Technical Memorandum #7

Review of Speciation Trends Network and IMPROVE Chemically Speciated Data

Prepared by Northeast States for
Coordinated Air Use Management
(NESCAUM)

For the MANE-VU Regional Planning
Organization

March 21, 2003

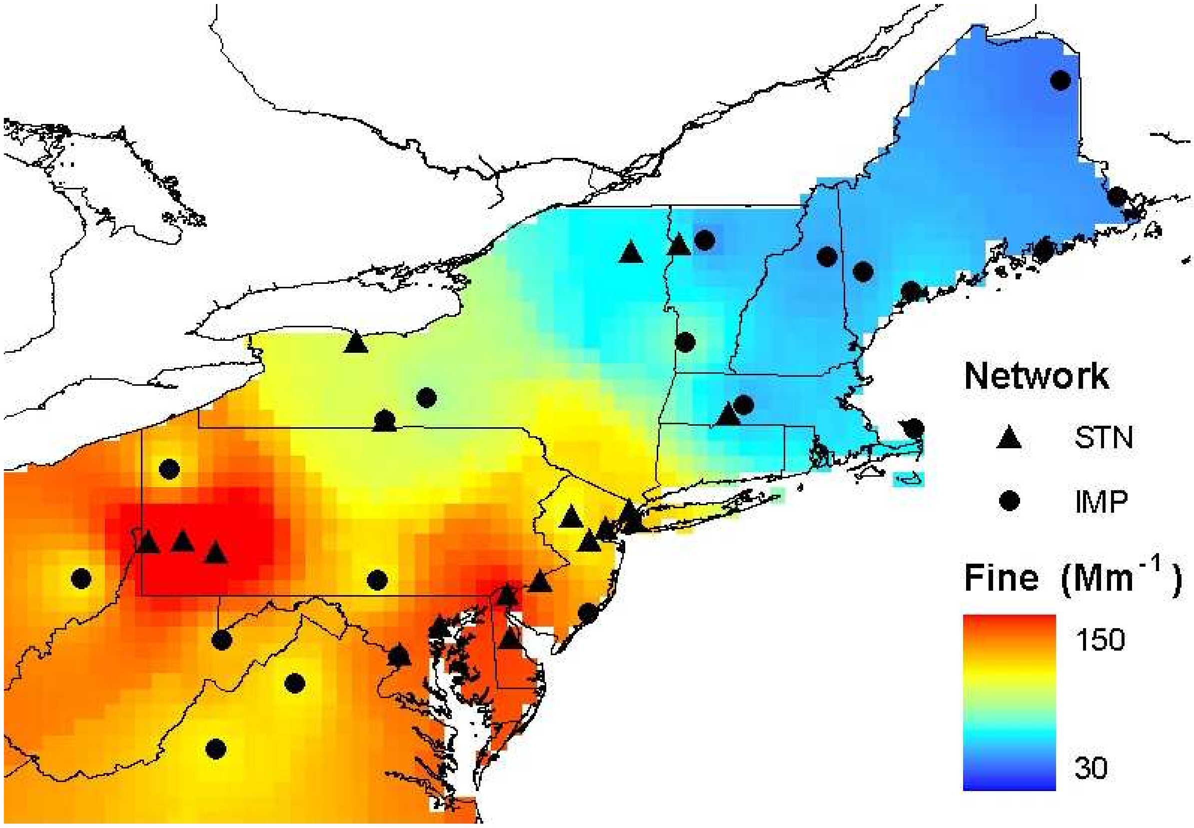

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Table of Contents

Executive Summary

In order to better support the USEPA's 1999 Regional Haze Rule, the Interagency Monitoring of Protected Visual Environments (IMPROVE) network was significantly expanded, extending spatial coverage of aerosol characterization throughout the MANE-VU domain. Simultaneous with the expansion of the IMPROVE network, USEPA developed the Speciation Trends Network (STN), in part to assist states in understanding fine particulate composition in urban areas and to illuminate rural/urban differences in fine particulate composition.

This memorandum examines region-wide speciated fine particulate measurements and their corresponding aerosol extinction using data from these two networks. Preliminary attempts were made to eliminate the substantial differences between the networks, yielding composite maps of aerosol extinction in the MANE-VU region. This investigation has shown the need for an in-depth assessment of the approaches utilized in combining these two data sets and the underlying data from each network if they are to be included in state or tribal SIPs. Nonetheless, the results from the spatially dense network combination are consistent with the historical findings from the IMPROVE program. Considerable detail is gained by including the urban data afforded by the STN sites. This information will play a key role as states prepare to comply with federal fine particulate and regional haze standards.

The pilot data comparison presented here between the two networks demonstrates the significant differences between the measurements obtained by IMPROVE and STN. Although the results are promising, they should be viewed with caution due to their preliminary nature. An ongoing study of collocated STN and IMPROVE monitors will provide more complete information than was available for this analysis. Additional investigations, beyond this, are needed to fully understand and subsequently resolve the differences.

I. Introduction

An interagency coalition composed of the National Park Service (NPS), the Fish and Wildlife Service (FWS), the Bureau of Land Management (BLM), the Forest Service (FS) and the United States Environmental Protection Agency (USEPA) established the Interagency Monitoring of Protected Visual Environments (IMPROVE) program in response to the 1977 amendments to the Clean Air Act. This monitoring network has collected speciated aerosol and related visibility data in or near Federal Class 1 areas in the United States since 1988. In order to better support the USEPA's 1999 Regional Haze Rule (hereafter, "the Haze Rule"), the network was significantly expanded, extending spatial coverage of aerosol characterization. Simultaneous with the IMPROVE expansion, USEPA developed the Speciation Trends Network (STN), in part to assist states in understanding fine particulate composition in urban areas and to illuminate rural/urban differences in fine particulate composition. Both of these monitoring programs are useful sources of information to states as they prepare implementation plans for fine particulate matter ($PM_{2.5}$) and regional haze; however, important differences between the networks' sampling and analysis protocol necessitate great care in comparing results between the two.

Twelve months of data covering the period June 2001 through May 2002 encompassing a total 22 IMPROVE and 26 STN sites located throughout the MANE-VU region are reviewed in this memo (Figure 1). This document compares urban and rural visibility, when possible, and attempts to explain important differences that must be accounted for when conducting such comparisons. A comprehensive picture of fine particle composition is developed using both data sets that increases our knowledge of the spatial influence of urban areas on ambient fine particulate. The network protocols are summarized and their differences highlighted below. Composite illustrations of visibility extinction from ionic and carbonaceous species are provided. Additional analyses focused on seasonal and annual visibility using the expanded IMPROVE network yield the broadest depiction of regional visual air quality available to date.

A. Interagency Monitoring of Protected Visual Environments (IMPROVE)

The IMPROVE measurement network is a cooperative effort between the federal land management agencies, USEPA and state air agencies. Under this program, a total of 110 aerosol samplers have been deployed with an additional fifty-two samplers that are operated using the IMPROVE protocol. (IMPROVE, 2002 newsletter Volume 11/Number 3)[1]. The main objectives of the program as listed in the third major summary report (Malm 2000) include (1) establishing current visibility and aerosol conditions in Class 1 areas, (2) identifying the chemical make up and the major contributing sources to observed aerosol, (3) documenting long-term trends in visibility, and (4) providing representative monitoring of regional haze in Class 1 areas. These goals are met, in part, by sampling air for particulate matter.

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[1] Six of the IMPROVE monitors have operated in the MANE-VU region in excess of eight years, with twelve additional monitors deployed since 2001.

Table 1. Formulae and assumptions used with IMPROVE sampling measurements to derive reconstructed particle light extinction (from Sisler and Malm 2000).

Species	Formula	Assumptions
SULFATE	4.125[S]	All elemental S is from sulfate. All sulfate is from ammonium sulfate.
NITRATE	1.29[NO3]	Denuder efficiency is close to 100%. All nitrate is from ammonium nitrate.
LAC (Light absorbing carbon)	[EC1] + [EC2] + [EC3] – [OP]	All high temperature carbon is elemental.
OMC (Organic mass from carbon)	1.4{[OC1] + [OC2] + [OC3] + [OC4] + [OP]}	Average organic molecule is 71% carbon.
SOIL (Fine Soil)	2.2[AL] + 2.49[SI] + 1.63[CA] + 2.42[FE] + 1.94[TI]	[Soil K] = 0.6[Fe]. FeO and Fe_2O are equally abundant. A factor of 1.16 is used for MgO, Na_2O, H_2O, CO_2.
RCFM (Reconstructed fine mass)	[SULFATE] + [NITRATE] + [LAC] + [OMC] + [SOIL]	Represents dry ambient fine aerosol mass for continental sites.
CM (Coarse Mass)	[MT] – [MF]	Consists only of insoluble soil particles.

[S]= Elemental sulfur   [NO3]= Nitrate
[AL]= Aluminum   [SI]= Silicon
[CA]= Calcium   [TI]= Titanium
[FE]= Iron   [MT]= Total mass (PM_{10})
[MF]= Fine mass ($PM_{2.5}$)
[OC#] = Detailed TOR organic species with bins (# = 1,2,3,4)
[OP] = Pyrolized Organic Carbon evolved in the first EC fraction
[EC#] = Detailed elemental carbon species measured by thermal optical reflectance (TOR) with three bins (# = 1,2,3)

The IMPROVE aerosol sampler has four channels for particle collection. The A and D channels collect $PM_{2.5}$ and PM_{10} on teflon filters and are weighed gravimetrically to yield the mass of fine and coarse particulate. The deposits on the fine fraction filter are also analyzed by three methods to determine the elemental particle composition: (1) Proton Elastic Scattering Analysis (PESA) for H, (2) Particle Induced X-ray Emission (PIXE) for elements Na to Mn, and (3) X-Ray

Fluorescence (XRF) for heavier elements Fe through Pb (IMPROVE Standard Operating Procedures (SOP) 301 and 326, 1997)[2].

The B channel uses a 25 or 37 mm nylon filter for sample collection, after the sample stream has passed through an annular sodium carbonate denuder to remove acid gases (IMPROVE QAPP). These filters are analyzed for major anions by Ion Chromatography (IC). The filters are extracted with a dilute sodium carbonate/sodium bicarbonate buffer and then analyzed for chloride (Cl^-), nitrite (NO_2^-), nitrate (NO_3^-), and sulfate (SO_4^{2-}). At a limited number of IMPROVE sites, including Shenandoah National Park and Dolly Sods Wilderness area, analysis includes Ammonium ion (NH_4^+). Deionized water is used for filter extractions of both anions and cations. (RTI anion SOP, 2000)

Finally, quartz filters are used in the C channel and analyzed for elemental (EC) and organic (OC) carbon. The demarcation between these two carbon components is defined based on the analytical technique and analysis protocol. The IMPROVE program uses Thermal Optical Reflectance (TOR) and splits EC and OC as the point during analysis at which the filter reflectance reaches its original value.[3]

The measurements obtained from the four filters may be combined and used to "reconstruct" the 24-hour integrated visibility conditions represented by the samples. Briefly, reconstructed mass and visibility are derived by using the measured concentrations of nitrate ion for ammonium nitrate, the element sulfur for ammonium sulfate, the EC and OC components for carbon, the difference in PM_{10} and $PM_{2.5}$ for coarse particulate and the elements aluminum, calcium, iron, silicon and titanium for soil. This derivation is based on a number of assumptions, which are listed in Table 1 and documented elsewhere (Malm et al., 1994, Malm, et al. 2000, NESCAUM 2001, 2002).

B. Speciation Trends Network (STN)

The National Ambient Air Quality Standards (NAAQS) for $PM_{2.5}$ requires the collection of fine particulate for speciated analysis. The rule specifies that at a minimum, measurements of metals, certain ions and carbon are to be conducted as part of a National Air Monitoring Stations (NAMS) network of fifty sites (62 FR 38763). These sites operate under uniform conditions nationally and will be used to establish trends in fine particle constituents and also serve as models for a more extensive network of speciation samplers. As of January 2002, 54 trends sites were in place with approximately 215 supplemental sites established by states. This broader network will provide vital information to states as they devise plans to address particulate pollution. Examples of data usage include tracking the progress of control programs, delineating annual and seasonal spatial characterization of aerosols; informing decisions in emission control strategy development and integrating with data collected from IMPROVE (USEPA 2001b).

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[2] Samples taken after December 1, 2001 use XRF with Cu tube excitation for the low Z elements, replacing the PIXE instrument. The original XRF instrument uses Mo excitation and is still used for the high Z elements. Elemental detection overlap between PIXE/Cu-XRF and Mo-XRF allows for quality assurance of the data.

[3] IMPROVE fractions are evolved as follows: Under inert He atmosphere, OC1 (ambient to 120 °C) OC2 (120 to 250 °C), OC3 (250 to 450 °C), OC4 (450 to 550 °C), add 2% oxygen, EC1 (550 °C in oxidizing atmosphere), EC2 (550 to 700 °C) and EC3 (700 to 800 °C)

Figure 1 Site Location Map for IMPROVE and STN

The measurements made by the STN samplers are similar to those of IMPROVE. There are three channels for collection of fine particulate using a teflon, nylon and quartz filter in respective channels. Unlike IMPROVE, PM_{10} is not collected as part of this program. In addition, there are four different sampler types used in the network: Andersen Reference Ambient Air Sampler-RAAS, MET ONE Spiral Aerosol Speciation Sampler-SASS, URG Mass Aerosol Speciation Sampler-MASS, and Rupprecht & Patashnick (R&P) 2300, each with its own sample flow rate and collection volume. Finally, STN filter samples are shipped cold[4] whereas IMPROVE filters are not.

The teflon filter is gravimetrically weighed for fine particulate mass. Elemental analysis is also performed on this filter by Energy Dispersive X-ray Fluorescence (EDXRF). The STN does not use PIXE, nor does it analyze for H. As a result, STN lacks the beneficial quality assurance of the IMPROVE approach. For example, IMPROVE compares organic carbon as determined by hydrogen versus the direct organic carbon as measured on the quartz filter.

The second filter is nylon and collects ionic species. Like IMPROVE, STN employs IC for detection, but the denuder is magnesium oxide for RAAS and SASS instruments. The target species are nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium (NH_4^+), sodium (Na^+) and potassium (K^+). Unlike IMPROVE, the extraction procedure uses deionized water that removes both positive (cations) and negative (anions) ions. The cations listed are not routinely determined by IMPROVE.

The particulate collected on the quartz filter is analyzed for both elemental and organic carbon. The STN uses a modified diesel particulate NIOSH method as its procedure. This method, like IMPROVE's, is based on thermal evolution of carbon from the filter. However, STN follows different temperature ramping[5] and employs Thermal Optical Transmittance (TOT) instead of Reflectance (TOR) to monitor charring on the filter and determine the split between OC and EC. Differences between IMPROVE and STN carbon measurements will be explored in more detail in the following section.

II. Intercomparison of IMPROVE and STN

Comparison of results from both networks would appear straightforward. Target species are primarily the same, and both collection and detection technology are similar. However, examination of data demonstrates the hidden complexity of this task. USEPA is currently investigating results from six pairs (one STN and one IMPROVE) of collocated samplers, with two in the east (Washington, DC and Dolly Sods), two in the northwest (Seattle Beacon Hill and Mt. Rainier NP), and two in the southwest (Phoenix and Tonto National Monument). The comparison study began in October 2001 and will continue through at least July 2003. New York State operates another pair of collocated samplers at Pinnacle State Park.

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[4] STN protocol requires filters to be shipped at a temperature less than 4 °C.

[5] STN fractions are evolved under an inert helium atmosphere from ambient to 310 °C, and ramps to 480, 615, 900 °C, the oxygen is added with temperature back to 600 °C then ramped to 675, 750, 825 and 920 °C.

This section explores and discusses some of the data from the New York samplers covering the period from mid-April 2001 to the end of May 2002. Of the possible 138 sample dates, 125 had available data from both sites. To increase the number of comparison points, two other sampler pairs have been analyzed: Washington, D.C. IMPROVE (elevation 16 m)/McMillan Reservoir STN (elevation 50 m) and Proctor Maple R.F. IMPROVE (elevation 403 m)/Burlington STN (elevation 81 m). The Washington pair includes data from June 2001 through mid-February 2002, with 62 of 85 possible sample pairs available. For Burlington, 105 of the possible 121 sample pairs were valid, covering June 2001 through the end of May 2002. These two site pairs do not have collocated monitors and are used only as supplemental data. The Washington DC pair and the Vermont pair are approximately 10 and 40 km apart respectively. Therefore, very local sources may influence the sites, causing differences that are not due to sampling or analytical methodologies.

A. Direct Comparison of Results

Significant differences exist in the reported results from all three analyzed sets of data. Certain constituents compare favorably, including $PM_{2.5}$ mass and the ionic species sulfate and nitrate. Others, like several of the elements that are used to derive the “soil” component and the carbon fractions agree less frequently or not at all.

A.1 MASS

The fine mass results are well correlated (coefficient of determination r^2= 0.95 NY, 0.96 DC, 0.85 VT, Figure 2). Measured fine mass from the NY samplers was also compared to an FRM sampler with favorable results. This demonstrates that the networks are recording similar fine particulate mass. However, the trends are not consistent for the three pairs, with the STN at New York yielding approximately ten percent less mass than the IMPROVE monitor, whereas the Washington, D.C. and Vermont STN predict four and twenty percent greater mass than does IMPROVE. In all cases there is a positive bias in STN measured mass as represented by a positive intercept (Table 2).

Firm conclusions cannot be drawn from this data. Although the IMPROVE samplers are all the same, the STN sampler in NY is an R & P 2300, Washington DC uses a RAAS and Burlington operates a SASS. The collocated site in NY suggests that IMPROVE underpredicts total mass relative to STN R & P 2300 samplers. One cannot determine if the different results among the sites are due to differences in sample flow rates and volumes, the fact that the Washington, D.C. and Vermont pairs are not collocated, merely nearby, or if some other factor plays a role.

A.2 SOIL ELEMENTS

Soil elements include aluminum (Al), calcium (Ca), Iron (Fe), silicon (Si) and Titanium (Ti). Measured concentrations of these five elements are multiplied by factors to represent the mass of their common oxides in crustal materials. Other multipliers are factored in (Table 1). Soil does not contribute significantly to regional haze due to its low concentrations and inefficient scattering properties. Since sulfur is also measured on the same filter and by the same techniques, comparisons among all of these elements are important. Lessons may be learned by assessing their observed similarities and differences.

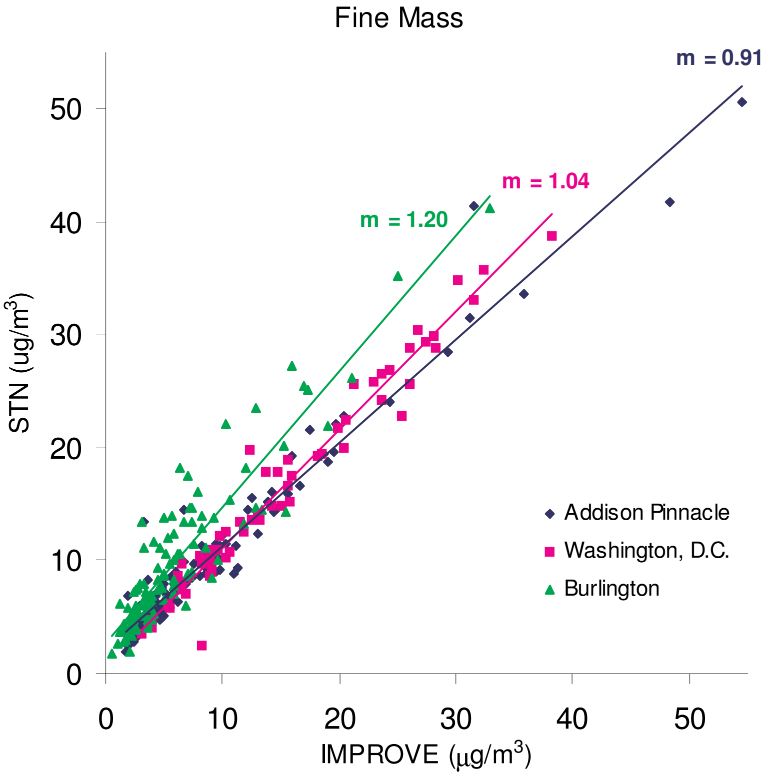

Figure 2. STN and IMPROVE fine mass comparison from Addison Pinnacle State Park in NY, Washington D.C. IMPROVE/McMillan Reservoir STN, and Proctor Maple RF IMPROVE/Burlington STN in VT.

Considerable scatter is seen for the soil composite and the individual elemental components for all three sites (Figures 3.1 and 3.2). The results in New York are the most promising. For soil in New York, the STN predicts about half the mass that IMPROVE does but has the highest coefficient of determination. The other two sites have higher slopes and intercepts. Both Washington and Burlington pairs are impacted differently by mobile sources, which are often associated with generation of dust. The following discussion focuses on the New York results since they seem to be the most consistent of the three sites. This is likely in part due to the collocation of the monitors. To understand what drives the differences seen, each element is reviewed individually.

Iron shows the best agreement of the five elements. There are no zero values or non-detects for this element, at least no cases where one network recorded a value while the other reported below detection levels. The data encompasses five seasons (Spring and Fall of 2001, Winter 2001-2 and Spring and Summer 2002). Six of the iron values recorded at the New York site in the spring 2001 are substantially higher than all of the other values. Since many of the other soil elements have extreme values on the same dates, it is possible some local soil disturbance contributed to these results. These six points have a significant influence on the slope of the linear fit. When spring 2001 is removed from the data set, the slope increases from 0.72 to 0.90, demonstrating the weakness of using a limited data set for this type of analysis.

Iron is the one element for which IMPROVE used the same detection over the entire data record. The other four elements were detected using PIXE through December 2001 and after that date, Cu-XRF (see footnote 2). However, no clear difference is seen analyzing the data before and after this change.

The calcium results are second only to iron with respect to agreement between the networks. Using all available points, the calcium results are comparable to those of iron. IMPROVE generally measures about thirty-five percent higher mass for this element on average, although the correlation is high and the intercept near zero. Comparing the results when spring 2001 Ca is removed shows little difference between the full and partial data sets, unlike iron.

Silicon displays more scatter than iron or calcium. There are some instances where STN detects Si and IMPROVE does not. This serves to increase the intercept. Results are progressively worse for titanium and aluminum, in large part due to the greater number of non-detects by either network.

The above soil comparisons used all available data, even if some elements on a particular date were below detection. Non-detects were treated as zero, rather than substituting half the detection limit in all cases. It was assumed that soil mass was small on those days. In order to determine if the observed results were sensitive to the inclusion of non-detects and lowest measured values, additional tests were performed. Removal of the non-detects generally moved the intercept closer to the origin but did not seem to have an appreciable effect on the slope or correlation for comparisons of individual elements. These comparisons also show no significant improvement when data points within twenty percent of the lowest recorded value were removed from the data set.[6] These tests imply that neither detection limits nor low values are responsible for the poor relationships discovered. Table 2 details the slope, intercept and coefficient of determination for all of the graphs produced for this data comparison.

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[6] Some of these results may be explained by limitations in the detection technique. For example, as in chromatographic methods, overlap exists between the detected fluorescent energy of some elements. Aluminum is routinely a merged or shoulder peak, resulting in unreliable concentration calculations. An uncertainty analysis was not performed to assess the impact of individual elemental uncertainty upon overall soil results. However, the soil is dominated by contributions from silicon and iron, so the low/highly variable Al values should not have an undue impact on soil results.

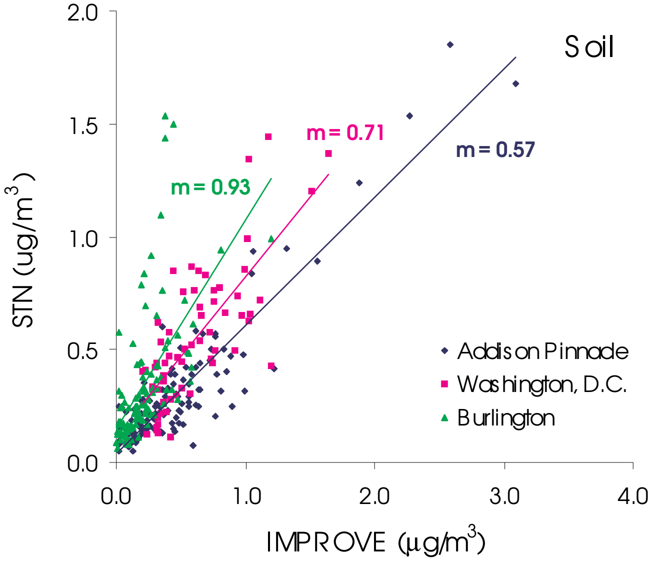
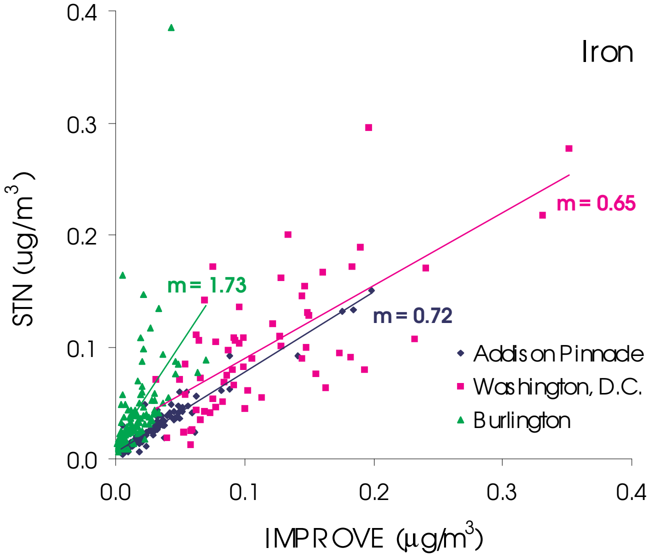


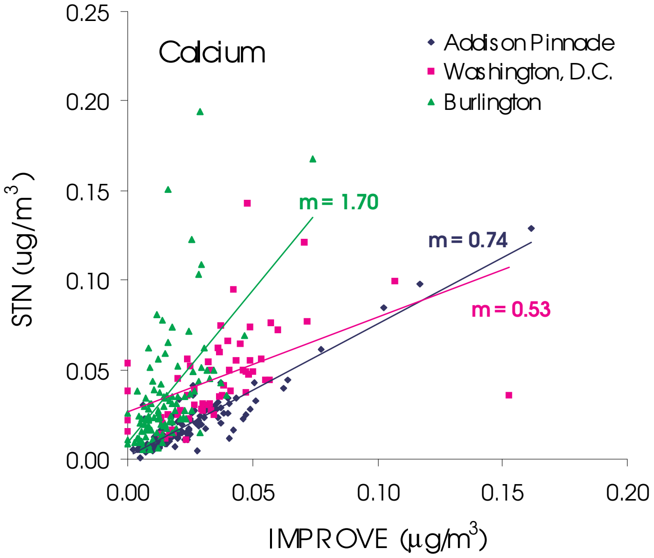
Figure 3.1 Elemental comparison of STN and IMPROVE from Addison Pinnacle State Park in NY, Washington D.C. IMPROVE/McMillan Reservoir STN, and Proctor Maple RF IMPROVE/Burlington STN in VT.

(a) Soil

(b) Elemental Iron

(c) Elemental Calcium





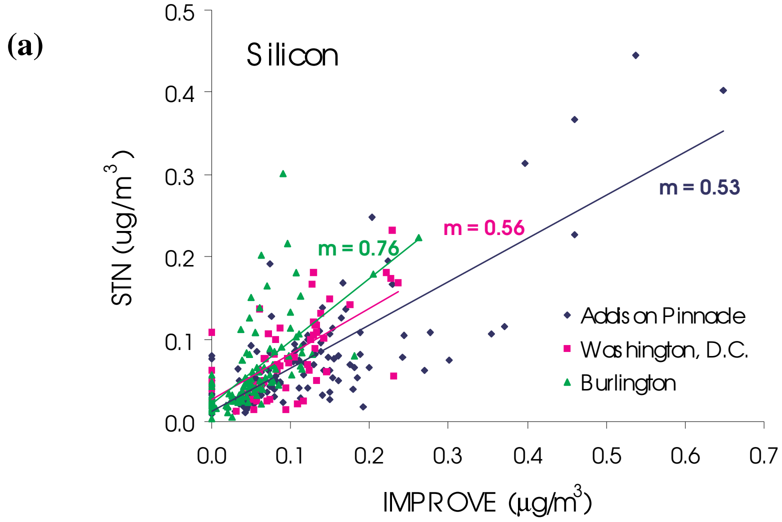
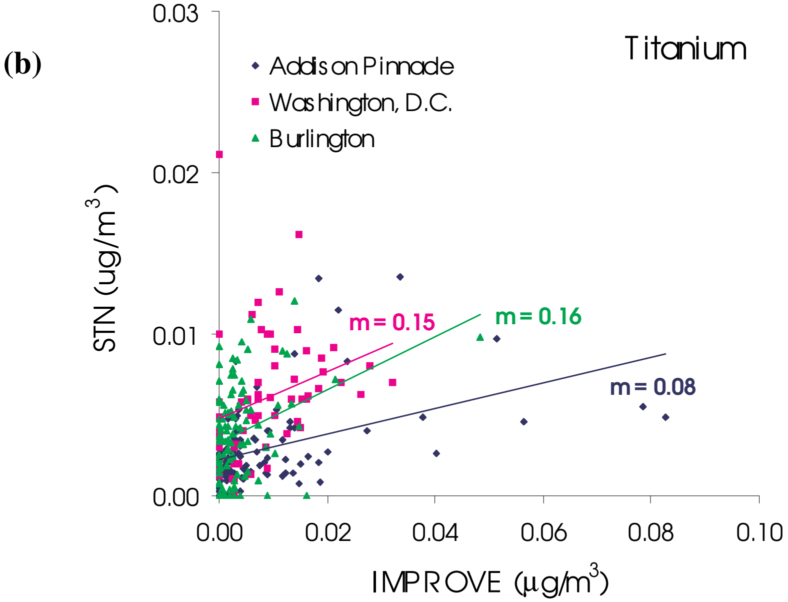


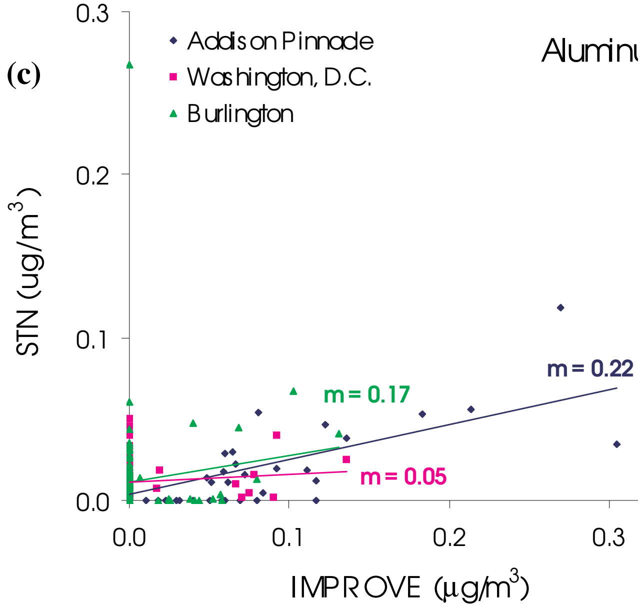
Figure 3.2 **Elemental comparison of STN and IMPR from Addison Pinnacle State Park in NY, Washingto D.C. IMPROVE/McMillan Reservoir STN, and Proc Maple RF IMPROVE/Burlington STN in VT.**

(a) Elemental Silicon

(b) Elemental Titanium

(c) Elemental Aluminum





A.3 IONS

Two measured ions common to both IMPROVE and STN, sulfate and nitrate, show the best agreement of all measured visibility relevant species. Sulfate compares most favorably, although the ionic measurement is not used for the regional haze program. Instead, elemental sulfur concentrations are used to determine extinction due to ammonium sulfate.

The plot of IMPROVE sulfate versus STN sulfate displays the agreement between the networks (Figure 4a). Both of the New York and Washington, D.C. pairs have slopes of unity, while the Vermont slope is 1.07, indicating a slight positive bias for the Burlington STN site. The intercepts are all close to zero with coefficients of determination all above 0.93. One data point representing the maximum sulfate measured in Washington, D.C. over the twelve months, was excluded from the linear fit, as it was far outside the range of other values for the site.

The elemental sulfur measurements hold greater importance for regional haze, since extinction calculations rely on them. The simple quality assurance test contrasting three times sulfur mass with sulfate mass provides confidence in both the teflon and nylon filter measurements, as the relationship holds for both networks. Despite this, the sulfur relationship is not as strong as that observed for sulfate. At all three sites STN measures, on average, higher sulfur than does IMPROVE, but the correlation remains high and the intercept is close to zero (Figure 4b). Sulfur compares much more favorably than the other soil elements. This may be due to its abundance and the wide range of observed concentrations.

Nitrate results also show high correlations and low intercepts for New York and Washington (Figure 4c). The Vermont plot does not share this close agreement, yielding greater scatter, with the STN values generally higher by an average of seventy percent. The other two pairs have STN with roughly fifteen percent greater mass on average than the corresponding IMPROVE value. Two points were excluded from the linear fit of the New York data. For these points, the measured STN values were one to two orders of magnitude greater than the corresponding IMPROVE values, far outside the factor of two range for all other points. In addition, the corresponding sulfate ion ratios were above the 95th percentile. This implies a potential problem existed with either the collection or analysis of these filters for one or both of the networks.

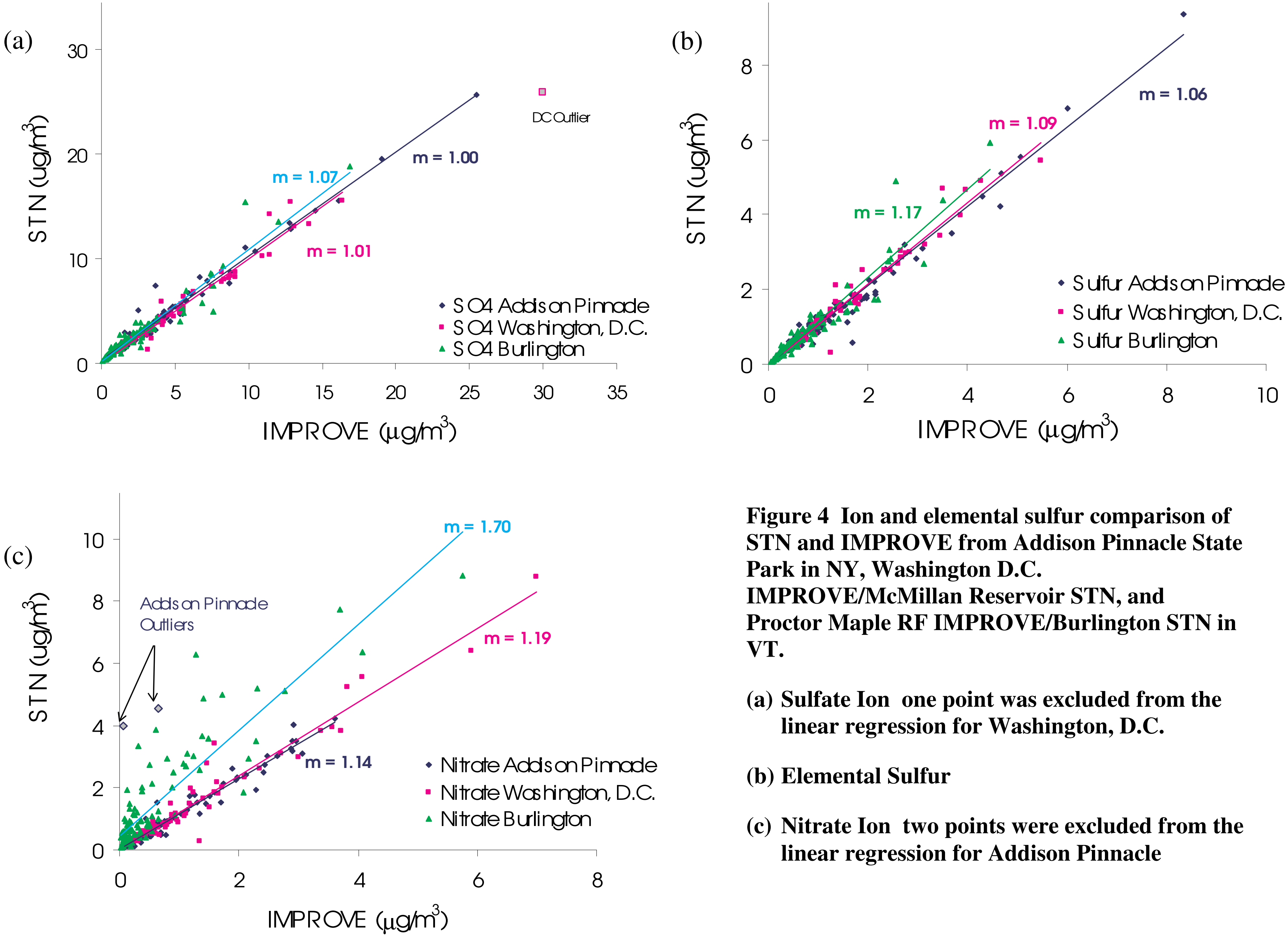


Figure 4 Ion and elemental sulfur comparison of STN and IMPROVE from Addison Pinnacle State Park in NY, Washington D.C. IMPROVE/McMillan Reservoir STN, and Proctor Maple RF IMPROVE/Burlington STN in VT.

(a) Sulfate Ion one point was excluded from the linear regression for Washington, D.C.

(b) Elemental Sulfur

(c) Nitrate Ion two points were excluded from the linear regression for Addison Pinnacle

Table 2 Linear least squares best fit parameters for the STN/IMPROVE comparison curves. These correspond to scatterplots in Figures 2 and 3.1; 3.2; 4, 5, 6 and 7 a, b, c.

Specie	Site	Slope (m)	Intercept (b)	Coefficient of Determination (r^2)
TC	Addison Pinnacle	0.98	0.03	0.50
TC	Washington, D.C.	1.17	0.54	0.86
TC	Proctor Maple RF	1.13	0.90	0.47
Calcium	Addison Pinnacle	0.74	0.00	0.86
Calcium	Washington, D.C.	0.53	0.03	0.25
Calcium	Proctor Maple RF	1.70	0.01	0.34
Aluminum	Addison Pinnacle	0.22	0.00	0.54
Aluminum	Washington, D.C.	0.05	0.01	0.01
Aluminum	Proctor Maple RF	0.17	0.01	0.02
Iron	Addison Pinnacle	0.72	0.01	0.93
Iron	Washington, D.C.	0.65	0.02	0.50
Iron	Proctor Maple RF	1.73	0.01	0.26
Titanium	Addison Pinnacle	0.08	0.00	0.20
Titanium	Washington, D.C.	0.15	0.00	0.09
Titanium	Proctor Maple RF	0.16	0.00	0.12
Silicon	Addison Pinnacle	0.53	0.01	0.65
Silicon	Washington, D.C.	0.56	0.03	0.44
Silicon	Proctor Maple RF	0.76	0.02	0.44
Soil	Addison Pinnacle	0.57	0.04	0.84
Soil	Washington, D.C.	0.71	0.11	0.57
Soil	Proctor Maple RF	0.93	0.15	0.35
S/$(NH_4)_2SO_4$	Addison Pinnacle	1.06	- 0.02	0.97
S/$(NH_4)_2SO_4$	Washington, D.C.	1.09	- 0.04	0.96
S/$(NH_4)_2SO_4$	Proctor Maple RF	1.17	- 0.04	0.90
Sulfate Ion	Addison Pinnacle	1.00	0.23	0.98
Sulfate Ion	Washington, D.C.	1.01	- 0.07	0.96
Sulfate Ion	Proctor Maple RF	1.07	0.17	0.93
Nitrate Ion	Addison Pinnacle	1.14	0.03	0.96
Nitrate Ion	Washington, D.C.	1.19	0.01	0.95
Nitrate Ion	Proctor Maple RF	1.70	0.43	0.79
EC + 40% OCX2	Addison Pinnacle	0.94	0.16	0.30
EC	Washington, D.C.	1.02	0.57	0.38
EC	Proctor Maple RF	0.89	0.53	0.22
EC + 100% OCX2	Addison Pinnacle	1.87	0.23	0.32
EC	Washington, D.C.	1.72	0.92	0.45
EC	Proctor Maple RF	2.11	0.78	0.26
EC Original	Addison Pinnacle	0.32	0.12	0.13
EC	Washington, D.C.	0.55	0.33	0.21
EC	Proctor Maple RF	0.08	0.37	0.005
OC - 40% OCX2	Addison Pinnacle	0.92	- 0.06	0.52
OC	Washington, D.C.	1.00	0.57	0.84
OC	Proctor Maple RF	1.10	0.41	0.55
OC -100% OCX2	Addison Pinnacle	0.67	- 0.15	0.55
OC	Washington, D.C.	0.80	0.17	0.85
OC	Proctor Maple RF	0.79	0.18	0.60
OC Original	Addison Pinnacle	1.09	0.00	0.50
OC	Washington, D.C.	1.13	0.85	0.81
OC	Proctor Maple RF	1.30	0.57	0.52

The poor results observed from the Vermont pair may be due to the proximity of Vermont's STN site to sources of nitrogen not seen at Proctor Maple R.F. In addition, the MET ONE sampler at Burlington has a substantially lower sample flow rate than the IMPROVE sampler, which may affect collection efficiency and volatilization from the filter. Other differences exist in the denuder and sample shipping protocols that may affect the data. The data were also analyzed on a seasonal basis to see what differences might exist from time of year. In all cases the STN measured significantly greater levels of nitrate than IMPROVE. As expected, the range of values differed from season to season, with summertime having the lowest, tightest range and winter the highest, widest range.

In future analyses, efforts may focus on testing the assumption of fully ammoniated sulfate. The STN monitors ammonium that can be used to determine aerosol acidity. Atmospheric availability of ammonia will be of great concern in future haze programs in MANE-VU as the balance between sulfate and nitrate may shift.

A.4 CARBON

Carbon measurements present the greatest challenge for comparing results between the networks. The total carbon measured by both the IMPROVE and modified NIOSH techniques is similar (USEPA 2002).[7] However, the split between EC and OC occurs at a different point in the carbon evolution. This discrepancy can have a substantial effect on the reported mass of EC and OC. Equally important are the implications for extinction calculations, as organic carbon is multiplied by 1.4 to obtain the organic mass. Then this value is multiplied by a dry scattering coefficient of 4 m^2/g while the EC mass is multiplied by 10 m^2/g to account for scattering.

Analysis of the calculated total carbon mass shows reasonable agreement. The IMPROVE OC/EC data calculation methodology is summarized in Table 1. The reported results for IMPROVE have been blank corrected based on OC levels recorded on backup filters. For the STN data, a sampler specific annualized blank correction factor was applied to the measured OC before summation with measured EC (personal communication, V. Rao). The plotted carbon sums reveal slopes close to one (0.98, 1.17 and 1.13 for New York, Washington, D.C. and Vermont) and intercepts of 0.03, 0.54 and 0.90 for the New York, Washington, D.C. and Vermont sites, respectively (Figures 5a, 6a and 7a). Since the New York site is collocated, it seems reasonable that on average that comparison should have the best agreement. However, a significant number of points lie above the best fit line for NY, which indicates STN often measures higher carbon mass than IMPROVE. There is still substantial scatter in the data that is likely due, in part, to the application of an annual blank correction factor rather than a sample or season specific value. Thus, individual measurement pairs may not compare favorably to the extent that the blank correction process lacks adequate temporal resolution.

The analytical temperature profiles and combustion atmosphere play a substantial role in creating the observed differences. Both methods start the process in an inert atmosphere of helium. Discrete temperature ramping is applied to mobilize the carbon into the gas phase. For IMPROVE, the temperature does not rise above 550 °C in this inert atmosphere. Four separate fractions are reported based upon the temperature and time of evolution (OC1, OC2, OC3 and OC4). Oxygen is then added to the mix, keeping the temperature constant. The carbon measured from the point of

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[7] Similarity is for total mass of carbon. This does not include the adjustment factor of 1.4 traditionally used to account for other elements like oxygen and nitrogen that contribute to the mass of organic carbon.

oxygen introduction to the time that the reflectance reaches the initial value defines a fifth fraction, OP. The first EC fraction includes OP and any further carbon evolved at 550 °C. The final two IMPROVE EC fractions evolve at higher temperatures.

The STN method ramps temperature to 900 °C during the inert phase. This high temperature drives off more carbon from the filter than is driven off in the corresponding inert atmosphere IMPROVE segment. Again, the delineation between EC and OC is set when the transmittance reaches its original value. Subsequently, oxygen is added and the remaining carbon is oxidized and detected. Therefore, OC determined by STN in general is greater than that for IMPROVE. Correspondingly, the EC is less for STN (Watson, 2002).

Filter blanks and gas adsorption onto filters constitute another problem area for carbon. In an ideal world, blank values would be minimal when compared to measured mass. Due to the ubiquitous nature of gas-phase carbon and the affinity of quartz for these gases, "clean" blanks are rare. Therefore, samples must be corrected for background levels of carbon. Both STN and IMPROVE use a system-wide average. The correction factor is inversely related to sample volume/flow rate (USEPA, 2001a). Depending on the sampler, correction for STN ranges from 0.56 to 1.40 $\mu g/m^3$. The IMPROVE factor is less as it operates under higher flow conditions. The STN preliminary blanks are intended as an annual correction factor and have been applied here for individual measurements. This potentially introduces substantial error when comparing data from an individual day, but may be less problematic when comparing the seasonal averages and best-fit slopes. A useful future approach, given the large annual blank correction factors, would include a seasonal analysis of blank values to determine any seasonal dependency.

Differences in optical technique may also play a small role in reported differences between the networks. The point of intersection defined by reflectance and transmittance may differ even when identical temperature and atmosphere conditions exist. The magnitude of these variations is expected to be substantially less than that from differences in thermal evolution conditions.

Although the STN methodology may come closer to reporting "true" EC and OC than does IMPROVE, not all light-absorbing carbon is elemental. For the purposes of visibility, the desired fractions are organic carbon and light-absorbing carbon, which includes all EC and some part of OC. The calculations specified for reconstructed light extinction were developed and optimized using data collected from the IMPROVE network. It is this approach that must be followed to comply with the regional haze regulations.

USEPA recognized early on that STN carbon results would be different from IMPROVE. With the contractor for STN carbon measurements, they pursued what was hoped to be a solution. Starting in March 2001, a new carbon fraction was reported by the STN (USEPA, 2002). This fraction, OCX2, was defined as carbon evolved between 550 and 900 °C in the inert atmosphere. This definition was based on the reasonable expectation that since the IMPROVE temperature for organic carbon does not surpass 550 °C, that this temperature should define the transition from OC to EC. Unfortunately, simply moving OCX2 to OC from EC does not create agreement between the carbon values of the two approaches.

Since on average the total measured carbon mass is equal, the possibility of devising a reasonably accurate conversion from STN to IMPROVE may exist. Using data from the collocated New York site and the Washington, D.C. pair, STN OC adjusted by removing OCX2 was compared to IMPROVE OC1, OC2, OC3 and OC4. This analysis demonstrates that on average, adjusted STN OC is at least as great as the sum of the first three IMPROVE fractions, but less than the combination of the first four fractions. The best fit, on average, includes OC1, OC2, OC3 and about forty percent of OC4. At the comparison sites, moving OCX2 to EC from OC apparently overcorrects the fractions for comparison to IMPROVE OC and EC.

Figure 5. Addison Pinnacle State Park STN and IMPROVE (a) Total Carbon = EC + OC (b) Organic Carbon: Original STN EC/OC split, OC with all OCX2 removed, OC with 40% OCX2 removed (c) Elemental Carbon: Original EC, EC with all OCX2 added, EC with 40% OCX2 added (d) B_{ext} from OC + EC: Original OC/EC, "IMPROVE-like" OC/EC with OCX2 considered EC, Adjusted "IMPROVE-like" with 40% OCX2 considered EC, Average B_{ext} = TC*7.8

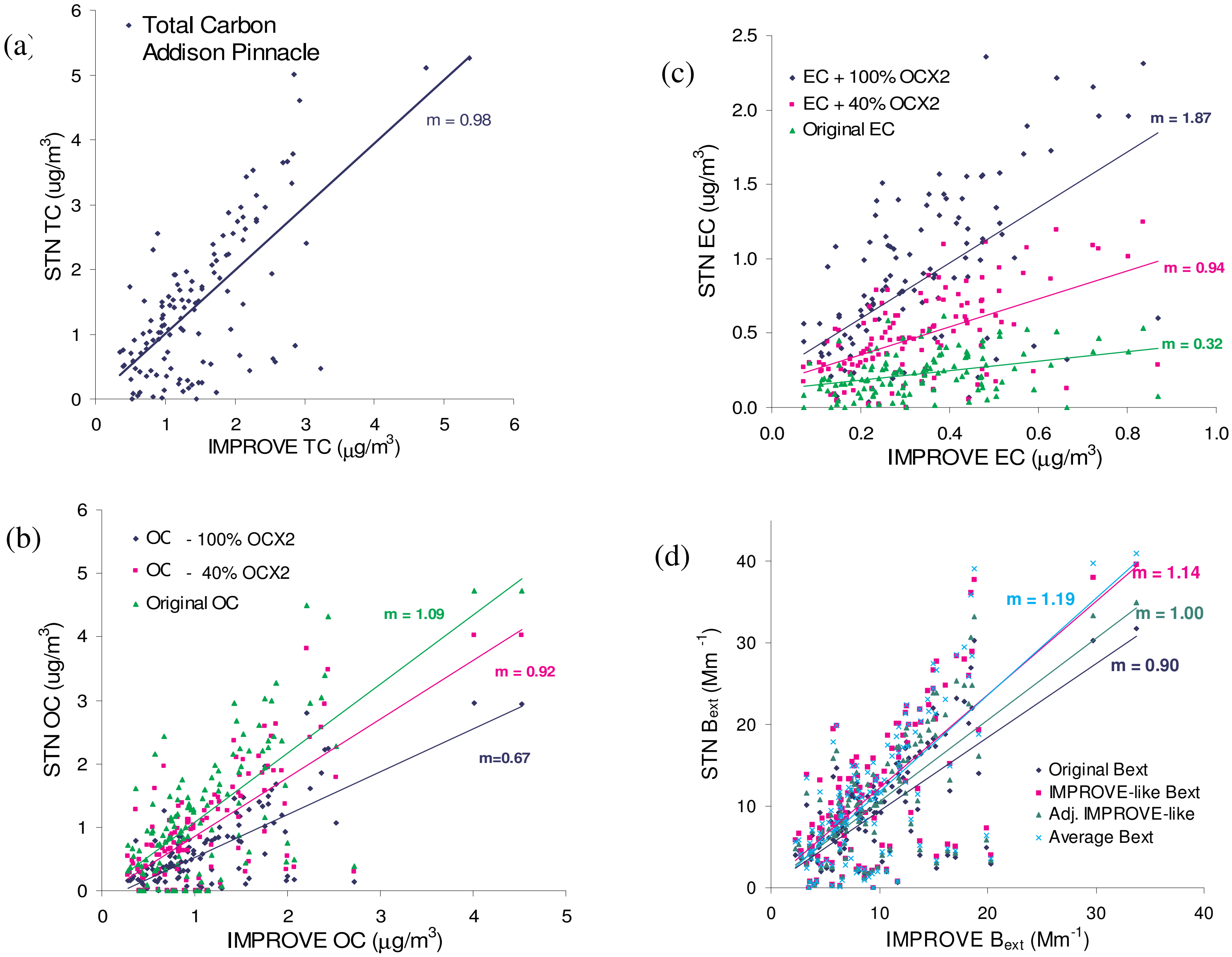


Figure 6. McMillan Reservoir STN and Washington, D.C. IMPROVE (a) Total Carbon = EC + OC (b) Organic Carbon: Original STN EC/OC split, OC with all OCX2 removed, OC with 40% OCX2 removed (c) Elemental Carbon: Original EC, EC with all OCX2 added, EC with 40% OCX2 added (d) B_{ext} from OC + EC: Original OC/EC, "IMPROVE-like" OC/EC with OCX2 considered EC, Adjusted "IMPROVE-like" with 40% OCX2 considered EC, Average B_{ext} = TC*7.8

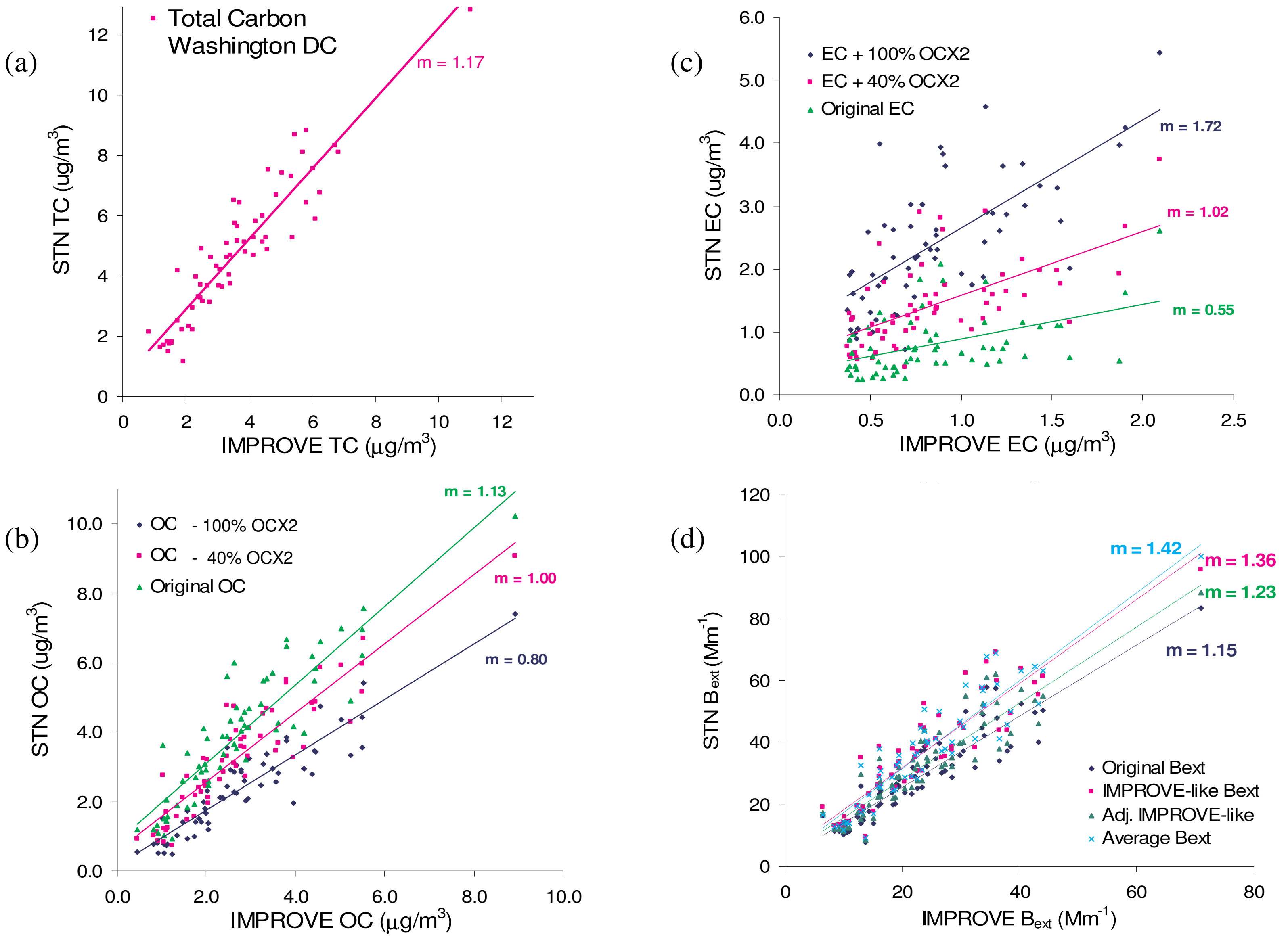
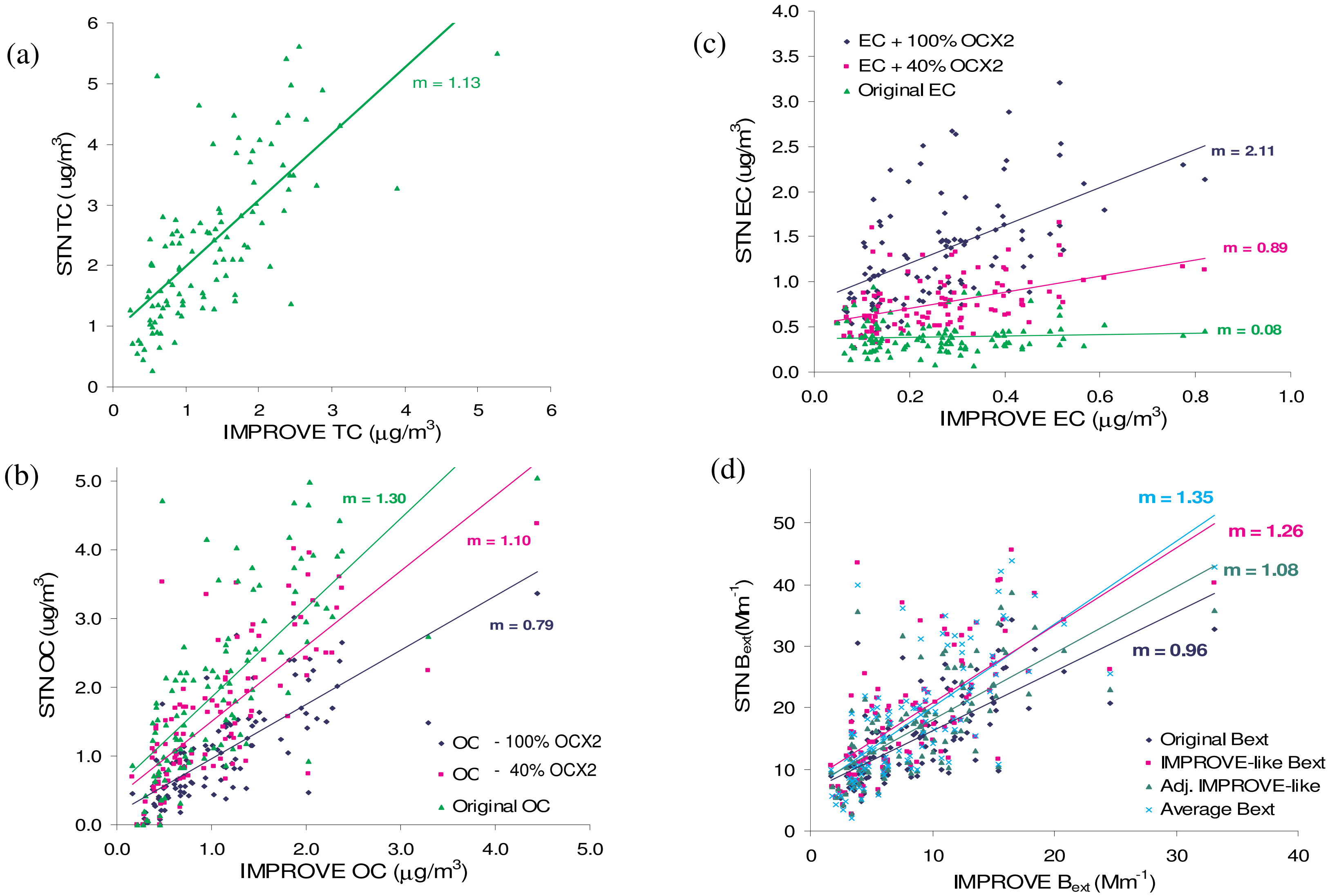


Figure 7. Burlington STN and Proctor Maple RF IMPROVE (a) Total Carbon = EC + OC (b) OC: Original STN EC/OC split, OC with all OCX2 removed, OC with 40% OCX2 removed (c) EC: Original EC, EC with all OCX2 added, EC with 40% OCX2 added (d) B_{ext} from OC + EC: Original OC/EC, "IMPROVE-like" OC/EC with OCX2 considered EC, Adjusted "IMPROVE-like" with 40% OCX2 considered EC, Average B_{ext} = TC*7.8



Further comparison of OC and EC was conducted to investigate different applications of the OCX2 factor. The initial step was to correct the STN data for the OC blank. Since reported OC includes OCX2, the blank correction must be apportioned between OCX2 and the remaining OC, denoted here as OC'.[8] In other words, the measured quantity OC is equal to the sum of OCX2 and OC'. In some cases the blank exceeded the recorded value of OC and consequently, mass of OC for those samples was recorded as zero.[9] This demonstrates the weakness in using an annual average blank to correct sample mass. However, no other information on blanks for STN carbon is available.[10]

After blank correction, two different sets of OC/EC mass splits were calculated from the STN data. These were then compared to the OC/EC split derived from the IMPROVE measurements, which served as a benchmark. The first set of STN OC/EC values used for comparison to this benchmark are the directly measured EC and OC, with OC corrected for the blank. In this case, the OC used is the reported OC value minus the blank (the OCX2 factor is not considered). The second OC/EC split is calculated following USEPA's initial thinking. Here, the reported OC value has been blank corrected and then the blank corrected OCX2 has been subtracted off. The EC value used is the reported EC with the blank corrected OCX2 factor added. Based on the plots of these data, it is apparent that neither of these two approaches for comparing network OC/EC splits results in a successful transformation relationship. Rather, making no adjustment to reported STN values yields greater STN OC relative to IMPROVE and subtracting the OCX2 factor yields STN values of OC significantly less than that measured by IMPROVE.

Based on these findings, a third OC/EC split was derived by partitioning the blank corrected OCX2 factor into both an OC and EC component. In order to develop the best fit between collocated data sets from the two networks, forty percent of the blank corrected OCX2 factor was transferred from the reported STN OC factor to the reported EC factor, leaving sixty percent as a component of STN OC. This derivation, while wholly subjective, does allow for a network comparison with total carbon apportioned in a similar manner at the few representative collocated sites that were available for analysis. The limitations in terms of number of sites, blank correction complexities, and limited number of carbon fractions reported by the STN (only three) will hopefully spur continued research and eventually the development of a more robust mechanism for direct comparison of STN to IMPROVE.

The results of the EC and OC carbon analyses are plotted in Figures 5b and c, 6b and c and 7b and c. The same general behavior is observed for all three site pairs for EC and OC. The various

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[8] The possibility exists that the blank correction should not be applied in a proportional manner and should be weighted more toward the lighter, OC' factor. However, the exact proportions are unknown and a proportional treatment is likely to be the lower bound for the quantity removed from the lighter fraction of OC. Therefore, blank corrected OC' equals uncorrected OC' minus [(OC')/(OC' + OCX2)]*(Blank Correction Factor) and blank corrected OCX2 equals uncorrected OCX2 minus [(OCX2)/(OC' + OCX2)]*(Blank Correction Factor).

[9] The blank exceeded OC in roughly ten percent of the cases in New York, three percent of the samples from Vermont and no samples from Washington.

[10] The average IMPROVE OC mass for the "zero" STN samples was 0.77 $\mu g/m^3$, while the average for the remaining samples was 1.22 $\mu g/m^3$.

treatments of the data have minimal effects on the coefficient of determination of the best-fit line for OC. However, both the slope and intercept are affected. The data without adjustment have the highest slopes (1.09 to 1.30) and intercepts, passing through the origin with the New York data and between 0.5 and 0.9 μg/m^3 for the other sites. The OCX2 correction changes the slopes to substantially less than one (0.67 to 0.80), but improves the intercepts to less than ±0.2 μg/m^3. The "optimized" approach attempts to adjust the slopes to one and yields a range of 0.92 to 1.10, with intercepts generally improved from the original OC/EC split.

The differences for EC are more dramatic since the magnitude of EC is generally less than OC. For the IMPROVE data, the ratio of OC to EC is three or four to one, whereas the blank corrected OC to EC ratio for STN is over five to one (and is almost twice that with no blank correction!). Since the OCX2 factor can be as much as half the recorded OC, shifting that entirely to EC impacts the OC to EC ratio considerably. Plots of the original EC value for STN versus IMPROVE EC show slopes substantially less than one (0.08 to 0.55) with intercepts ranging from 0.12 to 0.37 μg/m^3 and very low correlation. The OCX2 adjustment yields slopes of 1.7 to 2.1 and intercepts 0.2 to 0.9 μg/m^3. The "optimized" approach generates slopes of 0.9 to 1.0 with intercepts of 0.2 to 0.6 μg/m^3. Both corrected ratios improve the correlation, though abundant scatter remains.

Although the various carbon mass fractions compared poorly, the corresponding extension to atmospheric extinction was made. Figures 5d, 6d and 7d display the results, with four curves produced for each site. The first three are based on the previous mass apportionment schemes and the dry extinction coefficients for EC and OC (Original B_{ext}, "IMPROVE-like" B_{ext}, and Adjusted "IMPROVE-like" B_{ext}). In these three cases, the 1.4 mass factor for OC was incorporated into the calculation. The fourth approach assumes knowledge only of total carbon and uses an average extinction of 7.8 m^2/g.[11] This last result does not vary significantly from the "IMPROVE-like" curve, implying OC[12] and EC mass for that case is in a ratio of roughly 1.8 to 1.

The results are best for the New York site, as might be expected based on the total mass comparison. The slopes of all the curves are within 20% of unity and the intercept is within ±0.8 Mm^{-1}. For Washington, the slopes show STN averaging fifteen to forty percent higher than IMPROVE with intercepts on the order of 3 Mm^{-1}. Agreement for the Vermont results is also poor with slopes ranging from 0.96 to 1.34 and intercepts greater than 6.5 Mm^{-1}. In all cases, the agreement between the original and adjusted 'IMPROVE-like' is similar and better than the other two approaches. Since the original EC/OC split for STN is likely not comparable to the IMPROVE split and differences between the extinction calculations using the original and adjusted split are small, all STN carbon data used in subsequent analyses has been adjusted by allocating 40% of OCX2 to EC.

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[11] This represents the average of (4*1.4) and 10, where the 4 is the dry scattering for organic carbon mass, 1.4 accounts for organic mass associated with the measure mass (carbon only) and 10 is the dry scattering coefficient for elemental carbon. This approach does not split the mass of C into EC and OC and results in an average scattering of carbon mass. This is equivalent to assuming an even split between OC and EC.

[12] Here OC refers to mass of organic carbon, which includes mass of N, O and other elements.

III. Other Data Analysis

A. Period Analyzed

Data are available from the IMPROVE program from its inception through May 2002. The STN data are available from early 2000 through July 2002. Data from the most recent months available present the broadest spatial coverage. Therefore, data were analyzed for the period between summer 2001 and spring 2002. The difference in coverage obtained by not starting three months earlier is substantial, as the STN had roughly ten sites available during spring 2001 but almost twenty were available for the summer.

B. Missing Data

The general approach in comparing the STN and IMPROVE data is to keep as many measurements as possible. This maximizes the total number of points for comparison and minimizes the uncertainty in seasonal and annual average calculations to the extent possible. Some constituents have better data capture than others. Seasonal and annual averages are calculated for each component, but there may be an unequal number of data points in these averages. The USEPA draft guidelines for tracking reasonable progress (USEPA, 2001) include recommendations for data substitution which were used to fill in missing data when possible.

The guidelines recommend using the longest data record available, preferably five years worth, when considering data substitution. Only one year was considered in this study. Given this limitation, seasonal averages were calculated rather than calendar quarter averages as it is expected a seasonal average is more appropriate than the calendar quarter average. For example, December is more like January and February than is March. Once the seasonal average for a component was determined, this average was used to replace all of the valid data for that component. Then, overall extinction was calculated for all valid days using in one case, the original data and for the other, the average. The criterion used for allowable substitution is that the difference between these two extinction values must be within ten percent for ninety percent of the days. When this condition is met, the average value for that component may be substituted for missing values. For those cases where multiple components were missing, the evaluation was done simultaneously. The points evaluated were those that had valid data for every component. For example, if one day was missing one component and another day was missing a second component, neither day was used in the calculation of total extinction. However, all individual values were used in deriving the average of that component. For example, assume there are ten days in the period. Every day has soil and sulfate values, but nitrate is missing on the first sample date and OC/EC are missing on the second and sixth. Nine values are used to determine the nitrate average for the period and eight OC/EC values are used for their averages. However, only seven days have all components, so the comparison of total extinction for the period versus extinction calculated using substituted average values for missing components, uses only those seven dates.

Once the substitution test is complete, season and annual data capture can be evaluated. Valid seasonal averages are defined as having at least fifty percent data capture. Annual averages require seventy-five percent overall capture with four valid quarters. IMPROVE sites had fewer missing averages in general. Many of the STN sites lack annual averages since they lacked twelve months of data. These differences are shown in the tables in Appendix A and Figures in Appendix B.

Updated guidelines will be available in 2003. Some anticipated modifications included in this update will be a change from using averages to medians in missing data substitution. The overall procedure for substitution will also be clarified. Another change deals with the relative humidity (RH) factor used for hygroscopic aerosol constituents. Monthly averages were recalculated for the entire country using a ninety-five percent RH cut-off. Time periods with RH greater than this will average in a constant factor equal to that of the ninety-five percent RH factor.

These new f(RH) values will be publicly available in early 2003. USEPA permitted its contractor to release to MANE-VU these updated values for use in this memorandum. These are listed in Table 3.

C. Sampling Frequency Effects

The use of a limited data set can often introduce complications in the evaluation of results for data analyses. The analyses in this document consider data collected from nearly fifty sites. The majority of these sites operated on a one-in-three day sampling schedule. However, some STN sites operated one-in-six days and others operated on a varying schedule ranging from every day to a one-in-six day schedule.

A primary issue for one-in-three day sites is the representativeness of the available data. Can a once every third day sample schedule accurately capture daily variations? In general, the longer the time span considered the better the result. For an annual period, the difference in the annual average calculated from a daily sample schedule and from a one-in-three day schedule is small. When the period is shortened to a three months of data, the potential deviation between averages calculated from these two schedules increases. Similarly, comparison of one-in-six day schedule to daily sampling for a year-long period could have higher variability than one-in-three day samples. These potential deviations from the “true” average calculated by daily sampling are influenced by the distribution of the data. The further the data deviate from a normal distribution, and these data do not fit the normal distribution, the more likely averages calculated from a subset will deviate from the average of all data.

Any interpretations of the extinction plots in this memorandum should account for this dependence of calculated averages on sample collection schedule and data completeness. Two different analyses were performed to provide reasonable estimates for these effects. First, data from New Brunswick, NJ were examined, comparing collocated monitors on different sampling schedules (Table 4). Second, the effect of removing a discrete number of days from a sample set was investigated for MK Goddard, PA (Table 5). The results of these two examples will not represent every possibility and are meant to remind the reader of the order of magnitude deviations to be expected as a result of these sampling differences.

Table 3. Summary tables of the new monthly relative humidity factors used in calculating visibility extinction. The upper table lists values for IMPROVE sites and the lower for STN.

Site Name	State	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
Acadia National Park	ME	3.22	2.81	2.81	3.15	3.18	3.26	3.65	3.71	3.88	3.48	3.43	3.53
Addison Pinnacle	NY	3.05	2.82	2.83	2.65	2.88	2.93	3.10	3.32	3.48	3.19	3.03	3.13
Arendtsville	PA	2.84	2.59	2.68	2.40	2.78	2.84	2.98	3.12	3.21	2.95	2.73	2.89
Brigantine National Wildlife Refuge	NJ	2.89	2.64	2.73	2.59	2.92	2.99	3.19	3.37	3.37	3.24	2.84	2.91
Bridgton	ME	2.78	2.52	2.62	2.75	2.87	2.96	3.24	3.41	3.53	3.20	2.94	2.87
Casco Bay	ME	2.68	2.43	2.53	2.68	2.84	2.91	3.15	3.29	3.36	3.07	2.79	2.73
Cape Cod	MA	2.67	2.47	2.59	2.63	2.78	2.83	3.06	3.19	3.28	3.07	2.78	2.70
Connecticut Hill	NY	3.34	2.97	3.01	2.72	2.72	2.68	2.93	3.17	3.44	3.09	3.26	3.43
Great Gulf Wilderness	NH	2.81	2.55	2.64	2.76	2.86	2.99	3.27	3.47	3.60	3.24	3.00	2.92
Lye Brook Wilderness	VT	2.78	2.57	2.65	2.63	2.80	2.86	3.06	3.27	3.40	3.18	2.90	2.87
M.K. Goddard	PA	3.31	3.10	3.00	2.72	2.95	3.15	3.37	3.61	3.75	3.25	3.07	3.43
Mohawk Mt.	CT	2.77	2.56	2.66	2.61	2.82	2.88	3.07	3.24	3.35	3.16	2.85	2.82
Moosehorn NWR	ME	3.01	2.66	2.68	2.93	2.94	3.05	3.45	3.61	3.77	3.33	3.18	3.19
Old Town	ME	3.00	2.68	2.69	2.84	2.75	2.86	3.27	3.45	3.51	3.04	2.92	3.00
Proctor Maple R. F.	VT	2.81	2.60	2.66	2.71	2.76	2.84	3.09	3.35	3.48	3.18	2.98	2.95
Presque Isle	ME	2.93	2.57	2.56	2.84	2.79	2.89	3.09	3.44	3.63	3.47	3.32	3.25
Quabbin Summit	MA	2.75	2.54	2.65	2.61	2.77	2.84	3.04	3.22	3.33	3.13	2.85	2.82
Washington D.C.	DC	2.84	2.60	2.65	2.42	2.83	2.84	2.96	3.11	3.18	3.06	2.75	2.84
Dolly Sods	WV	2.95	2.74	2.74	2.49	3.45	3.09	3.24	3.45	3.52	3.12	2.84	3.06
James River Face	VA	2.85	2.66	2.64	2.39	2.90	3.06	3.16	3.34	3.41	3.03	2.73	2.95
Quaker City	OH	3.16	2.92	2.85	2.55	2.96	3.10	3.22	3.46	3.52	3.01	3.06	3.26
Shenandoah	VA	2.89	2.62	2.65	2.41	2.91	3.11	3.24	3.47	3.46	2.97	2.71	2.94

Site Name	State	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sept	Oct	Nov	Dec
Dover	DE	2.82	2.58	2.66	2.46	2.82	2.87	3.04	3.20	3.24	3.09	2.75	2.85
MLK Wilmington	DE	2.87	2.59	2.70	2.49	2.84	2.83	3.03	3.16	3.24	3.16	2.78	2.84
McMillan Reservoir	DC	2.84	2.60	2.65	2.42	2.83	2.84	2.96	3.11	3.18	3.06	2.75	2.84
Baltimore	MD	2.85	2.60	2.68	2.47	2.84	2.89	3.03	3.20	3.26	3.11	2.77	2.89
Fort Meade	MD	2.87	2.61	2.70	2.48	2.85	2.88	3.00	3.16	3.23	3.15	2.79	2.90
Essex	MD	2.85	2.60	2.68	2.47	2.84	2.89	3.04	3.20	3.27	3.11	2.77	2.88
Chicopee	MA	2.72	2.52	2.63	2.58	2.77	2.84	3.04	3.21	3.33	3.14	2.83	2.80
Roxbury	MA	2.61	2.46	2.61	2.63	2.78	2.73	2.91	3.04	3.13	2.98	2.68	2.61
Manchester	NH	2.82	2.60	2.69	2.75	2.93	2.99	3.24	3.41	3.59	3.31	3.02	2.94
Portsmouth	NH	2.74	2.52	2.64	2.71	2.87	2.92	3.17	3.32	3.43	3.16	2.88	2.80
Camden	NJ	2.79	2.55	2.63	2.45	2.77	2.79	2.97	3.11	3.18	3.10	2.74	2.81
New Brunswick	NJ	2.78	2.55	2.65	2.53	2.82	2.86	3.04	3.17	3.23	3.09	2.75	2.80
Chester	NJ	2.79	2.56	2.66	2.55	2.82	2.86	3.04	3.19	3.26	3.11	2.78	2.81
Elizabeth	NJ	2.71	2.49	2.58	2.51	2.76	2.72	2.85	2.96	2.99	2.93	2.65	2.68
Botanical Garden	NY	2.69	2.48	2.59	2.55	2.79	2.77	2.89	3.00	3.03	2.94	2.66	2.66
IS 52	NY	2.69	2.48	2.59	2.55	2.79	2.77	2.89	3.00	3.03	2.94	2.66	2.66
Buffalo	NY	3.10	3.01	2.89	2.68	2.77	2.90	2.92	3.14	3.31	3.05	3.06	3.23
Whiteface	NY	2.73	2.60	2.60	2.82	2.76	2.65	2.86	3.30	3.40	3.21	2.99	2.79
Rochester	NY	2.96	2.94	2.92	2.78	2.83	2.92	3.06	3.27	3.50	3.23	3.15	3.15
Queens College	NY	2.70	2.50	2.62	2.59	2.84	2.82	2.92	3.00	3.00	2.95	2.66	2.66
Addison Pinnacle	NY	3.05	2.82	2.83	2.65	2.88	2.93	3.10	3.32	3.48	3.19	3.03	3.13
Lawrenceville	PA	3.07	2.86	2.83	2.55	2.92	3.02	3.16	3.36	3.50	3.11	2.94	3.16
Philadelphia	PA	2.65	2.42	2.51	2.33	2.64	2.73	2.93	3.08	3.18	3.02	2.63	2.76
Florence	PA	3.09	2.88	2.84	2.54	2.90	2.99	3.11	3.31	3.46	3.05	2.94	3.19
Greensburg	PA	3.08	2.87	2.84	2.57	2.94	3.04	3.19	3.41	3.53	3.14	2.95	3.17
Burlington	VT	2.78	2.59	2.64	2.67	2.70	2.77	3.00	3.29	3.41	3.14	2.94	2.93

Table 4 assesses the effects of one-in-three versus one-in-six day sampling at New Brunswick, New Jersey. Two collocated monitors sample at this site, each operating under one of the two schedules. Annual and seasonal average extinction is calculated for both monitors for five components and their sum, fine extinction. The one-in-three day monitor collected 96 days of valid samples, with 22 samples for the fall, 26 for the summer and 24 each for winter and spring. The corresponding one-in-six day sample collection was 11 days for fall and winter, 13 for summer and 15 for spring. A third group of averages was calculated from the one in three monitor, using only those days that corresponded to days collected by the other monitor.

Relative percent difference (RPD) calculations were performed to evaluate differences between a one-in-three and one-in-six day schedule using the first monitor. By comparing values from one monitor, the differences in monitoring precision are not a factor. The RPDs are as high as 22% for summer nitrate and average 8% for the individual components. The overall differences in fine extinction are somewhat less, as the variation within the five components are offset when summed.

Comparison of the two one-in-six day data sets gives an estimate of instrumental precision. As shown in the table, the overall precision of the measurements does not differ greatly from the differences between sampling schedule averages. The greatest deviation of 41% was observed for summer organic carbon, while the average deviation was 7%. For this site, it appears that the differences in average extinction calculations are of the same magnitude for both a one-in-six day versus a one-in-three day schedule and for values measured by two different samplers.

The second approach for evaluating differences caused by averaging different sample dates is contained in Table 5. The data used in the calculations are from the MK Goddard site, where every sampling day (121 total) in the 12 month period was valid. Two different data subsets were created and the corresponding averages calculated. The first case, which may represent a "worst-case" scenario, removes from the data the two days with the highest fine extinction in each season. Most of the relative percent differences between averages from the full data set and the "worst-case" data set are within 10%. The largest difference is seen in sulfate averages and implies that 24-hour sulfate measurements are skewed more than the other constituents.

By removing three randomly selected days from each season, another data set was generated. For this case the differences in the averages were small and mostly under 3%. This implies that if missed sampling days are randomly distributed within a network, the effect on averages calculated from the measurements will be minimal.

The IMPROVE network had very high data collection rates for the period studied. Based on the previous analysis, inter-site comparisons should be valid. However, data collection at STN sites was more varied and site to site comparison may be substantially affected. Even so, large differences between site averages likely represent true differences.

Table 4. Data from New Brunswick STN site. Two monitors are located here, sampling on a 1 in 3 and 1 in 6 day schedule, for monitors 1 and 2 respectively. There were 96 valid sample days from monitor 1 and 50 valid days from monitor 2. The first data block [1a] displays averages in Mm^{-1} for all sample days from monitor 1. The second block [1b] includes the 50 days from monitor 1 that coincide with the 50 days from monitor 2. Monitor 2's data are in the third block [2]. Relative percent differences (RPD) are shown in the fourth [3a] and fifth [3b] blocks, where RPD equals the difference divided by the average of two values. Results in the fourth block give RPD for monitor 1 when comparing the averages of the full data set to a subset of 50 days (one possible 1 in 6 schedule). In the fifth block, RPD of the 1 in 6 day averages from monitor 1 and monitor 2 are presented.

Season	b_{ext} soil	b_{ext} EC	b_{ext} OC	B_{ext} SO_4	B_{ext} NO_3	B_{ext} Fine
[1a] Monitor 1 frequency 1 in 3 days						
Winter	0.33	9.47	9.33	28.87	24.71	72.71
Spring	0.46	8.79	6.35	37.74	15.14	69.83
Summer	0.40	11.05	13.98	73.86	10.77	110.07
Fall	0.47	12.07	11.20	44.45	14.42	84.38
Annual	0.42	10.31	10.20	46.63	16.16	84.78
[1b] Monitor 1 frequency 1 in 6 days						
Winter	0.29	9.85	9.04	24.71	24.93	68.82
Spring	0.44	9.30	5.97	38.19	14.31	68.21
Summer	0.37	10.58	12.91	80.79	13.41	118.07
Fall	0.41	11.08	10.00	52.03	13.99	87.50
Annual	0.38	10.15	9.34	49.35	16.34	85.55
[2] Monitor 2 frequency 1 in 6 days						
Winter	0.28	9.83	9.37	24.73	25.32	69.53
Spring	0.53	8.92	7.04	39.16	14.46	70.10
Summer	0.38	12.49	19.52	84.32	13.39	130.10
Fall	0.42	11.55	11.30	52.76	14.70	90.73
Annual	0.41	10.63	11.73	50.72	16.62	90.11
[3a] Relative Percent Difference [1a] & [1b]						
Winter	13%	-4%	3%	16%	-1%	5%
Spring	5%	-6%	6%	-1%	6%	2%
Summer	8%	4%	8%	-9%	-22%	-7%
Fall	14%	9%	11%	-16%	3%	-4%
Annual	9%	2%	9%	-6%	-1%	-1%
[3b] Relative Percent Difference [1b] & [2]						
Winter	5%	0%	-4%	0%	-2%	-1%
Spring	-18%	4%	-16%	-3%	-1%	-3%
Summer	-3%	-17%	-41%	-4%	0%	-10%
Fall	-2%	-4%	-12%	-1%	-5%	-4%
Annual	-7%	-5%	-23%	-3%	-2%	-5%

Table 5. Example calculations demonstrating the effect of including different sample days in annual and seasonal averages. Data are from MK Goddard with units of Mm^{-1}. Data block [a] gives averages from the complete 121 day sample set. The two highest visibility days were removed from [a] for each season (8 points overall) and the resulting averages are displayed in Block [b]. Similarly, Block [c] gives averages when three randomly chosen dates were removed from each season. Relative Percent Difference calculations are given in [d] and [e], representing the difference between [a] and [b]; and [a] and [c], respectively.

Season	b_{ext} soil	b_{ext} EC	b_{ext} OC	B_{ext} SO_4	B_{ext} NO_3	B_{ext} Fine
[a] Complete data capture of 1 in 3 day schedule						
Winter	0.31	4.24	8.31	33.07	26.76	72.68
Spring	0.45	4.13	7.43	37.33	11.71	61.05
Summer	0.44	6.59	11.29	86.40	5.89	110.60
Fall	0.41	5.54	10.68	53.27	10.55	80.45
Annual	0.40	5.13	9.44	52.52	13.70	81.19
[b] Same data with two worst (by season) visibility days removed.						
Winter	0.31	4.12	7.94	31.04	24.91	68.31
Spring	0.43	3.98	7.14	33.99	11.86	57.41
Summer	0.42	6.41	11.03	76.20	6.15	100.22
Fall	0.38	5.22	10.58	41.82	10.89	68.89
Annual	0.39	4.94	9.18	45.73	13.43	73.67
[c] Same data with three randomly selected dates per season removed						
Winter	0.31	4.32	8.27	33.64	26.16	72.70
Spring	0.45	4.01	7.28	36.59	11.62	59.95
Summer	0.45	6.78	11.50	87.10	5.96	111.79
Fall	0.40	5.46	10.56	48.04	10.29	74.75
Annual	0.40	5.14	9.41	51.31	13.48	79.75
[d] Relative Percent Difference [a] & [b]						
Winter	3%	3%	5%	6%	7%	6%
Spring	4%	4%	4%	9%	-1%	6%
Summer	4%	3%	2%	13%	-4%	10%
Fall	7%	6%	1%	24%	-3%	15%
Annual	5%	4%	3%	14%	2%	10%
[e] Relative Percent Difference [a] & [c]						
Winter	1%	-2%	0%	-2%	2%	0%
Spring	1%	3%	2%	2%	1%	2%
Summer	-3%	-3%	-2%	-1%	-1%	-1%
Fall	4%	1%	1%	10%	2%	7%
Annual	1%	0%	0%	2%	2%	2%

D. Contour Plots

Data tables (Appendix A) and contour plots (Appendix B) were created from the IMPROVE and STN particulate matter measurements. The tables provide both the values used in the plots and the number (n) of valid[13] measurement days included in the averaging period. Annual[14] and seasonal averages were calculated for coarse, soil, elemental carbon, organic carbon, ammonium sulfate and ammonium nitrate. Total fine extinction and total extinction averages were also calculated. The interpretation of these plots should be made in the context of the previous sections and limitations of the chosen ArcView parameters discussed next.

D.1 ArcView settings

Data tables were imported into ArcView (version 8.2) for plotting contour maps. Since $1/r^2$ is common for these types of plots, inverse distance weighting with a power of two was chosen. ArcView provides two options for radius of influence, constant or variable. The constant radius option allows the user to set a maximum distance of influence for a measurement and its inverse weighting is applied to any point within that area. When choosing variable radius, the user selects the number of points, n, to be used in calculating a location's value. The nearest n points are then weighted accordingly[15]. These maps use the nearest 6 values, where highly clustered sites influence a small area compared to those which are more sparsely distributed.

D.2 Plots

Appendix B contains all of the extinction plots, with five maps per figure (annual plus one for each season). Plots were created first for IMPROVE and then for STN sites. A merged plot of both networks was created for fine aerosol extinction, ammonium sulfate, ammonium nitrate, elemental carbon and organic carbon. The common plot scale for the three figures/fifteen maps in each group was determined based on the range of all values from both networks. Total extinction and coarse extinction maps were created only for IMPROVE, since STN does not determine coarse particulate mass. Merged plots for soil were not created based on the poor comparison discussed in section II A.2.

The IMPROVE sites are distributed more evenly across the MANE-VU region than are the STN sites. The more dense spatial coverage afforded by the expanded IMPROVE network permits a much better representation of the rural aerosol burden. STN sites available for this memo are clustered along the Washington to New York City metropolitan areas with several other urban locations throughout the region. When combined, these networks vastly improve our knowledge of the spatial and temporal distribution of speciated particulate matter.

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[13] A valid measurement day was considered to be a day in which all measured constituents were valid. Substituted values are considered valid for this purpose.

[14] Annual average weights each day equally as opposed to taking the average of the four seasons. These two calculation approaches could differ significantly if the number of valid points per season varies.

[15] A maximum radius of influence can be selected such that if n points are not within that radius, only those points that are will be used to determine the value at each location.

Conclusions regarding small-scale spatial patterns should not be drawn based on these contour plots. Although the expanded network improves the knowledge of the particulate composition throughout the region, it is impossible to know the true particulate makeup between monitored points. Additionally, the surrounding area accurately represented by a monitor will vary depending on the individual site characteristics and its environs (local sources, terrain, dominant meteorology).

Summed extinction plots

Appendix Figures 1a, b, and c show fine aerosol extinction plots. Appendix Figure 1d displays total extinction as determined by the IMPROVE network.[16] Based on the merged map (Figure 1a), visibility extinction is worst during the summer months and best during the spring. The general trend shows the worst visibility in the southwestern part of the region improving through the northeastern edge. Within this broad trend, it appears that urban areas have greater extinction then do the surrounding rural regions, as might reasonably be expected. This urban/rural difference is most pronounced for the Pittsburgh area and along the eastern seaboard with Washington, Baltimore and New York City and is seen to a lesser extent with Boston and Rochester.

The individual network maps (Figures 1b, c) show the same general behavior as does the merged map. Washington D.C., the only urban IMPROVE site, displays the greatest fine aerosol extinction for IMPROVE with the other sites generally cleaner in the northeastern-most part. Figure 1c. has the rural STN sites like Addison Pinnacle and Whiteface with the least impaired visibility levels.

Figure 1d represents the most important figure to some extent, because the haze rule is based on total aerosol extinction. The trend here is the same as that for fine extinction, which makes sense considering the relatively low contribution that coarse mass has to visibility reduction. This figure can also be compared to the three year averages (Figures S.4 and S.5, roughly years 1996-8) plotted in the May 2000 IMPROVE report (Malm, 2000). The trends are similar with extinction values generally within ±10%.

Ammonium Sulfate

The greatest contribution to visibility degradation in the MANE-VU region comes from ammonium sulfate (Appendix Figures 2a, b, c). The overall trend of highest values in the southwestern part to lowest in the northeastern occurs here and contributes this dependence to the overall fine extinction. Seasonal variation in sulfate also seems to drive the variation in fine extinction. The differences in urban and rural ammonium sulfate are small, although Pittsburgh appears to have a somewhat higher sulfate burden than the surrounding areas.

Ammonium Nitrate

Ammonium nitrate behaves differently from ammonium sulfate. Four figures are produced for this constituent. Appendix Figure 3a displays the merging of Appendix Figures 3b and 3c.

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[16] Fine extinction is the sum of the five fine parameters: ammonium sulfate, ammonium nitrate, elemental carbon, organic carbon and soil. Total extinction equals fine extinction plus extinction due to coarse matter and Rayleigh scattering. Within this memo, Rayleigh scattering is considered to be 10 Mm^{-1} for all sites.

Appendix Figure 3c and 3d are both derived from the STN values for nitrate and differ by 14%.[17] Measured nitrate values are greatest in the wintertime and least in the summer. This behavior is driven by temperature and ammonium nitrate stability. The annual behavior is primarily a function of wintertime values as they are much greater than the other seasons. Urban areas show higher concentrations than rural ones, with a slight west to east gradient superimposed on top. This is apparent in all seasons, with summer showing the lowest gradients. Both individual network maps show the same behavior, with urban or near urban sites possessing the highest values.

Elemental Carbon

The plots for elemental carbon display a third, different driver for visibility extinction (Appendix Figures 4a, b, c). Much less seasonal variation exists, with the seasonal figures and annual plot displaying similar trends and magnitudes. Urban areas dominate the elemental carbon burden and are especially concentrated in along the eastern seaboard metropolitan areas. Inspection of the IMPROVE only EC figures shows little variation for the plot scale (Figure 4b). The STN figure, on the other hand, confirms the urban/rural contrast as Whiteface and Addison Pinnacle have the lowest EC values (Figure 4c). This observation adds confidence to the result since the EC plotted for STN has been adjusted from the actual measured values. This adjustment was applied network-wide, so the urban/rural difference should not be an artifact of the transformation.

Organic Carbon

Organic carbon shares many of the characteristics seen with elemental carbon (Appendix Figures 5a, b, c). Seasonal variation is somewhat greater than that seen with EC, but less than that noted for the ionic components. Urban areas have the highest OC levels. The plots from the individual networks share more in common than the corresponding EC plots. The gradient tends from the south and east to the north, with highest values seen in the summer and fall. The seasonal relationship may indicate the influence of secondary organic aerosols and higher atmospheric oxidizing capacity in the summer months.

Soil

Separate figures for IMPROVE (Appendix Figure 6a) and STN (Appendix Figure 6b) soil extinction are shown. Soil contributes a very small percentage of the overall extinction budget so lower emphasis was placed on obtaining a consistent picture for the networks. IMPROVE soil shows seasonal and spatial variations, with the highest values observed in summer and spring. Spatially, the lowest extinction due to soil occurs in the western New England region. Unlike the other plots, extinction for soil in Maine is greater than the areas immediately west. The STN network shows a similar seasonal dependence with the highest values seen in urban areas.

Coarse

Coarse extinction is given in Appendix Figure 7. The greatest values are observed at Brigantine, which is consistent with historical observations and has been attributed to oceanic

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[17] The adjustment of 14% is made based on section II A3, where the collocated monitors at Addison Pinnacle showed STN nitrate = 1.14 IMPROVE nitrate. Since STN measures higher nitrate values, a correction was applied so the merged plot would represent a similar scale. Although it is unknown which network better measures nitrates, since the haze rule is based on IMPROVE measurements, IMPROVE values were determined as most appropriate.

influence. Otherwise, the distribution of coarse extinction closely resembles that of soil. The southwestern and northeastern corners yield the highest values with the central region having the lowest.

IV. Conclusions

The recent expansion of the IMPROVE network, combined with the development of the STN, have significantly increased the spatial density of speciated particulate measurements in the MANE-VU region and throughout the country. These data provide fundamental information regarding the composition of particulate matter and will be instrumental in aiding states as they prepare Regional Haze and $PM_{2.5}$ State Implementation Plans (SIPs). The IMPROVE network provides details on rural aerosol and will serve as the basis for regional visibility characterization. STN supplies complementary information regarding urban aerosol composition where the greatest potential for exceeding the fine particulate standard exists.

Individually, these two networks give a consistent regional picture of the fine particulate burden. Preliminary attempts to take advantage of the measurements from both of these networks show promise, as demonstrated in this memorandum. In the broadest sense, visibility in the MANE-VU region is most impaired to the southwest, improving toward the northeast. Superimposed on this general trend are the excess contributions from urban centers, primarily due to carbonaceous species and nitrate. Although this conceptual picture is consistent with prior analyses and current understanding of fine particulate, many issues need resolution before the results of these two networks can be used simultaneously with a high level of confidence.

Currently, only sulfate measurements may be combined outright. The union of nitrate measurements is more problematic than sulfate. Despite the high degree of correspondence evidenced by the comparison at collocated monitors, the STN results consistently predict higher nitrate concentrations than does IMPROVE. The comparison of carbon measurements between the two networks presents numerous complexities and requires further scientific inquiry before the results of these networks can be reliably considered together. The methodologies for comparison of EC/OC used in this memo are simplistic and should be viewed only as an initial best guess. The final component measured, soil, is a composite of five individual elements. Due to its small contribution to both fine mass and aerosol extinction, understanding the discrepancies between the soil results of the two networks is less pressing, and should receive lower priority than understanding the carbon differences.

The USEPA recognizes the importance of understanding the differences between IMPROVE and STN. Additionally, they realize certain problems exist and questions remain unanswered for particulate collection and characterization in general. Plans exist for an STN/IMPROVE method intercomparison for carbon analyses of identical quartz filters. Additional effort will address differences in shipping protocols. Further studies of filter blank corrections, focused on carbon, are expected. As of March 2003, it is anticipated that the STN will be adapted to IMPROVE protocol methods.[18] However, as demonstrated in this document, many uncertainties exist in the measurements for both networks. A thorough evaluation should help to identify the successes and deficiencies, with the resultant knowledge being applied to strengthen the measurements nationwide.

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[18] Remarks given by Joann Rice for Rich Sheffe at U.S. EPA sponsored OC/EC workshop in Durango, Colorado on March 4 and 5 2003. Presentation is available online at: http://ocs.fortlewis.edu/aerosols/ocec/scheffe.pdf

This memorandum provides a first look at region-wide speciated fine particulate measurements using the newly established STN and recently expanded IMPROVE networks. The combined networks afford high-density coverage, previously unavailable, and will significantly augment the current understanding of local and regional fine aerosol composition. With the increased availability of data, states will be able to more effectively address fine particle pollution and protect the health and welfare of their citizenry.

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Appendix A
B_{ext} Data Tables

Table A1. Annual Average b_{ext} values for IMPROVE and STN sites. The number (n) of samples for each average is given. EC and OC are adjusted from the measured STN values by allocating 40% of the OCX2 fraction to EC and the remaining 60% to OC. Total extinction is the sum of the six aerosol fractions in the table plus extinction due to Rayleigh Scattering (10 Mm^{-1}). Total fine extinction is the sum of the five fine aerosol fractions. AdjNO$_3$ represents STN NO$_3$ reduced by 14% (see footnote 17) comparison to IMPROVE NO$_3$.

LOCATION	n	Coarse	Soil	EC	OC	SO$_4$	NO$_3$	Total	Total Fine	NW	AdjNO$_3$
Washington D.C.	118	3.2	0.7	7.9	13.8	55.0	13.3	103.9	90.7	IMP	NV
Dover	57	NV	0.3	9.4	12.6	56.7	21.3	NV	100.4	STN	18.7
MLK Wilmington	57	NV	0.6	13.9	17.5	65.1	28.0	NV	124.2	STN	24.6
Cape Cod	117	NV	NV	NV	NV	33.6	NV	NV	NV	IMP	NV
Chicopee	64	NV	NV	6.5	8.7	NV	NV	NV	NV	STN	NV
Quabbin Summit	114	1.8	0.3	3.5	7.4	32.5	7.1	62.7	50.8	IMP	NV
Roxbury	62	NV	0.5	NV	NV	35.6	NV	NV	NV	STN	NV
Essex	118	NV	0.6	13.8	17.1	51.3	19.8	NV	101.9	STN	17.3
Acadia National Park	121	1.2	0.3	2.1	4.8	27.6	4.5	50.4	39.3	IMP	NV
Bridgton	114	1.2	0.3	3.0	6.7	28.4	3.3	52.9	41.6	IMP	NV
Casco Bay	119	1.5	0.4	4.0	7.9	27.8	4.9	56.4	44.9	IMP	NV
Moosehorn NWR	109	1.4	0.2	2.4	5.8	24.5	3.4	48.1	36.7	IMP	NV
Presque Isle	121	3.7	0.6	3.3	8.1	22.6	3.1	51.4	37.7	IMP	NV
Great Gulf Wilderness	117	NV	0.3	2.1	5.2	27.0	2.4	NV	36.9	IMP	NV
Brigantine National Wildlife Refuge	101	5.9	0.5	4.4	9.6	53.6	9.4	93.3	77.4	IMP	NV
Camden	85	NV	0.5	11.4	13.1	50.8	23.1	NV	97.7	STN	20.2
Chester	93	NV	0.3	7.6	8.8	47.1	15.9	NV	79.7	STN	14.0
Elizabeth	92	NV	0.7	23.4	19.4	48.5	23.6	NV	114.3	STN	20.7
New Brunswick	96	NV	0.4	10.3	10.2	46.6	16.2	NV	84.8	STN	14.2
Addison Pinnacle	119	1.5	0.4	3.3	6.3	47.2	8.8	77.5	66.0	IMP	NV
Addison Pinnacle	105	NV	0.3	4.6	5.3	48.3	11.5	NV	70.6	STN	10.1
Botanical Gardens	106	NV	0.6	19.2	15.0	48.2	22.0	NV	101.8	STN	19.3
Connecticut Hill	119	1.4	0.4	3.1	6.1	43.9	9.6	74.4	63.0	IMP	NV
Queens College	105	NV	0.6	11.2	11.4	50.3	22.3	NV	97.0	STN	19.5
Rochester	108	NV	0.4	6.4	7.0	45.6	22.8	NV	83.3	STN	20.0
Whiteface	105	NV	0.2	3.4	3.6	31.6	5.7	NV	44.9	STN	5.0
Quaker City	118	2.3	0.4	4.4	8.6	56.7	8.5	90.9	78.6	IMP	NV
Arendtsville	120	2.8	0.5	4.7	9.4	54.1	14.8	96.3	83.5	IMP	NV
Lawrenceville	109	NV	1.0	13.0	14.8	77.5	19.4	NV	125.9	STN	17.0
M.K. Goddard	121	2.1	0.4	5.1	9.4	52.5	13.7	93.3	81.2	IMP	NV
James River Face	114	2.3	0.5	5.4	11.8	52.7	6.4	89.6	77.3	IMP	NV
Shenandoah	109	1.9	0.3	3.6	8.1	54.4	7.8	86.0	74.2	IMP	NV
Burlington	115	NV	0.3	7.6	8.9	29.4	14.6	NV	60.9	STN	12.8
Lye Brook Wilderness	107	1.0	0.3	2.3	4.9	31.3	5.4	55.2	44.2	IMP	NV
Proctor Maple R. F.	113	1.4	0.2	2.6	6.1	28.3	6.0	54.6	43.2	IMP	NV
Dollysods	117	1.3	0.4	3.2	8.2	55.0	4.4	82.5	71.2	IMP	NV

Table A2 Summer Average b_{ext} values for IMPROVE and STN sites. The number (n) of samples for each average is given.

LOCATION	n	Coarse	Soil	EC	OC	SO_4	NO_3	Total	Total Fine	NW	AdjNO_3
McMillan Reservoir	24	NV	0.6	13.6	19.2	101.0	9.9	NV	144.3	STN	8.6
Washington D.C.	27	2.8	0.7	10.3	15.0	98.1	7.4	144.4	131.5	IMP	NV
Dover	13	NV	0.4	11.1	17.9	103.2	11.4	NV	143.9	STN	10.0
MLK Wilmington	15	NV	0.5	14.2	20.4	101.8	19.4	NV	156.3	STN	17.0
Cape Cod	28	3.6	0.6	3.1	7.4	55.3	4.1	84.2	70.6	IMP	NV
Chicopee	21	NV	0.2	6.4	10.6	45.2	4.9	NV	67.7	STN	4.3
Quabbin Summit	28	2.1	0.3	3.4	7.7	42.5	2.9	68.8	56.8	IMP	NV
Roxbury	5	NV	0.5	NV	NV	49.1	NV	NV	NV	STN	NV
Essex	24	NV	0.6	13.7	18.2	101.0	12.4	NV	144.1	STN	10.9
Acadia National Park	30	1.2	0.4	2.7	7.3	42.8	2.5	66.9	55.7	IMP	NV
Bridgton	30	1.8	0.5	3.3	8.4	44.0	1.9	69.9	58.1	IMP	NV
Casco Bay	29	2.0	0.5	5.0	10.0	48.2	3.8	79.6	67.5	IMP	NV
Moosehorn NWR	22	2.0	0.3	2.2	9.0	39.3	2.1	64.8	52.9	IMP	NV
Presque Isle	30	4.2	0.5	3.5	11.7	30.0	1.4	61.4	47.2	IMP	NV
Great Gulf Wilderness	28	NV	0.5	2.6	8.5	43.4	1.0	NV	56.1	IMP	NV
Brigantine National Wildlife Refuge	22	7.3	0.8	5.2	13.4	103.0	4.5	144.2	126.8	IMP	NV
Camden	14	NV	0.4	11.1	16.5	88.2	16.7	NV	133.6	STN	14.7
Chester	22	NV	0.3	8.7	12.8	74.1	8.6	NV	104.5	STN	7.6
Elizabeth	20	NV	0.6	23.8	20.4	70.4	16.9	NV	127.3	STN	14.8
New Brunswick	26	NV	0.4	11.1	14.0	73.9	10.8	NV	110.1	STN	9.4
Addison Pinnacle	30	1.6	0.7	4.2	8.5	82.6	3.0	110.5	98.8	IMP	NV
Addison Pinnacle	24	NV	0.3	5.3	7.5	86.5	4.2	NV	104.5	STN	3.6
Botanical Gardens	20	NV	0.6	16.4	17.5	72.2	13.8	NV	111.5	STN	12.1
Connecticut Hill	28	1.5	0.6	3.7	8.0	71.7	3.2	98.8	87.2	IMP	NV
Queens College	27	NV	0.5	11.8	14.6	77.4	17.3	NV	123.2	STN	15.2
Rochester	28	NV	0.5	7.9	9.7	70.2	9.6	NV	99.9	STN	8.5
Whiteface	23	NV	0.2	5.1	7.1	56.4	1.3	NV	70.1	STN	1.2
Quaker City	30	3.1	0.5	5.3	10.1	92.4	3.5	124.9	111.8	IMP	NV
Arendtsville	30		0.7	4.9	10.7	85.2	7.7	122.2	109.3	IMP	NV
Florence	35	NV	0.7	9.9	13.6	136.8	4.7	NV	165.6	STN	4.1
Greensburg	34	NV	0.4	12.1	14.8	126.3	6.3	NV	161.4	STN	5.6
Lawrenceville	35	NV	1.9	13.7	17.0	135.6	8.4	NV	176.7	STN	7.4
M.K. Goddard	30	2.5	0.4	6.6	11.3	86.4	5.9	123.1	110.6	IMP	NV
James River Face	25	2.9	0.7	5.5	12.2	89.1	4.2	124.6	111.7	IMP	NV
Shenandoah	28	1.7	0.4	3.8	8.1	95.0	4.5	123.4	111.7	IMP	NV
Burlington	30	NV	0.5	8.9	12.7	45.8	5.5	NV	73.4	STN	4.8
Lye Brook Wilderness	22	1.4	0.6	3.2	8.6	67.4	2.0	93.1	81.8	IMP	NV
Proctor Maple R. F.	28	1.6	0.2	2.7	7.6	39.2	1.3	62.7	51.1	IMP	NV
Dollysods	26	1.3	0.7	3.1	7.2	108.1	1.7	132.1	120.8	IMP	NV

Table A3. Fall Average b_{ext} values for IMPROVE and STN sites. The number (n) of samples for each average is given.

LOCATION	n	Coarse	Soil	EC	OC	SO_4	NO_3	Total	Total Fine	NW	AdjNO_3
Mohawk Mt.	22	2.0	0.3	3.9	8.8	31.4	8.5	64.9	52.9	IMP	NV
McMillan Reservoir	24	NV	0.6	15.6	18.1	47.8	16.6	NV	100.0	STN	14.6
Washington D.C.	31	3.6	0.6	7.9	14.2	43.5	12.9	92.6	79.1	IMP	NV
Dover	15	NV	0.3	9.9	14.0	51.4	18.8	NV	94.3	STN	16.5
MLK Wilmington	15	NV	0.7	17.2	21.3	67.3	28.3	NV	134.9	STN	24.8
Cape Cod	30	2.9	0.2	2.5	5.7	32.0	5.5	58.7	45.8	IMP	NV
Chicopee	18	NV	0.3	7.0	8.3	36.6	9.9	NV	58.2	STN	8.6
Quabbin Summit	28	1.9	0.3	3.8	8.4	36.9	7.9	69.1	57.2	IMP	NV
Roxbury	23	NV	0.5	19.4	21.9	39.8	14.9	NV	97.9	STN	13.1
Essex	23	NV	0.6	14.8	17.3	45.5	16.3	NV	94.5	STN	14.3
Fort Meade	9	NV	0.4	13.4	19.7	43.1	12.4	NV	88.0	STN	10.9
Acadia National Park	31	1.3	0.2	1.9	4.4	25.4	4.9	48.2	36.9	IMP	NV
Bridgton	31	1.3	0.2	3.0	6.2	27.0	3.1	50.9	39.6	IMP	NV
Casco Bay	31	1.6	0.2	4.1	6.8	26.7	5.0	54.3	42.7	IMP	NV
Moosehorn NWR	29	1.4	0.2	3.2	5.8	25.9	3.7	51.3	39.8	IMP	NV
Old Town	29	2.5	0.3	4.2	10.0	33.0	4.5	64.5	52.0	IMP	NV
Presque Isle	31	3.1	0.4	3.2	7.3	26.8	3.2	54.0	40.8	IMP	NV
Great Gulf Wilderness	30	1.7	0.2	2.2	5.0	28.6	2.2	49.8	38.1	IMP	NV
Portsmouth	20	NV	0.4	9.0	11.2	34.3	9.5	NV	64.9	STN	8.4
Brigantine National Wildlife Refuge	30	5.7	0.4	4.0	8.7	42.6	7.2	78.7	63.0	IMP	NV
Camden	23	NV	0.5	13.1	16.3	51.3	19.6	NV	100.7	STN	17.2
Chester	22	NV	0.3	7.7	8.8	44.2	15.7	NV	76.6	STN	13.7
Elizabeth	23	NV	0.7	28.1	21.9	46.7	22.8	NV	120.2	STN	20.0
New Brunswick	22	NV	0.5	12.1	11.2	44.5	14.4	NV	84.4	STN	12.7
Addison Pinnacle	31	1.7	0.3	3.5	6.5	42.3	7.0	71.3	59.6	IMP	NV
Addison Pinnacle	24	NV	0.2	4.9	5.9	38.2	8.3	NV	59.8	STN	7.3
Botanical Gardens	27	NV	0.6	20.9	17.1	43.1	21.2	NV	103.0	STN	18.6
Connecticut Hill	31	1.6	0.3	3.3	6.3	41.2	7.6	70.3	58.7	IMP	NV
IS 52	25	NV	0.7	17.8	15.7	45.3	23.1	NV	102.7	STN	20.3
Queens College	23	NV	0.7	12.9	11.8	43.5	20.1	NV	91.0	STN	17.6
Rochester	27	NV	0.4	6.5	6.9	42.4	17.5	NV	75.3	STN	15.4
Whiteface	27	NV	0.1	3.7	3.8	30.8	5.4	NV	44.9	STN	4.8
Quaker City	31	2.6	0.4	4.8	9.5	54.9	5.2	87.4	74.8	IMP	NV
Arendtsville	30	3.3	0.4	5.0	10.8	50.7	14.1	94.4	81.0	IMP	NV
Florence	14	NV	0.5	9.0	11.0	49.1	6.6	NV	76.2	STN	5.8
Greensburg	13	NV	0.4	14.0	13.0	47.3	12.2	NV	87.0	STN	10.7
Lawrenceville	24	NV	0.6	16.0	18.3	60.1	15.2	NV	110.3	STN	13.3
M.K. Goddard	31	2.3	0.4	5.5	10.7	53.3	10.6	92.8	80.5	IMP	NV
Philadelphia	28	NV	0.5	15.8	18.0	42.0	20.9	NV	97.3	STN	18.4
James River Face	31	2.6	0.4	6.1	12.8	51.7	5.4	89.0	76.4	IMP	NV
Shenandoah	31	2.3	0.3	4.2	9.7	50.5	7.7	84.7	72.4	IMP	NV
Burlington	28	NV	0.3	7.5	7.8	24.4	12.1	NV	51.9	STN	10.7
Lye Brook Wilderness	27	1.0	0.2	2.4	5.0	30.9	5.1	54.5	43.5	IMP	NV
Proctor Maple R. F.	31	1.7	0.2	2.8	6.3	28.9	4.5	54.4	42.7	IMP	NV
Dollysods	31	1.5	0.3	3.5	9.9	48.2	3.9	77.4	65.8	IMP	NV

Table A4. Winter Average b_{ext} values for IMPROVE and STN sites. The number (n) of samples for each average is given.

LOCATION	n	Coarse	Soil	EC	OC	SO_4	NO_3	Total	Total Fine	NW	AdjNO_3
Mohawk Mt.	29	1.33	0.30	3.47	6.29	22.88	10.85	55.12	43.78	IMP	NV
McMillan Reservoir	18	NV	0.45	12.69	16.31	34.19	25.92	NV	89.08	STN	22.74
Washington D.C.	30	3.17	0.57	7.17	15.35	31.06	21.88	89.21	76.04	IMP	NV
Dover	14	NV	0.22	10.16	13.82	30.94	36.31		91.46	STN	31.85
MLK Wilmington	12	NV	0.48	14.25	17.98	33.60	41.10	NV	101.46	STN	36.05
Cape Cod	29	2.16	0.17	2.58	6.12	19.01	8.45	48.50	36.34	IMP	NV
Chicopee	20	NV	0.27	7.27	10.12	27.92	14.72	NV	61.40	STN	12.91
Quabbin Summit	28	1.48	0.23	3.82	8.06	21.53	11.44	56.57	45.09	IMP	NV
Roxbury	19	NV	0.41	16.96	21.13	27.11	22.23	NV	87.83	STN	19.50
Essex	51	NV	0.52	14.29	19.25	30.13	25.89	NV	90.08	STN	22.71
Fort Meade	45	NV	0.32	8.32	11.00	27.32	16.50	NV	63.45	STN	14.48
Acadia National Park	30	0.96	0.15	2.09	4.15	18.80	6.37	42.53	31.56	IMP	NV
Bridgton	29	0.82	0.15	3.12	6.95	17.47	5.45	43.96	33.14	IMP	NV
Casco Bay	29	0.94	0.16	4.61	10.05	18.51	6.55	50.82	39.88	IMP	NV
Moosehorn NWR	28	0.97	0.12	2.86	5.40	15.90	4.68	39.62	28.65	IMP	NV
Old Town	22	3.51	0.53	5.17	11.15	26.40	6.51	63.28	49.77	IMP	NV
Presque Isle	30	3.29	0.57	3.86	8.32	15.65	5.53	47.21	33.93	IMP	NV
Great Gulf Wilderness	29	0.83	0.11	1.99	4.03	14.35	4.55	35.85	25.02	IMP	NV
Portsmouth	22	NV	0.39	11.50	14.19	25.76	13.87	NV	66.24	STN	12.17
Brigantine National Wildlife Refuge	21	2.84	0.31	5.50	9.93	30.13	19.98	78.68	65.85	IMP	NV
Camden	23	NV	0.39	12.85	14.25	32.57	34.33	NV	94.39	STN	30.11
Chester	24	NV	0.22	6.47	6.87	26.03	21.80	NV	61.39	STN	19.13
Elizabeth	24	NV	0.56	21.25	21.43	32.58	29.49	NV	105.30	STN	25.86
New Brunswick	24	NV	0.33	9.47	9.33	28.87	24.71	NV	72.71	STN	21.67
Addison Pinnacle	30	1.29	0.25	2.92	5.21	26.48	15.20	61.36	50.07	IMP	NV
Addison Pinnacle	29	NV	0.21	4.04	4.08	29.17	19.57	NV	57.08	STN	17.17
Botanical Gardens	29	NV	0.57	21.08	15.45	34.27	30.22	NV	101.59	STN	26.51
Connecticut Hill	30	1.00	0.22	2.57	4.91	26.90	16.92	62.53	51.53	IMP	NV
IS 52	29	NV	0.58	15.00	13.22	36.75	31.08	NV	96.63	STN	27.26
Queens College	27	NV	0.55	10.90	11.82	36.21	30.95	NV	91.28	STN	27.15
Rochester	28	NV	0.36	5.50	6.25	29.09	40.87	NV	82.07	STN	35.85
Whiteface	29	NV	0.13	2.38	1.94	17.36	11.18	NV	33.01	STN	9.81
Quaker City	30	1.28	0.27	3.77	7.33	32.01	17.17	71.83	60.55	IMP	NV
Arendtsville	30	1.84	0.32	4.31	8.05	29.94	21.62	76.08	64.24	IMP	NV
Lawrenceville	21	NV	0.54	11.04	15.32	41.72	45.09	NV	113.98	STN	39.55
M.K. Goddard	30	1.60	0.31	4.24	8.31	33.07	26.76	84.28	72.68	IMP	NV
Philadelphia	27	NV	0.40	13.83	15.69	29.20	35.89	NV	95.01	STN	31.48
James River Face	29	1.67	0.35	5.03	11.51	29.94	10.01	69.52	57.83	IMP	NV
Shenandoah	22	1.25	0.20	2.96	7.96	24.45	11.85	58.67	47.42	IMP	NV
Burlington	29	NV	0.25	7.68	9.15	21.15	29.85	NV	68.07	STN	26.18
Lye Brook Wilderness	29	0.87	0.13	1.99	3.53	13.25	9.05	38.82	27.95	IMP	NV
Proctor Maple R. F.	30	0.92	0.15	2.77	5.48	17.26	13.44	50.02	39.10	IMP	NV
Dollysods	30	0.82	0.17	3.08	7.48	24.09	6.98	52.62	41.79	IMP	NV

Table A5. Spring Average b_{ext} values for IMPROVE and STN sites. The number (n) of samples for each average is given.

LOCATION	n	Coarse	Soil	EC	OC	SO_4	NO_3	Total	Total Fine	NW	AdjNO_3
Mohawk Mt.	27	2.4	0.4	2.4	5.0	28.4	5.8	54.4	42.0	IMP	NV
Washington D.C.	30	3.2	0.8	6.7	10.7	52.2	10.4	93.9	80.8	IMP	NV
Dover	15	NV	0.3	6.9	5.6	45.8	18.4		77.0	STN	16.1
MLK Wilmington	15	NV	0.6	10.1	10.6	53.3	25.1	NV	99.6	STN	22.0
Cape Cod	30	NV	NV	NV	NV	28.8	NV	NV	NV	IMP	NV
Chicopee	5	NV	NV	5.5	6.1	NV	NV	NV	NV	STN	NV
Quabbin Summit	30	1.8	0.4	3.1	5.6	29.4	6.2	56.5	44.7	IMP	NV
Roxbury	15	NV	0.7	15.6	16.3	32.0	10.9	NV	77.2	STN	9.5
Baltimore	55	NV	0.8	15.1	15.3	49.6	24.5	NV	105.2	STN	21.5
Essex	20	NV	0.7	11.3	9.9	49.6	17.5	NV	89.7	STN	15.3
Fort Meade	13	NV	0.4	7.8	7.5	49.3	10.6	NV	75.4	STN	9.3
Acadia National Park	30	1.2	0.3	1.6	3.3	23.6	4.0	44.2	32.9	IMP	NV
Bridgton	24	0.9	0.3	2.3	4.7	23.6	2.9	44.8	33.8	IMP	NV
Casco Bay	30	1.6	0.8	2.3	4.8	18.0	4.2	41.7	30.2	IMP	NV
Moosehorn NWR	30	1.2	0.3	1.5	4.0	20.6	2.9	40.6	29.4	IMP	NV
Old Town	21	4.0	0.5	3.9	6.0	25.7	4.0	54.2	40.2	IMP	NV
Presque Isle	30	4.4	0.8	2.5	5.0	18.0	2.3	42.9	28.6	IMP	NV
Great Gulf Wilderness	30	1.3	0.3	1.6	3.4	22.4	1.7	40.6	29.4	IMP	NV
Manchester	14	NV	0.5	8.1	6.6	25.9	9.9	NV	51.0	STN	8.7
Portsmouth	26	NV	0.4	7.2	6.7	29.6	7.9	NV	52.3	STN	6.9
Brigantine National Wildlife Refuge	28	7.3	0.5	3.2	7.4	44.0	7.7	80.1	62.8	IMP	NV
Camden	25	NV	0.5	8.5	7.2	44.7	19.5	NV	78.0	STN	17.1
Chester	25	NV	0.3	7.8	7.0	46.0	16.9	NV	78.0	STN	14.8
Elizabeth	25	NV	0.8	20.9	14.1	46.1	24.5	NV	107.2	STN	21.5
New Brunswick	24	NV	0.5	8.8	6.4	37.7	15.1	NV	69.8	STN	13.3
Addison Pinnacle	28	1.4	0.4	2.7	4.9	37.0	10.0	66.4	55.0	IMP	NV
Addison Pinnacle	28	NV	0.3	4.4	3.9	43.4	13.1	NV	65.0	STN	11.5
Botanical Gardens	30	NV	0.6	18.2	10.5	43.7	21.5	NV	94.4	STN	18.9
Buffalo	35	NV	0.5	8.9	12.5	45.4	15.5	NV	84.0	STN	13.6
Connecticut Hill	30	1.4	0.4	2.7	5.2	37.6	10.4	67.8	56.3	IMP	NV
IS 52	26	NV	0.7	13.1	9.6	45.0	23.2	NV	92.1	STN	20.3
Queens College	28	NV	0.6	9.5	7.5	43.2	20.2	NV	82.1	STN	17.8
Rochester	25	NV	0.4	5.4	5.1	39.6	22.9	NV	74.6	STN	20.1
Whiteface	26	NV	0.3	2.8	2.1	26.4	3.5	NV	35.7	STN	3.1
Quaker City	27	2.4	0.6	3.6	7.3	46.5	8.0	78.4	66.0	IMP	NV
Arendtsville	30	3.1	0.6	4.4	8.1	50.6	15.7	92.5	79.3	IMP	NV
Florence	9	NV	0.6	8.3	6.9	53.2	13.1	NV	81.9	STN	11.5
Greensburg	10	NV	1.2	10.9	9.1	42.6	12.3	NV	76.1	STN	10.8
Lawrenceville	29	NV	0.7	11.0	8.9	49.1	16.5	NV	86.1	STN	14.4
M.K. Goddard	30	2.0	0.5	4.1	7.4	37.3	11.7	73.1	61.1	IMP	NV
James River Face	29	2.1	0.5	5.1	10.7	46.0	5.7	80.1	68.1	IMP	NV
Shenandoah	28	2.0	0.5	3.1	6.4	41.7	8.0	71.6	59.6	IMP	NV
Burlington	28	NV	0.4	6.3	5.5	25.6	11.4	NV	49.1	STN	10.0
Lye Brook Wilderness	29	0.8	0.3	1.7	3.3	22.4	4.7	43.3	32.4	IMP	NV
Proctor Maple R. F.	24	1.3	0.3	2.1	4.7	28.7	4.1	51.2	39.9	IMP	NV
Dollysods	30	1.5	0.5	3.2	8.2	46.7	4.6	74.7	63.2	IMP	NV

Appendix B
Figures of average annual and seasonal b_{ext}

Figure 1a. Fine Aerosol Extinction Based on merged STN and IMPROVE Data

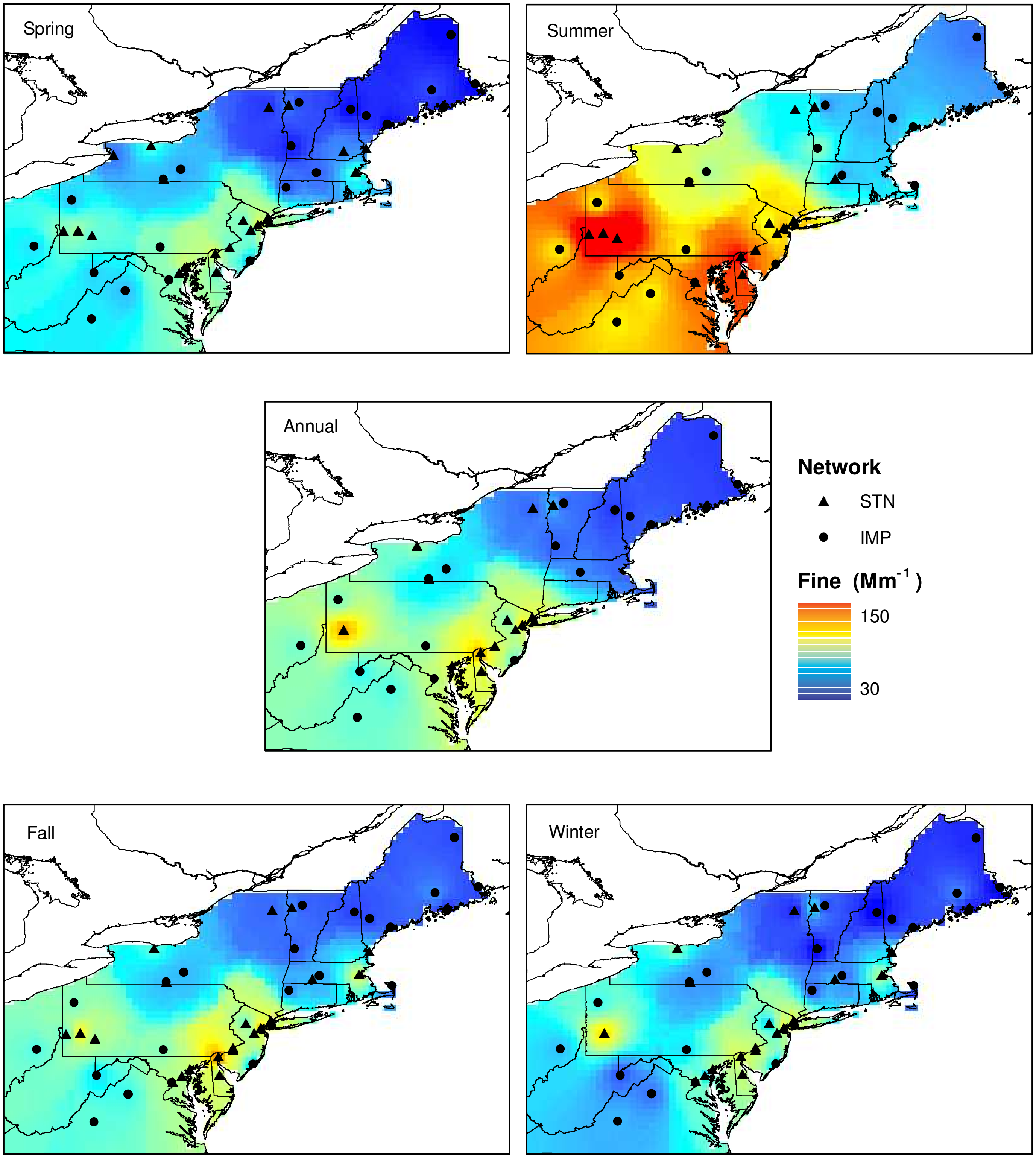

Figure 1b. Fine Aerosol Extinction Based on IMPROVE Data

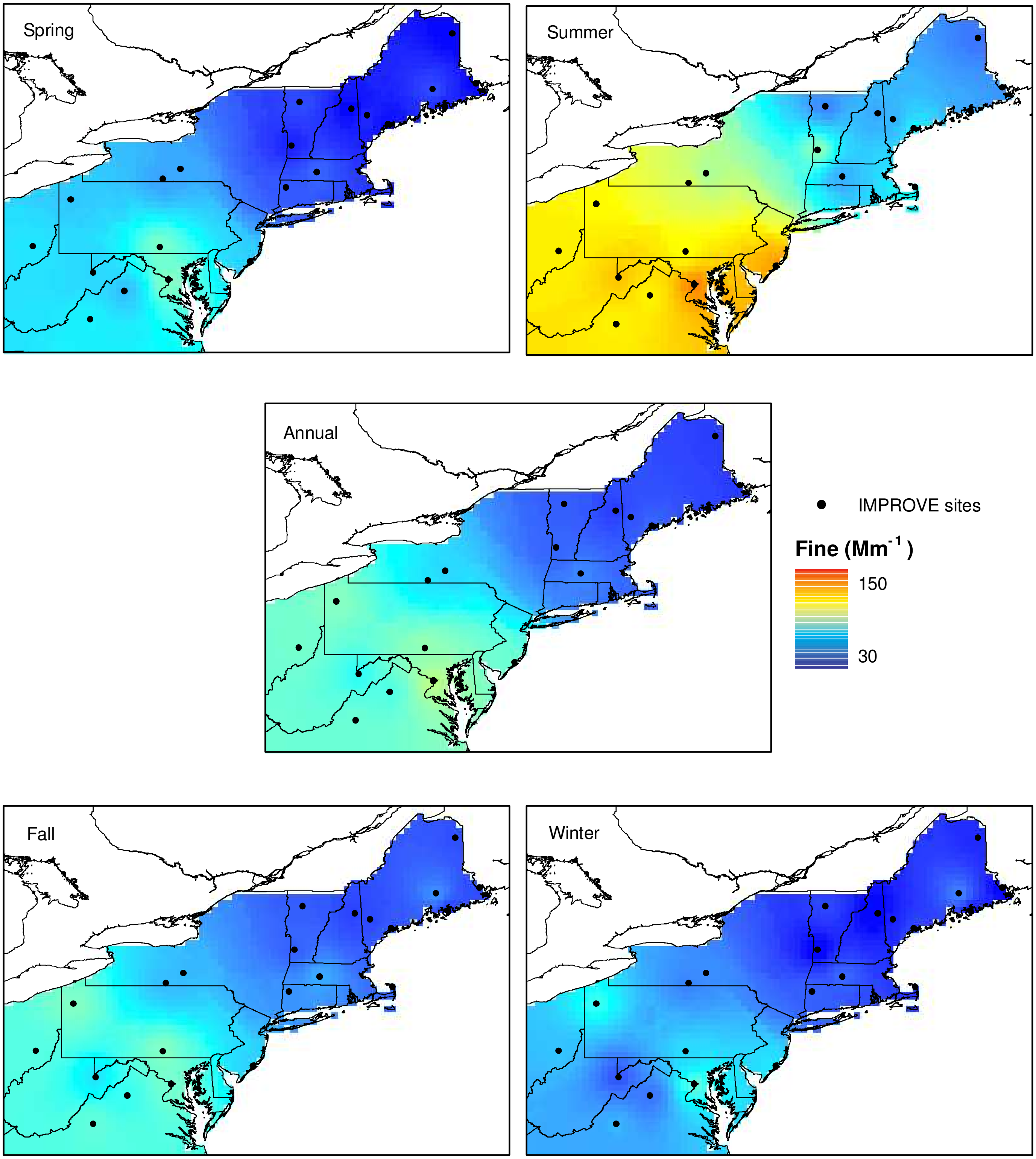

Figure 1c. Fine Aerosol Extinction Based on STN Data

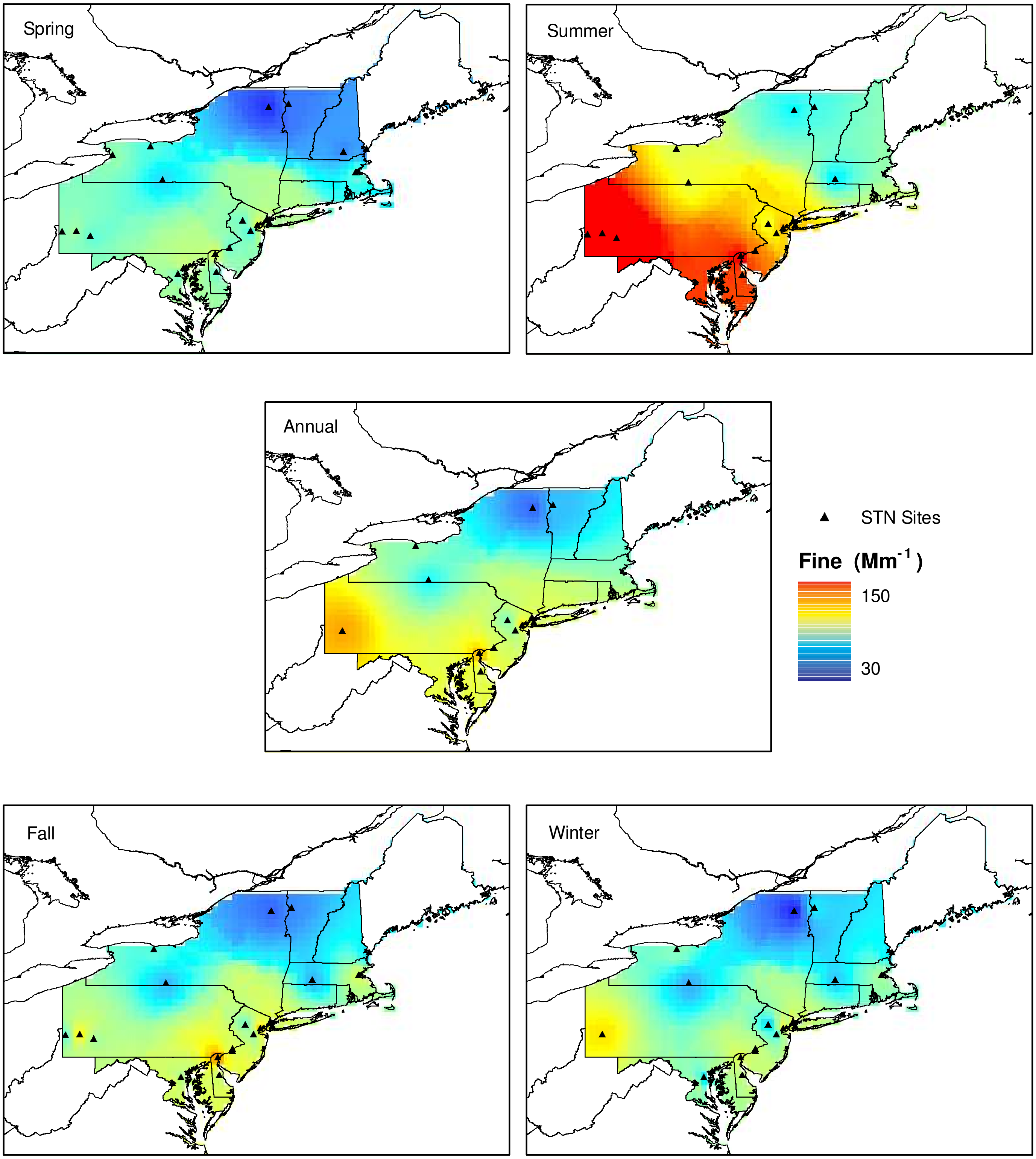

Figure 1d. Total Aerosol Extinction Based on IMPROVE Data

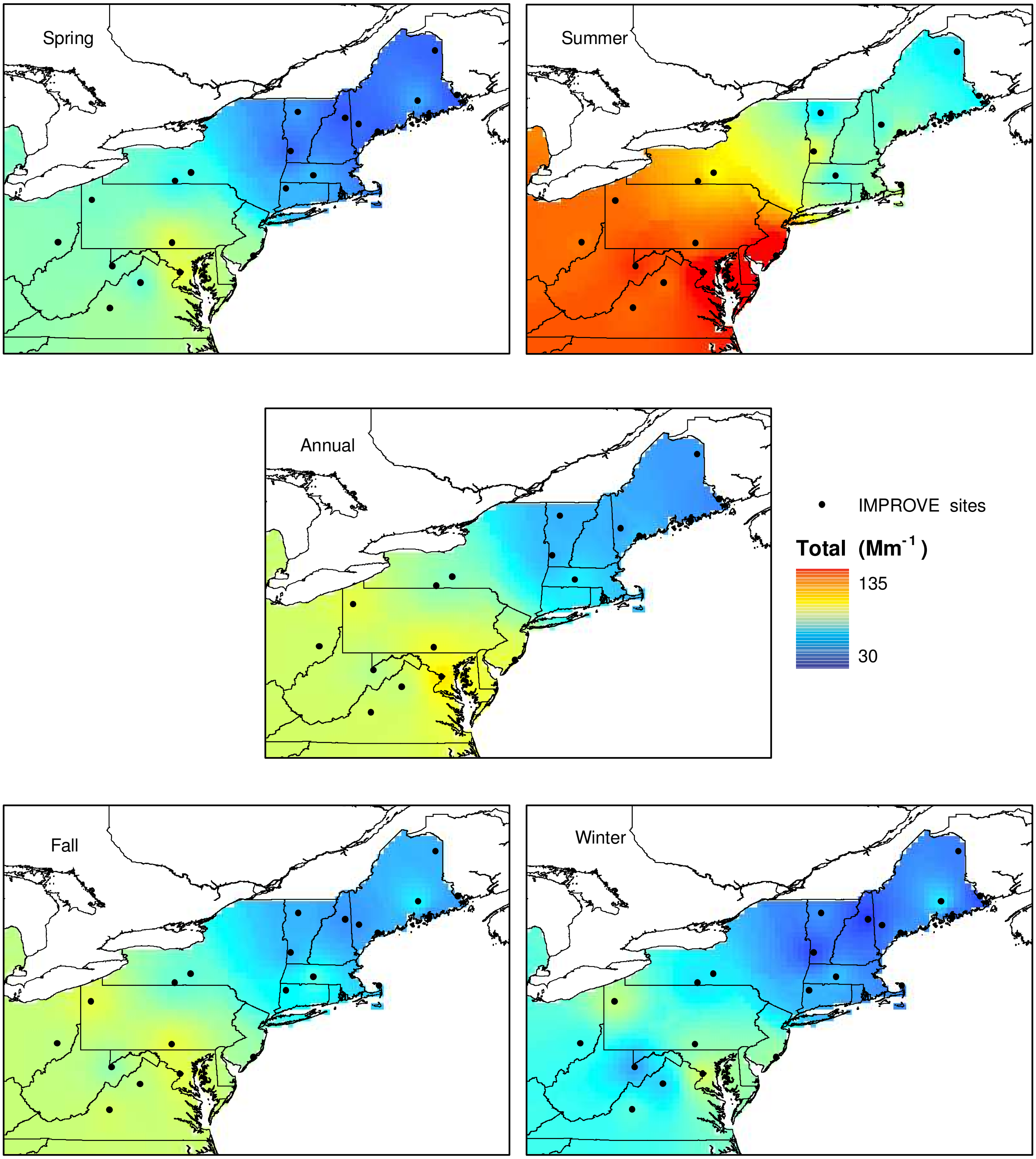

Figure 2a. Ammonium Sulfate Extinction Based on merged STN and IMPROVE Data

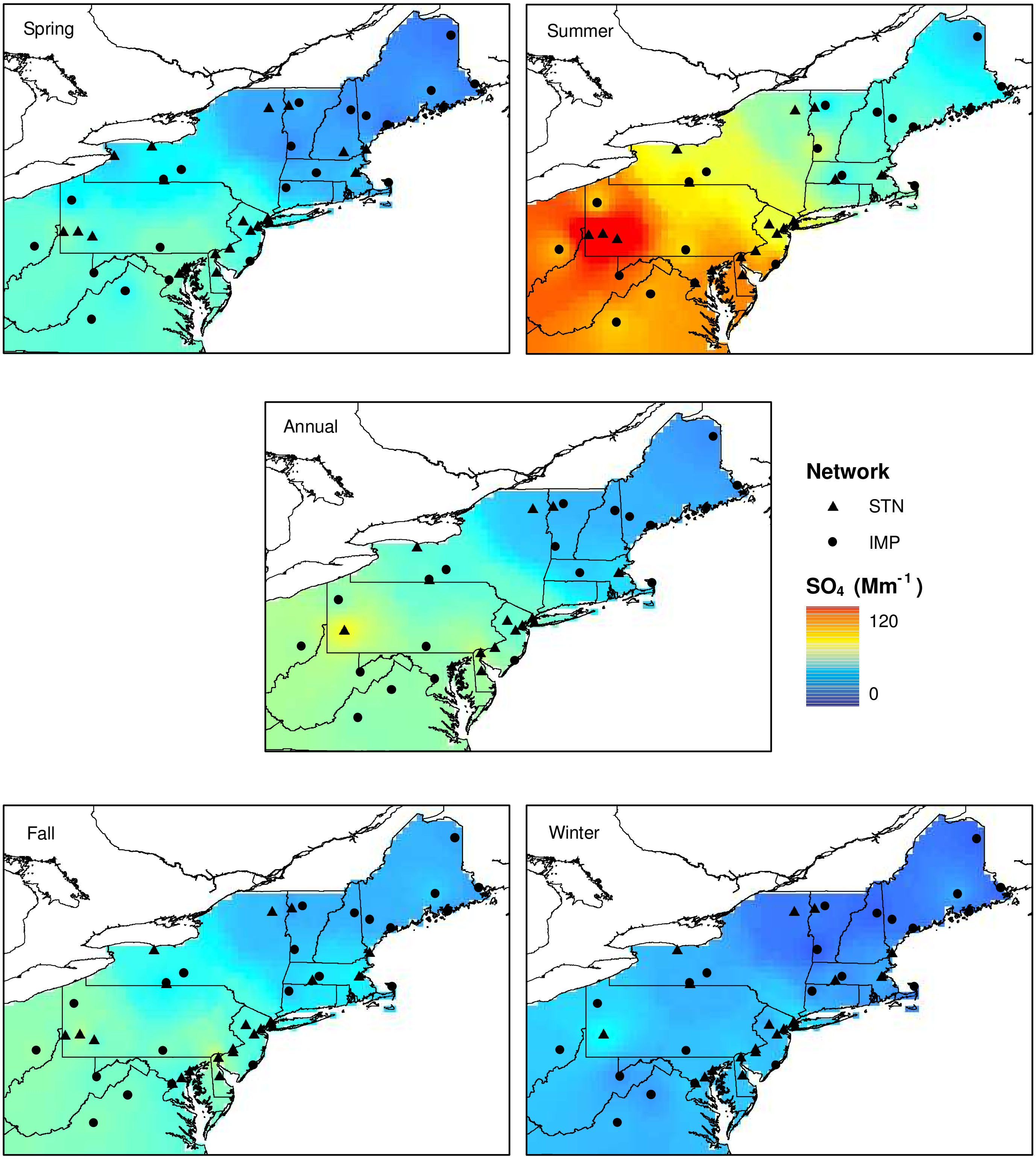

Figure 2b. Ammonium Sulfate Extinction Based on IMPROVE Data

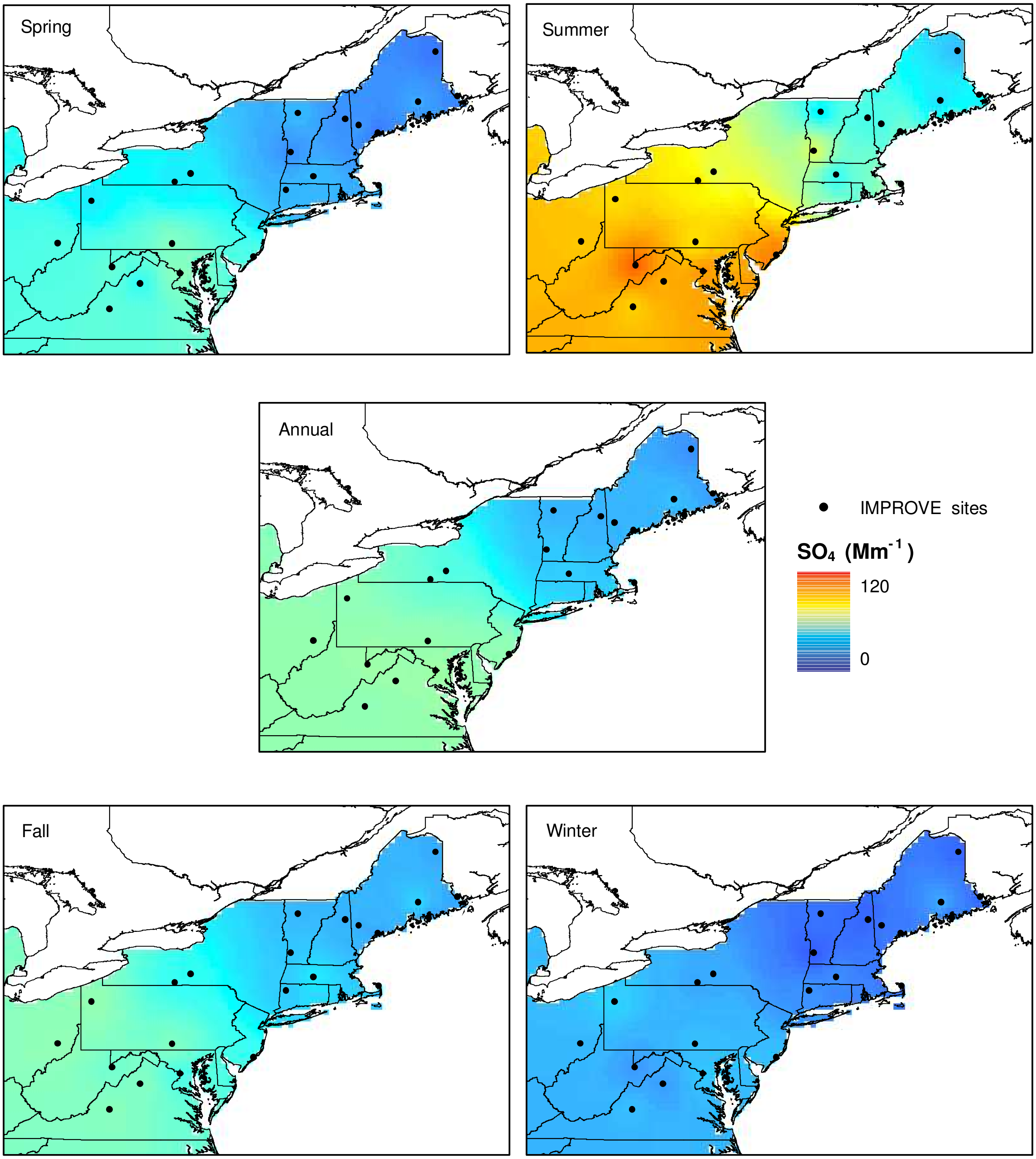

Figure 2c. Ammonium Sulfate Extinction Based on STN Data

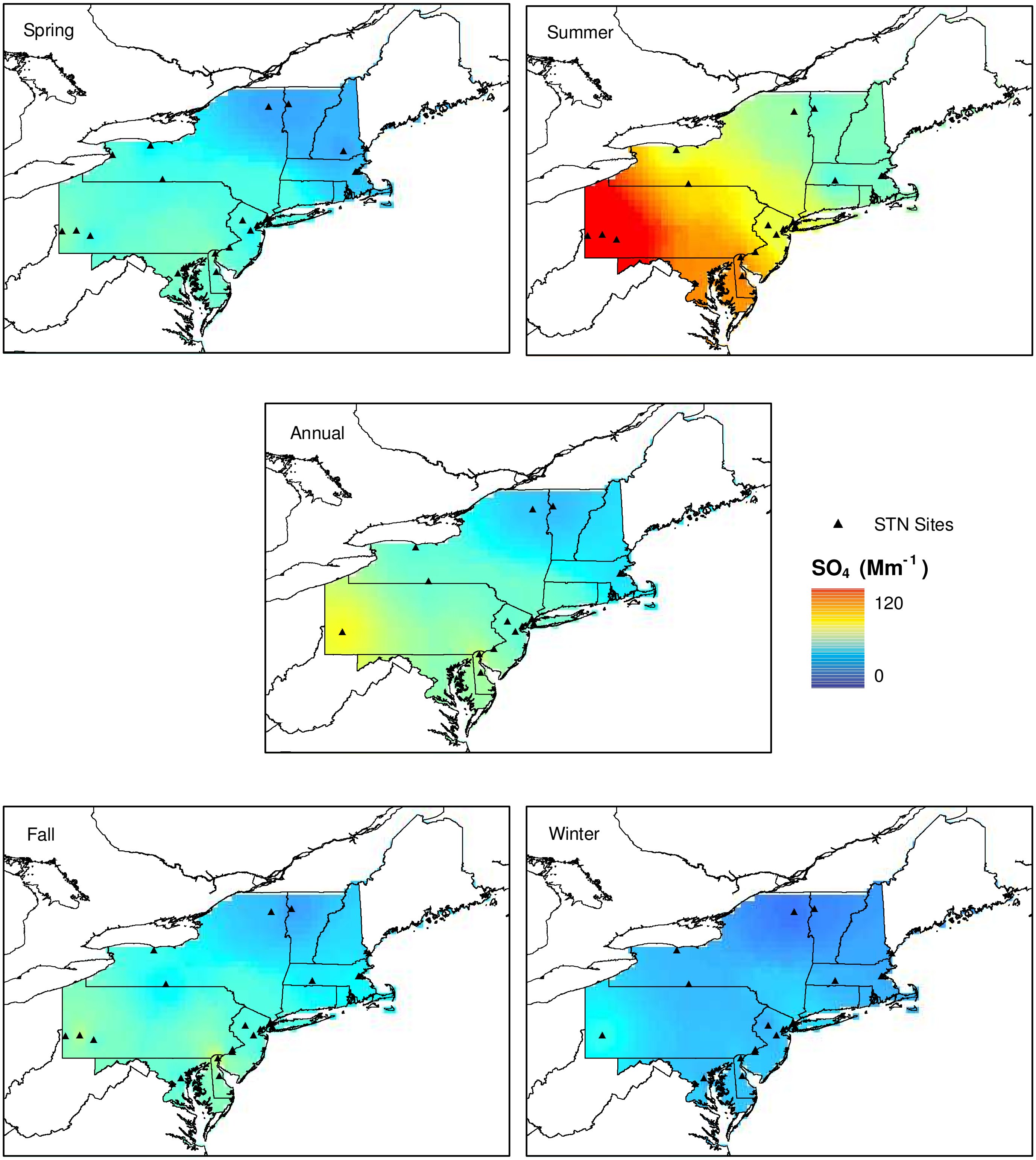

Figure 3a. Ammonium Nitrate Extinction Based on merged STN and IMPROVE Data

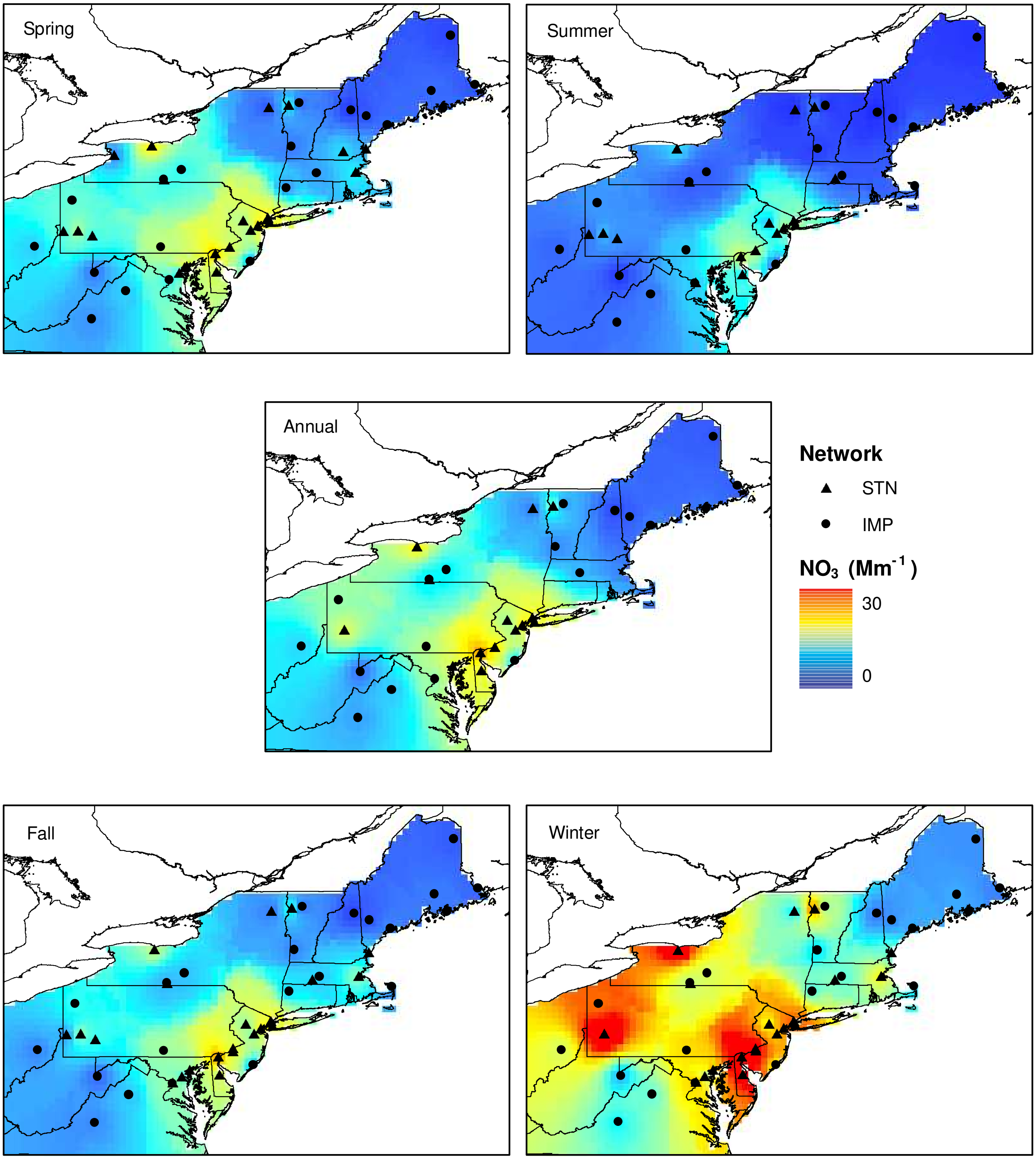

Figure 3b. Ammonium Nitrate Extinction Based on IMPROVE Data

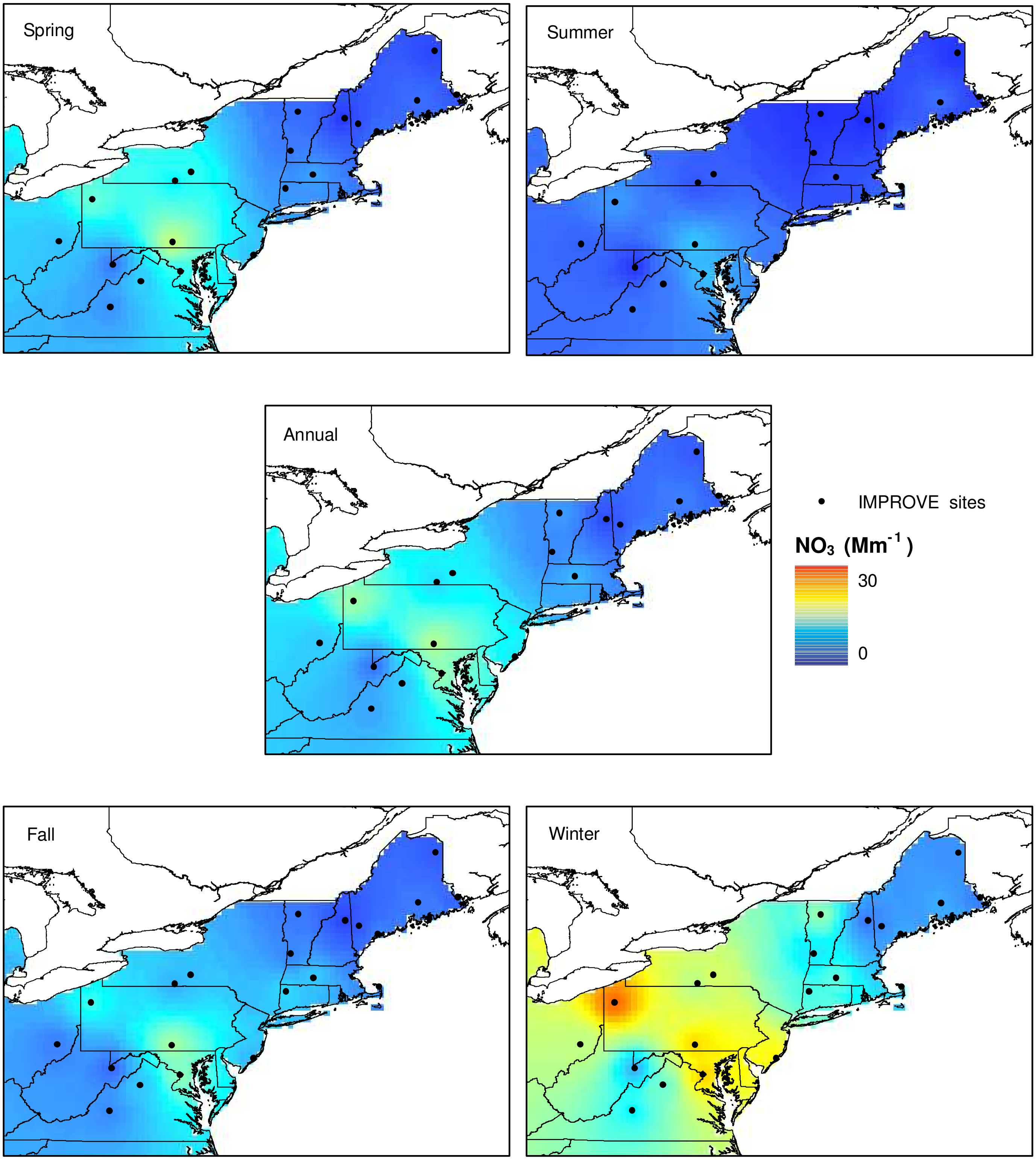

Figure 3c. Ammonium Nitrate Extinction Based on Adjusted STN Data

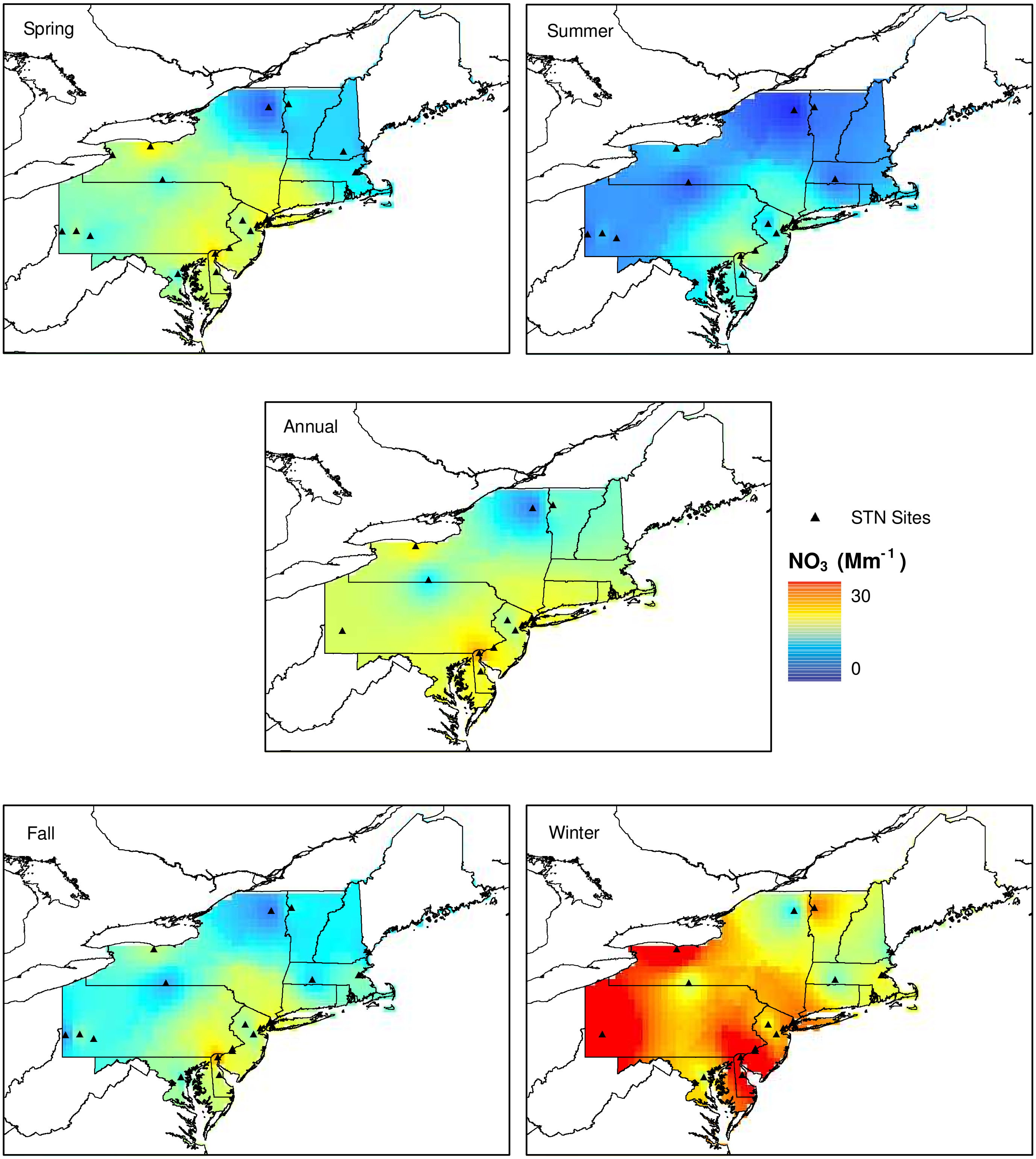

Figure 3d. Ammonium Nitrate Extinction Based on Unadjusted STN Data

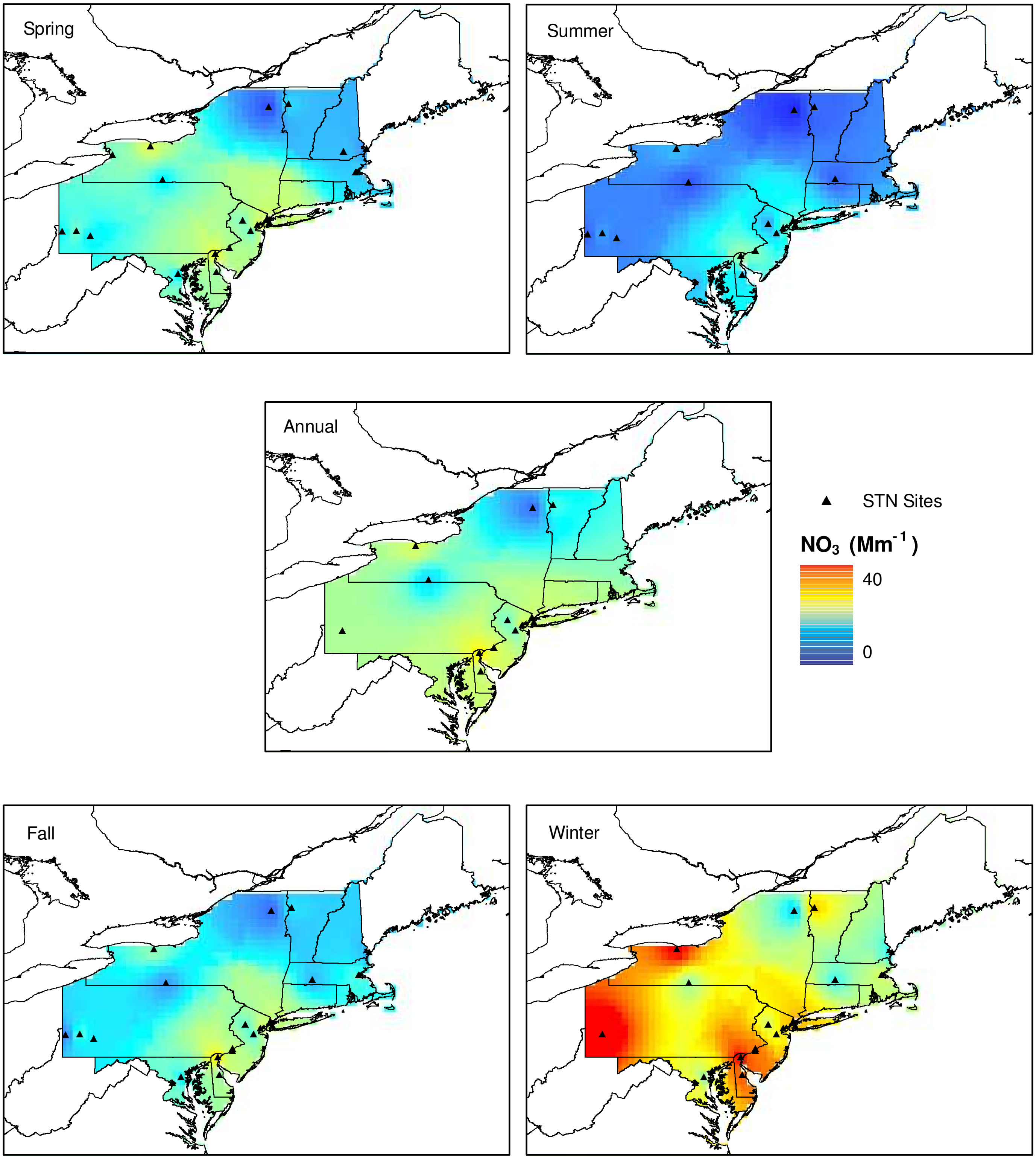

Figure 4a. Elemental Carbon Extinction Based on merged STN and IMPROVE Data

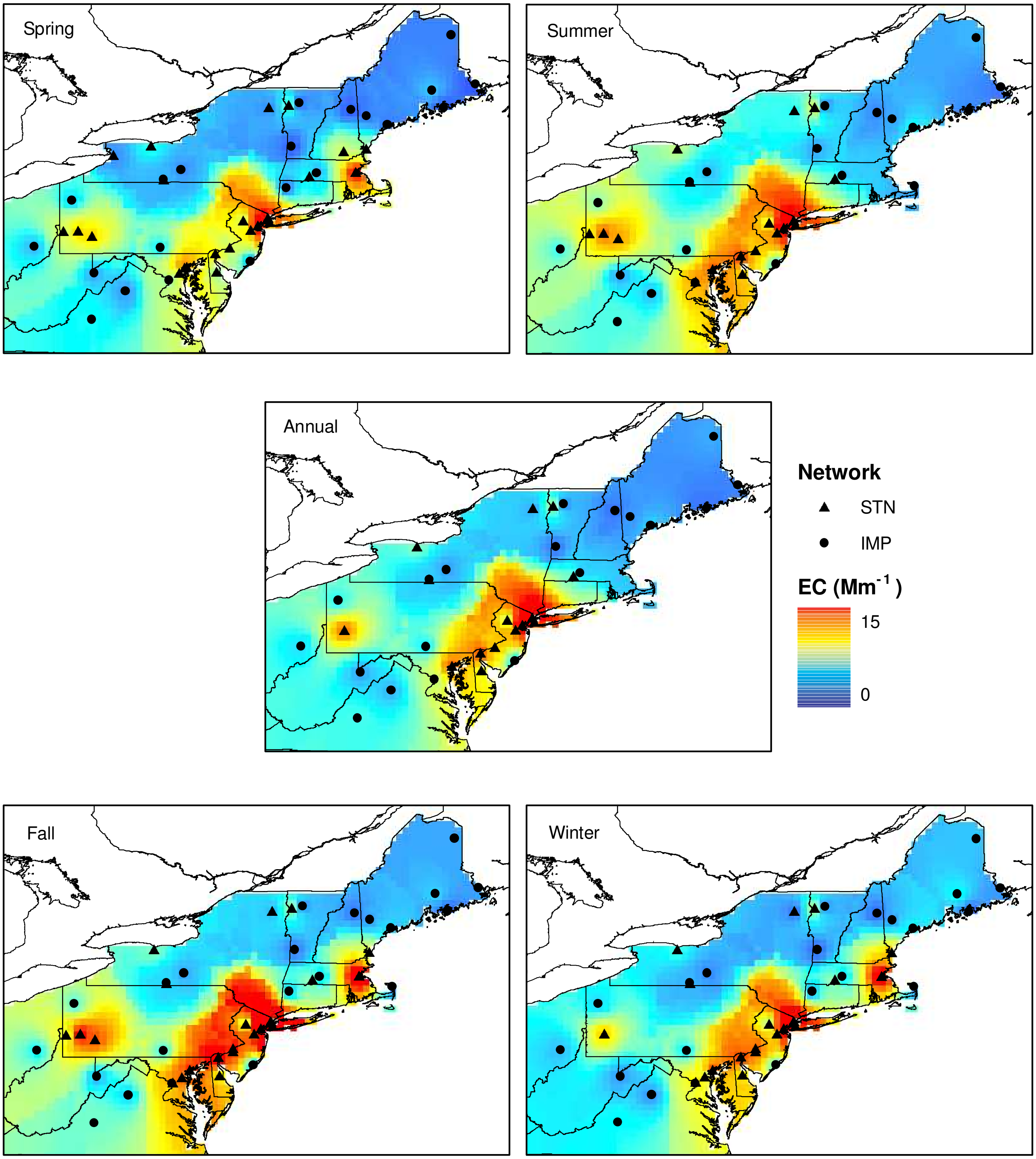

Figure 4b. Elemental Carbon Extinction Based on IMPROVE Data

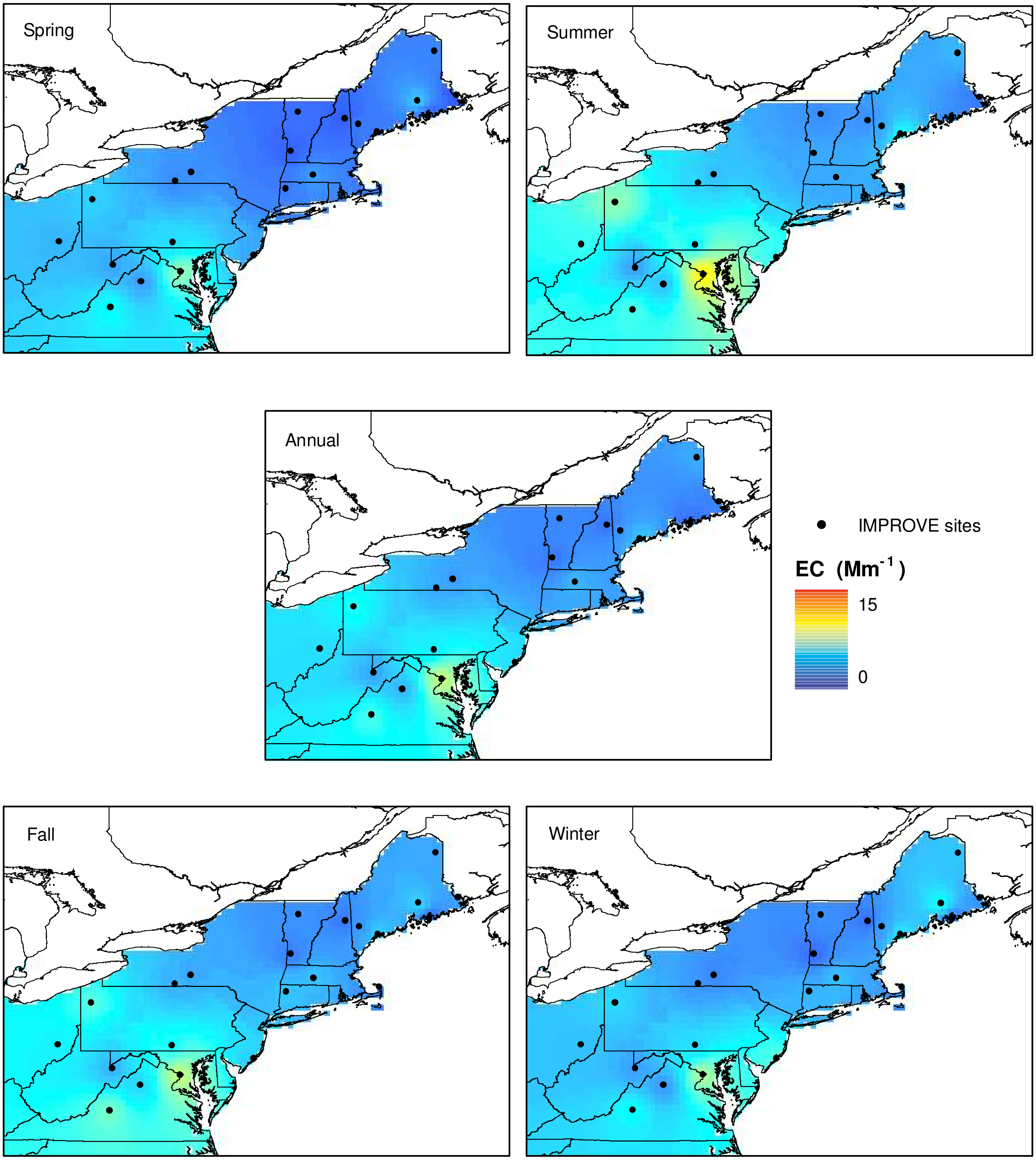

Figure 4c. Elemental Carbon Extinction Based on STN

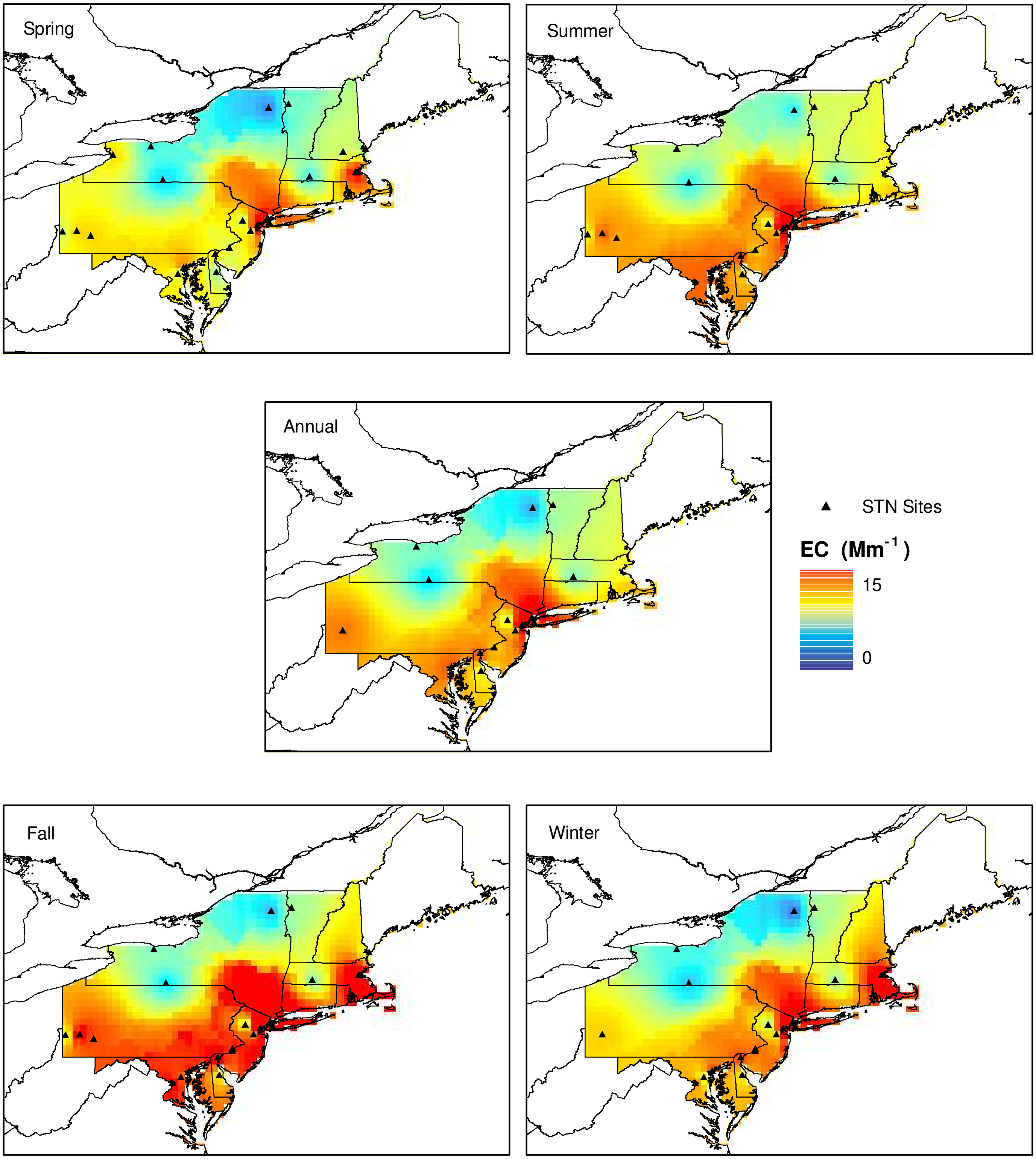

Figure 5a. Organic Carbon Extinction Based on merged STN and IMPROVE Data

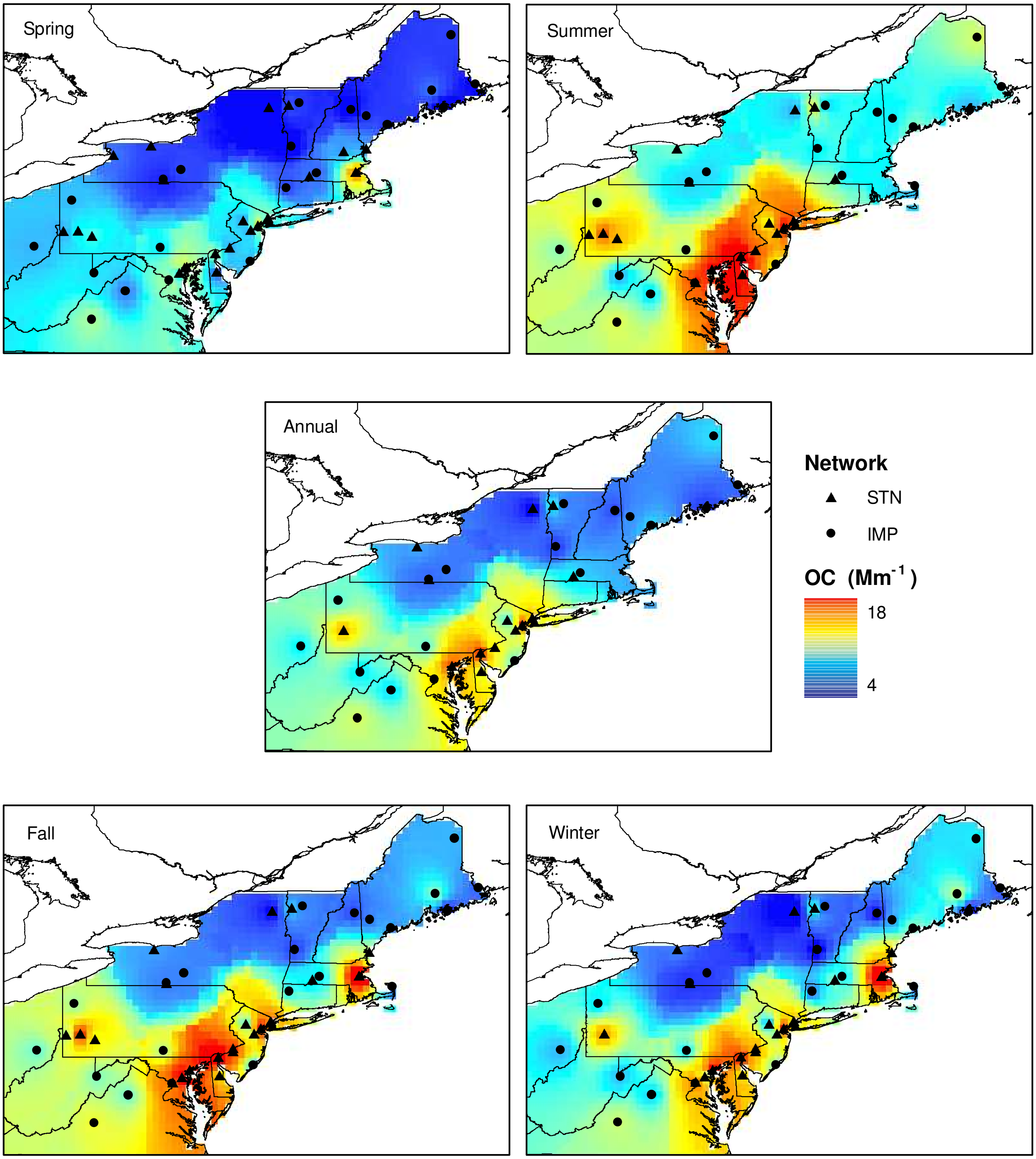

Figure 5b. Organic Carbon Extinction Based on IMPROVE Data

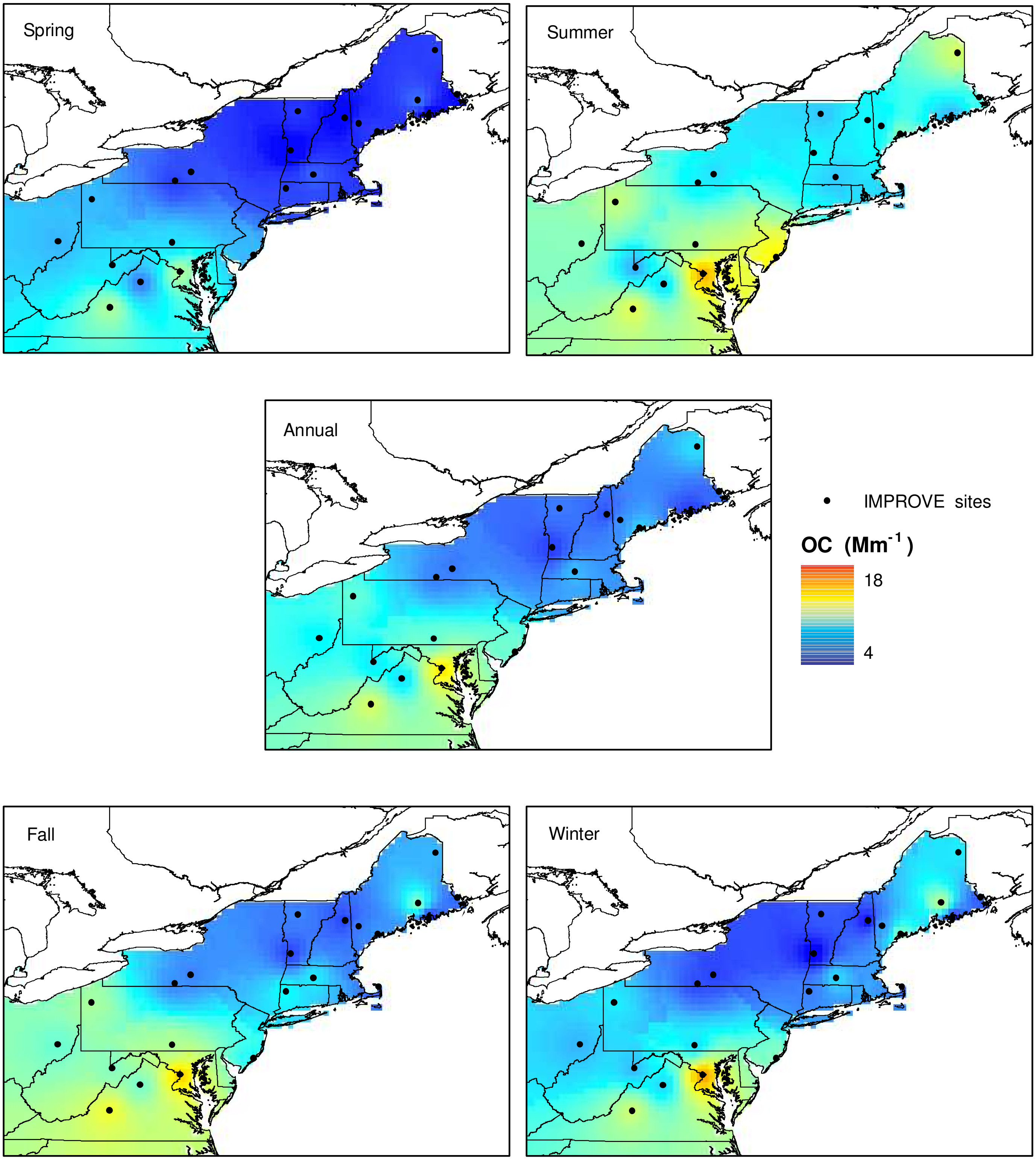

Figure 5c. Organic Carbon Extinction Based on STN Data

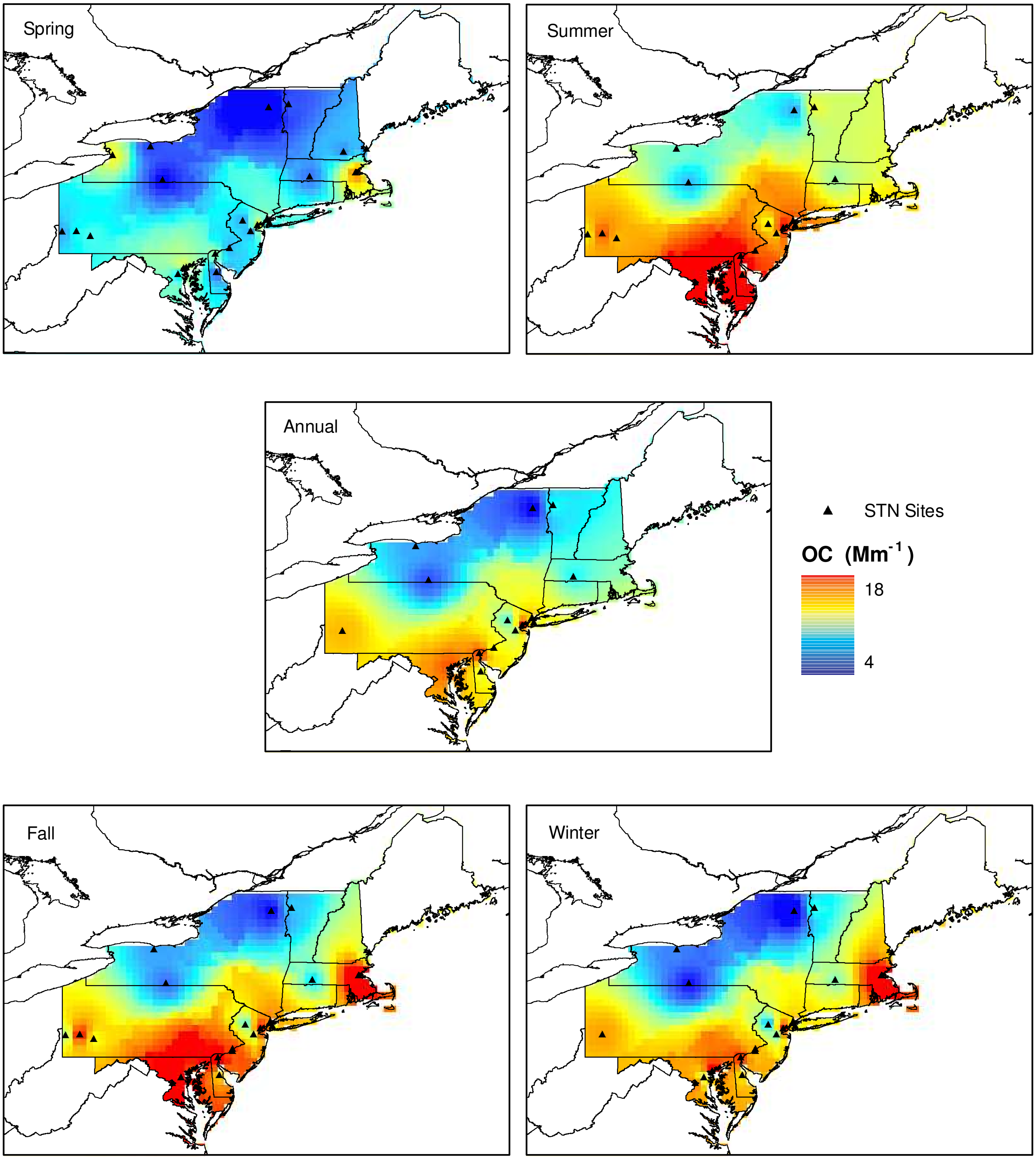

Figure 6a. Soil Extinction Based on IMPROVE Data

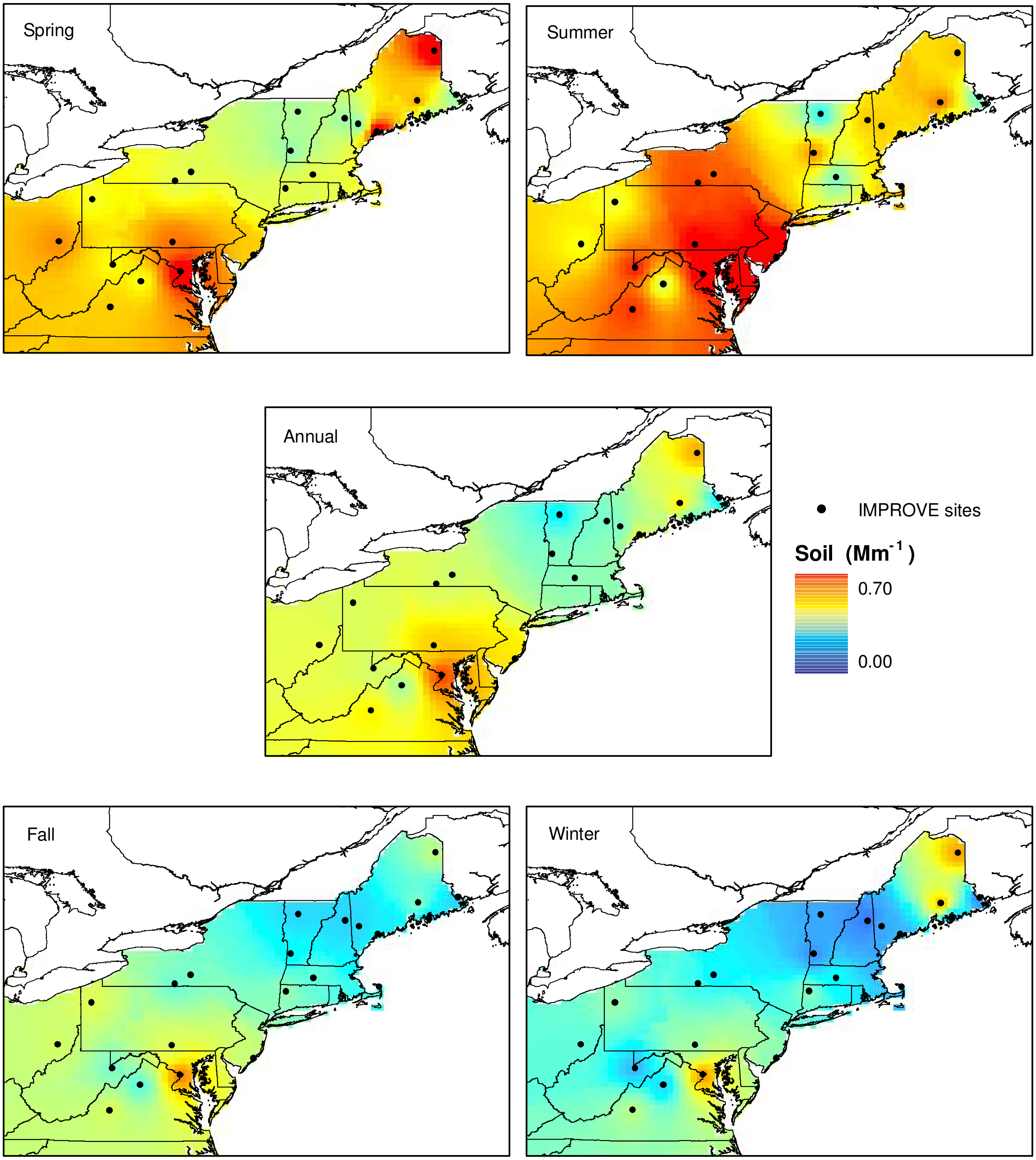

Figure 6b. Soil Extinction Based on STN Data

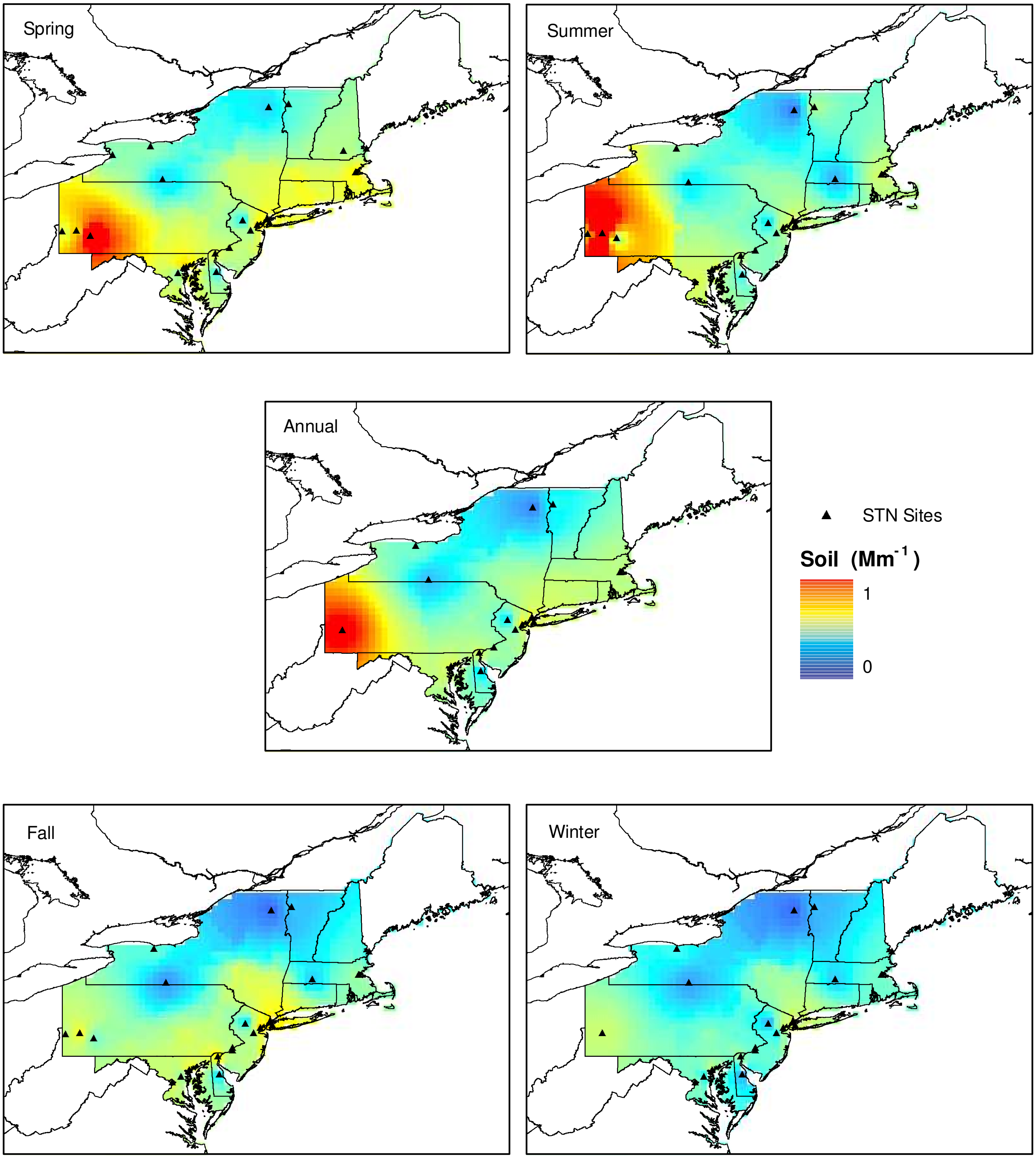

Figure 7. Coarse Extinction Based on IMPROVE Data