Salt

3.6.1. Overview

[47] Important findings regarding PM$_{2.5}$ sea salt include the following: (1) coastal regions were the only locations with non-negligible sea salt concentrations. Meaningful comparisons between urban and rural concentrations were difficult because of significant relative biases between data from the two networks; (2) contributions of sea salt to RCFM were 10–20% or higher at some coastal regions and low elsewhere (<5%); (3) sea salt concentrations were highest in winter and spring.

3.6.2. Absolute Concentration

[48] The only urban and rural regions in the United States with non-negligible sea salt concentrations corresponded to rural coastal regions. In the eastern United States, coastal sea salt concentrations were comparable to soil concentrations (0.2–0.5 μg m^{-3}) and reached up to 1.98 μg m^{-3} at the Virgin Islands in January (Figure 2a). Recall, like soil concentrations, the relative bias between IMPROVE and CSN sea salt concentrations, with higher IMPROVE concentrations at collocated sites (relative bias of −72.8%). However, sea salt concentrations at urban coastal regions were similar to rural regions in the eastern United States (0.2–0.3 μg m^{-3} in the Florida region; see Figure 2b). Sea salt concentrations at coastal regions in the northwestern United States corresponded to somewhat lower concentrations than in the East (approximately 0.2 μg m^{-3} or less; see Figure 3a). Interestingly, the Northern Great Plains region showed non-negligible concentrations of sea salt to RCFM in December (0.20 μg m^{-3}) compared to the annual mean of 0.03 μg m^{-3}, possibly associated with a long-range transport event [*White et al.*, 2010]. The highest rural concentrations of sea salt occurred at the California Coast (~1 μg m^{-3}) and Hawaii regions (~2 μg m^{-3}; see Figure 4a). Urban coastal regions such as San Francisco and San Diego had the highest urban concentrations in the southwestern United States (0.1–0.5 μg m^{-3}; see Figure 4b). While sea salt concentrations were low relative to other species, they can be an important contributor to visibility degradation, especially at remote coastal locations, because of their hygroscopic nature [e.g., *Quinn et al.*, 2001; *Lowenthal and Kumar*, 2006; *Hand et al.*, 2011].

3.6.3. Relative Concentration

[49] Not surprisingly, based on its relatively low concentrations, sea salt contributions to RCFM were generally negligible (<1%); however, at coastal urban and rural regions contributions reached 5–10%. Higher contributions occurred at the rural Virgin Islands region (57%; see Figure 2a) and the rural Alaska region (40–50%; see Figure 3a) because of coastal vicinity. Urban contributions at Hawaii reached 52%, much higher than at the rural Hawaii region (10–20%; see Figures 3b and 3a, respectively).

3.6.4. Seasonality

[50] Although sea salt concentrations were low throughout the United States, they were typically highest during winter and spring months for noncoastal, rural, and urban regions, possibly because of the application of road salt. Coastal regions demonstrated relatively low seasonality in sea salt concentrations.

4. Summary

[51] We examined the 2005–2008 monthly and annual mean aerosol concentrations for the remote/rural IMPROVE network and the urban/suburban CSN. Specifically, we investigated the seasonal and spatial variability for PM$_{2.5}$ ammonium sulfate (AS), ammonium nitrate (AN), particulate organic matter (POM), light-absorbing carbon (LAC), soil and sea salt. One of the major purposes of this paper was to compare and contrast rural and urban aerosol concentrations at nearby regions. The major findings include the following:

4.1. Ammonium Sulfate

[52] Urban and rural absolute and relative AS concentrations and seasonal trends were generally similar across the United States, although urban concentrations were somewhat higher. AS concentrations were highest in the central eastern United States, with concentrations that were 5–10 times higher than in the West. AS dominated the rural and urban PM$_{2.5}$ mass budget in the East, contributing roughly 40% year-round and up to 60% in summer; contributions decreased to roughly 20% in the West. With the exception of spring maxima observed in the northwestern United States, summer and winter concentrations were typically the highest and lowest, respectively, with the range in seasonality near 2.

4.2. Ammonium Nitrate

[53] Urban AN concentrations were 4–5 times higher than nearby rural regions and both regions corresponded to the highest concentrations in the Midwest and California. AN contributions to the PM$_{2.5}$ budget reached 50% at urban and rural regions in the Midwest and West and decreased to 10–15% at regions far from major sources. AN concentrations peaked in winter at nearly every urban and rural region and demonstrated a high degree of seasonality.

4.3. Particulate Organic Matter

[54] Urban concentrations were 2–5 times higher relative to rural concentrations and both were highest in the southeastern and northwestern United States. Urban and rural POM contributions to RCFM ranged from 20% to 30% in the East to over 50% in the West. POM concentrations were highest in the summer in the eastern United States for both urban and rural regions but were associated with opposite

seasonality in the West, with urban winter maxima and rural summer maxima.

4.4. Light-Absorbing Carbon

[55] Urban LAC concentrations were significantly higher than rural concentrations, with the differences being the greatest for any species. LAC concentrations were quite low relative to other species, and contributions to RCFM were a few percent at rural regions and slightly higher at urban regions. The seasonality in rural and urban LAC was similar in the East but with maxima occurring in different seasons than POM. In the West, LAC seasonality followed the urban and rural POM seasonality, with urban winter maxima and rural summer maxima.

4.5. Soil

[56] Soil concentrations were highest in the southwestern United States for both rural and urban regions, although relative biases between the data made comparisons between the two networks difficult. Contributions of soil to RCFM reached 40% at urban and rural regions in the southwestern United States in spring and were lower elsewhere. Concentrations and contributions of soil to the $PM_{2.5}$ budget were highest in spring and summer, depending on region.

4.6. Sea Salt

[57] Non-negligible sea salt concentrations occurred only at urban and rural coastal regions and meaningful comparisons between networks were difficult because of significant relative biases. Contributions of sea salt to RCFM were 10–20% or higher at some coastal regions and low elsewhere (<5%). Sea salt concentrations were highest in winter and spring.

[58] The seasonal and spatial patterns in rural and urban aerosols emphasized the importance for understanding the variability in the concentrations of major aerosol species and their contributions to the $PM_{2.5}$ budget. The close agreement between data from collocated IMPROVE and CSN sites suggested that for most species (except soil and sea salt), comparisons between urban and rural concentrations at similar or adjacent regions provided meaningful insights into aerosol sources and transport and the role of urban influences on nearby rural regions. Uncertainties certainly could affect these comparisons. The potentially most significant uncertainty unaccounted for in this analysis is associated with elevation differences. We did not correct for elevation differences, as *Rao et al.* [2003] showed that elevation corrections were small for nearby urban and rural sites, assuming the same aerosol mass was sampled. However, elevation corrections do not account for the possibility that different air masses might be sampled at higher altitude sites relative to sites at lower elevations because of boundary layer heights or ventilation processes [*Chow et al.*, 2006]. Nevertheless, the integration of data from over 300 sites from the IMPROVE network and the CSN provided extensive spatial coverage of surface aerosol concentrations and allows for further investigation into issues such as urban excess.

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