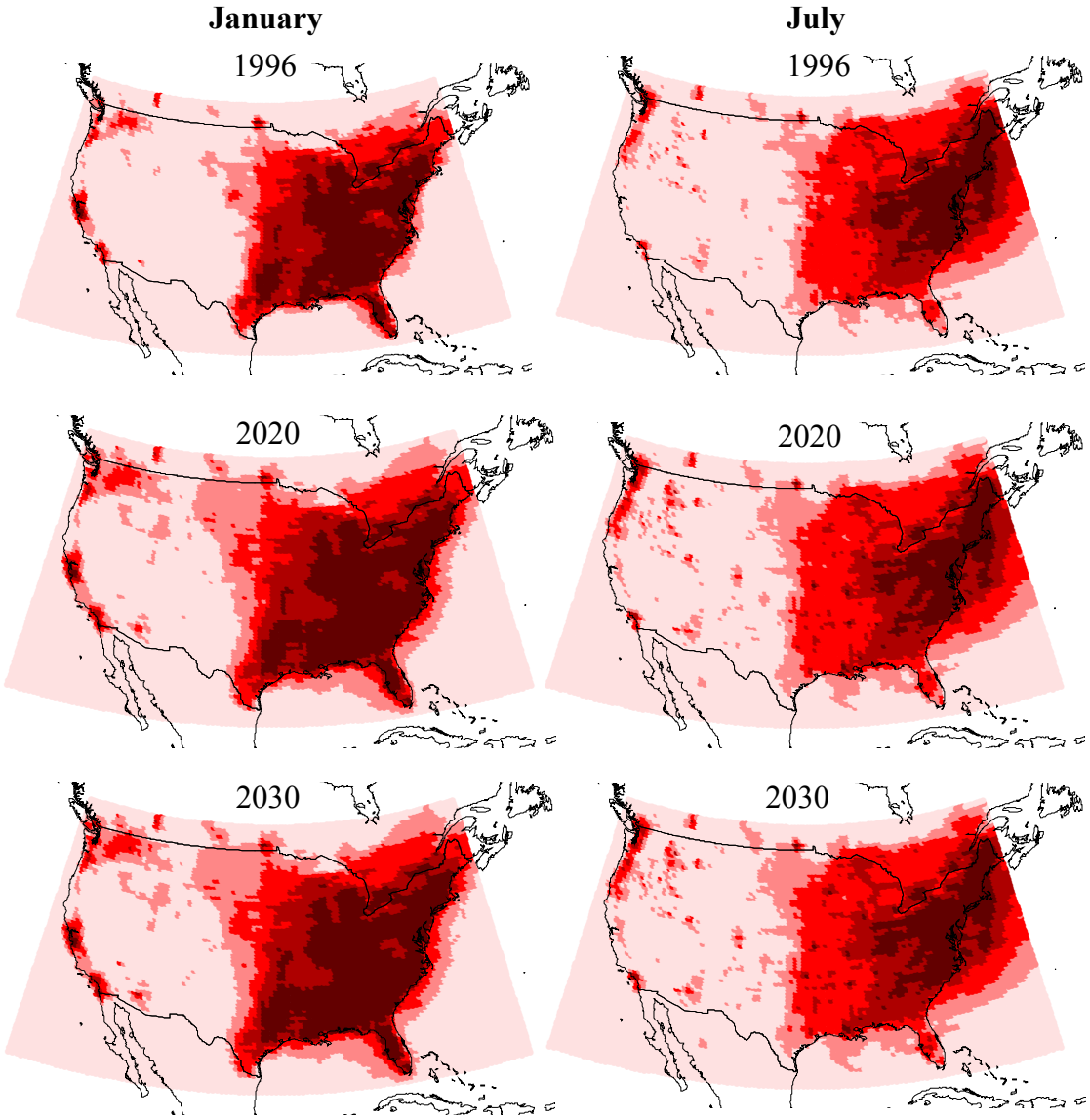
Technical Memorandum #2

REMSAD Modeling Exercises

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

For the MANE-VU Regional Planning
Organization

February 15, 2002



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Submitted to the United States Environmental Protection Agency, Region III in partial fulfillment of requirements for EPA grant X-983384-01-0 to the Ozone Transport Commission

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Acknowledgements

We gratefully acknowledge the funding support provided by the United States Environmental Protection Agency (USEPA) under cooperative agreement with the Ozone Transport Commission.

We thank Tom Braverman of the USEPA for providing USEPA REMSAD output and helpful descriptions of several REMSAD routines. In addition, we would like to thank Brian Timin of USEPA for providing clarification and guidance in installing REMSAD on the PC Windows NT platform. Finally, we would like to thank Tom Myers of SAI/ICF Consulting for providing us with speciated REMSAD output and a binary to ascii conversion program.

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Units, Species, Acronyms

Acronyms

ATDM – Aerosol and Toxics Deposition Model
IMPROVE – Interagency Monitoring of Protected Visual Environments
MANE-VU – Mid-Atlantic/Northeast Visibility Union
MARAMA – Mid Atlantic Regional Air Management Association
NADP – National Atmospheric Deposition Program
NESCAUM – Northeast States for Coordinated Air Use Management
NET – National Emission Trends Database
REMSAD – Regulatory Modeling System for Aerosols and Deposition
USEPA – United States Environmental Protection Agency

Chemical Species

ASO4 – sulfate formed through aqueous processes
GSO4 – sulfate formed through gaseous processes
NH4N – ammonium associated with nitrate particles
NH4S – ammonium associated with sulfate particles
PEC – primary elemental carbon
$PM_{2.5}$ – all particulate matter up to 2.5 μm in diameter
PMFINE – primary particulate matter up to 2.5 μm in diameter (excluding PEC and POA)
PNO3 – particulate nitrate
POA – primary organic aerosol
SOA – secondary organic aerosol

Units/Symbols

Mass

μg – micrograms (0.000001 x g; 10^{-6} g)

Concentration

$\mu g/m^3$ – micrograms per cubic meter

Visibility

dv – deciview
km – visual range in kilometers
Mm^{-1} – extinction in inverse megameters
b_{ext} – reconstructed extinction coefficient

Executive Summary

In order to design control strategies for state or tribal implementation plans (SIPs or TIPs) required under the regional haze rule, it is anticipated that detailed modeled simulations of current and future air quality conditions will be needed. REMSAD may be an appropriate tool to begin assessing future trends in annual visibility conditions under various growth and control scenarios. As a regional air quality model, REMSAD may also be useful for defining the geographic scope of the source regions for Class I areas in the MANE-VU region.

In order to develop capacity to run the REMSAD model in-house, a series of modeling exercises were designed and carried out to replicate modeling work that had been done at USEPA. A formal comparison, as such, was not possible due to differences in versions of the REMSAD code available to NESCAUM and that which was used by USEPA. However, a comparison of results between REMSAD version 4.0 on a Windows based PC (NESCAUM) and version 4.1 running on a Sun Workstation (USEPA) does lend confidence in the successful application of the model. Three separate simulations were conducted using meteorological and emissions input data from a 1996 base year and projected emissions data for 2020 and 2030. Spatially, the patterns of PM2.5 concentration compare well between platforms with differences in monthly averages of no more than 1 to 2 $\mu g/m^3$.

The observed differences may be partially due to rounding errors and compiler differences between computer platforms, but are likely to be due to changes in the REMSAD code between version 4.0 and 4.1. As expected, the extinction coefficient patterns are similar, spatially, to the patterns produced for PM2.5 concentrations. Along the East Coast, significant regions exist where the NESCAUM version 4.0 runs performed on a PC have substantially under-predicted atmospheric extinction relative to the USEPA version 4.1 runs performed on a Sun workstation.

Results of this exercise highlight differences between versions 4.0 and 4.1 of REMSAD but demonstrate successful installation, compilation and execution of REMSAD code on a Windows based computing platform. A formal intercomparison using the same model code on both platforms is needed to identify differences between platforms and to fully understand their specific cause. Nevertheless, the work undertaken to date has advanced MANE-VU's ability to examine detailed fine particle simulation results on a species-specific basis. Next steps for use of this model will include model validation exercises, sensitivity studies, and additional platform intercomparison.

I. Introduction

The regional haze rule, promulgated in July of 1999 by the United States Environmental Protections Agency (USEPA), requires states and interested tribes to submit state or tribal implementation plans (SIPs or TIPs) by December 2008 at the latest. These plans must contain strategies that take initial steps toward remedying any existing and preventing any future visibility impairment experienced at federal mandatory class I areas (heretofore Class I areas). In order to design these control strategies, it is anticipated that states and tribes will need to produce detailed modeled simulations of current air quality conditions as well as simulations of future air quality conditions under a variety of control scenarios. These modeled simulations will need to be robust enough to stand up to legal challenge and demonstrate convincingly that emissions reduction strategies, if implemented, will result in the desired visibility improvement.

Given the strong dependence of visibility conditions on ambient fine particle concentrations, modeled simulations will require the use of a validated fine particle model. Currently two regional air quality models have been advocated for use by USEPA. These include the CMAQ/Models-3 modeling system and the Regulatory Modeling System for Aerosols and Deposition (REMSAD). CMAQ/Models-3 has the advantage that this system was designed as a "one atmosphere" model with more rigorous treatment of gas and aqueous phase chemistry as well as heterogeneous chemistry occurring on the surface of particles. This model, when fully developed, should be able to accurately simulate fine particles and thus visibility conditions on both an episodic and an annual average basis. In addition, CMAQ/Models-3 is being designed to reliably predict tropospheric ozone concentrations and eventually ambient levels of toxic air components. REMSAD was developed by Systems Applications International, Inc. (SAI) for USEPA to understand the deposition, sources and removal processes of fine particles, toxics and other airborne pollutants. REMSAD has been shown to do an "excellent" job simulating annually averaged fine particle mass and a "credible" job simulating annually averaged light extinction (Wayland, 1999). [1] Given the requirements of the regional haze rule, REMSAD is an effective intermediate tool for performing sensitivity analyses and screening runs for haze planning. REMSAD may be an effective tool for defining the geographic scope of the source regions for Class I areas and will enable MANE-VU to begin assessing future trends in visibility under various growth and control scenarios. States and tribes that may be required to comply with regulations dealing with haze, tropospheric ozone and fine particulate in similar time frames will undoubtedly want to develop a "one atmosphere" modeling platform for regulatory work. Initial investigations targeted at gaining a more complete understanding of aerosol chemistry and regional aerosol modeling utilizing the REMSAD platform will allow for a more rapid development of advanced modeling systems in the future.

[1] This study compared IMPROVE aerosol sites in the contiguous United States to REMSAD model results. REMSAD over-predicted $PM_{2.5}$ mass by 4% and under-predicted light extinction as measured by transmissometer by 20%. When compared to reconstructed light extinction, REMSAD under-predicted by only 2%. It should be noted the study employed an earlier version of REMSAD with 1990 MM4 data with a spatial resolution of 60 km by 60 km.

This memorandum describes a series of "proof of concept" exercises designed to demonstrate that the REMSAD system could be installed, compiled and executed on a Windows NT based PC and provide a stable platform for performing future modeling simulations. We performed, as part of these exercises, three 7-month REMSAD version 4.0 simulations. These 7 month periods (January-July) were simulated using meteorological and emissions input data from a 1996 base year and projected emissions data for 2020 and 2030.[2] Results from these runs are compared to results obtained through USEPA that were produced by a contractor using REMSAD version 4.1.[3] Ambient levels of primary and secondary haze-associated pollutants are compared. Given potential differences in the modeling protocols employed by NESCAUM and USEPA's contractors and different versions of the model used, a strict comparison is not possible here, but qualitative comparisons can lend confidence in the initial installation and execution of the model. Resulting changes in projected visibility are analyzed and presented. Finally, recommendations for future REMSAD work are provided.

[2]While annual simulations had been intended for this exercise, we were unable to obtain a complete year of meteorological input files from USEPA for all of the 1996 base year. Missing meteorological files included wind files from August 1 through August 20, September 1 through September 27 and October 1 through October 22. Given that the model requires a few days of 'spin-up' time before stable simulations are possible, we truncated the year after 7 months for these initial tests.

[3] NESCAUM had initially intended to install and compare results from the same version of REMSAD for which results were available on the USEPA heavy duty diesel ftp site (ftp://ftp.epa.gov/modelingcenter/Heavy_Duty_Diesel/REMSAD/). However, we have recently learned that the directory containing results from REMSAD version 4.1 and Sun executable for REMSAD version 4.1 also contains REMSAD code for version 4.0 which was the basis of NESCAUM's installation. Thus NESCAUM results are based on a different model version than the available USEPA results, but comparison with these results will still provide qualitative information regarding model performance.

II. REMSAD Model Description

REMSAD is a three-dimensional Eulerian photochemical grid model that consists of a meteorological pre-processor, the aerosol and toxic deposition model (ATDM) and a series of post-processors.[4] More recent versions of REMSAD are currently available; however, the present analysis is based on REMSAD version 4.0 which was obtained from USEPA (USEPA, 2001).[5] The model may be run with varying degrees of horizontal and vertical resolution and provides a choice of horizontal grid. Both USEPA's contractor and NESCAUM employ a latitude/longitude horizontal grid with resolution of 0.5 degrees longitude by 0.33 degrees of latitude (approximately 36 km on a side) and a sigma-pressure vertical grid with 8 vertical layers. The continuity equation is solved for each chemical species simulated by the model, providing the basis for determining species concentration.

Particles[6] that contribute to visibility degradation include: sulfate, nitrate, organic aerosol, elemental carbon, and crustal material. The concentrations of these species vary seasonally and spatially across the country. Haze pollutants are emitted directly into the atmosphere (primary particles) and created in the atmosphere through chemical transformation (secondary particles). Studies have shown secondary sulfate to be a predominant contributor to poor visibility in the Northeast and Mid-Atlantic States (NESCAUM, 2001). This is, in part, due to the hygroscopic nature of both the sulfate and nitrate portions of aerosol. This affinity for water tends to cause sulfate and nitrate containing particles to grow rapidly under conditions of high relative humidity, swelling in size until they populate that portion of the aerosol size distribution most associated with the scattering of visible light. It is, therefore, particularly important that REMSAD reliably simulate sulfate concentrations in order to calculate visibility conditions in the Northeast and Mid-Atlantic region.

Table 1. lists the major haze-related pollutants simulated by REMSAD. The model relies on input files to estimate the emission of sulfur dioxide, nitrogen oxides, volatile organic compounds, secondary organic aerosol (SOA), carbon dioxide, ammonia, and primary particulate matter into the atmosphere.[7] Secondary organic aerosol concentrations are calculated in a preprocessor.[8] Thus SOA formation processes are not accounted for in the model and SOA in the model is simply subject to meteorological

[4] When the term "REMSAD" is used in this document, we refer to the ATDM portion of the modeling system. Pre- and post-processing functions are specifically referenced in the text.

[5] This version of REMSAD is based on UAM-V regional-scale photochemical model version 1.23 with the following two alterations: no plume-in-grid and a modified Carbon Bond IV routine (micro-CB4). This modified routine incorporates a portion of VOC species compared to the full CB4

[6] A more detailed discussion of aerosol microphysics, including assumptions and methods used in REMSAD's physics and chemistry routines, can be found in the REMSAD User Guide (SAI,1998).

[7] Primary particulates includes coarse particles with an aerodynamic diameter greater than 2.5 μm but less than or equal to 10 μm and primary fine particulates including primary organic particles (POA), primary elemental carbon (PEC), and other primary $PM_{2.5}$ (PMFINE) all of which have an aerodynamic diameter less than or equal to 2.5 μm.

[8] The SOA concentration is determined from an estimated SOA yield from VOC emissions. This deficiency is being addressed in version 7 of the model.

transport. The simplified treatment of SOA is largely due to the tremendous uncertainties in its chemical formation mechanisms and is a deficiency in most air quality models.

Table 1. The major haze-related pollutants simulated by REMSAD

Species	Abbrev	Gas or Particle [9]	Emission Input File[10]	Calculated by Model Output[10]
Sulfur Dioxide	SO_2	Gas	X	
Nitrogen Dioxide	NO_2	Gas	X	
Ammonia	NH_3	Gas	X	
Total Sulfate[11]	SO_4	Particle	X	X
Particulate Nitrate	PNO_3	Particle	X	X
Elemental Carbon	PEC	Particle	X	
Total Organic Aerosol[12]	TOA	Particle	X	
Particles whose diameter < 2.5 μm[13]	PMFINE	Particle	X	
Particles with diameters between 2.5 and 10 μm	PMCOARSE	Particle	X	
Ammonium	NH_4	Particle		X

REMSAD decomposes primary particulate matter into primary elemental carbon, primary organic aerosols, primary sulfate, primary nitrate, crustal matter (considered to be $PM_{2.5}$) and primary coarse particles. The model simulates the formation of the haze contributing secondary particles, sulfates and nitrates, through a select number of chemical pathways. Only the dominant chemical pathways are included. This conserves both computational time and memory. REMSAD simulates the chemical transformation of sulfur dioxide and nitrogen oxides (supplied through emission input files at every grid cell in the model) into secondary particles (i.e. sulfates and nitrates) through gas and aqueous phase chemistry schemes. REMSAD considers all particles, once formed, inert.

In the gas phase, predominant sulfate formation mechanisms include the reaction of sulfur dioxide with the hydroxyl radical to form sulfuric acid. Sulfuric acid has a very low vapor pressure and hence, will almost immediately change to the liquid phase.

[9] Particle may be solid or liquid. REMSAD and other regional models assume homogeneous particles, though particles do exist in nature as heterogeneous.

[10] Those particles included in the input file are considered primary pollutants while those formed in the atmosphere from gaseous precursors are considered secondary pollutants. With the exception of secondary organic material, other secondary components of particulate are calculated by REMSAD.

[11] Total Sulfate = aqueous phase created sulfate + gaseous phase created sulfate

[12] Total Organic Aerosol = primary organic aerosol (POA) + secondary organic aerosol (SOA)

[13] PMFINE includes all directly emitted (primary) particle species with a diameter less than 2.5 μm other than directly emitted organic (POA) and elemental carbon (PEC).

REMSAD allows for this chemical reaction and considers the sulfuric acid produced to be particulate sulfate.[14,15]

When available, the aqueous phase presents a significantly more efficient pathway for sulfate formation. When relative humidity is sufficiently high, sulfur dioxide dissolves in available water droplets where it may react with ozone, hydrogen peroxide or oxygen in the presence of metals to form sulfuric acid. [16] These reactions are dependent on such properties as the cloud pH[17] and the availability of oxidants. The sulfur dioxide and hydrogen peroxide reaction occurs quickly and is considered pH independent.[18] The species with the lowest concentration will be the limiting reactant. The ozone reaction is favored at high cloud pH and becomes relevant when pH is above 4.[19] This reaction is considered to be 10 times faster than the hydrogen peroxide reaction at a pH of 6. Though the ozone reaction may dominate in specific regions where pH is both high and the ozone concentrations are sufficient, the reaction is self-quenching.[20] Sulfur dioxide may also react with oxygen in the presence of metals and may be a dominant pathway at high pH though the current understanding of the rate constants is uncertain. It has been shown that the hydrogen peroxide reaction is dominant for pH less than roughly 4 to 5 (Seinfeld and Pandis, 1999). When the pH increases above this level, the ozone pathway begins to dominate, becoming stronger as the pH increases.

REMSAD version 4.0 only includes the hydrogen peroxide reaction. The liquid-phase sulfur dioxide concentrations available to react through the aqueous phase chemistry is not dependent on cloud liquid water content, as would be expected, but rather a reactant-limited sulfur dioxide-hydrogen peroxide oxidation method (Kasibhatla et al., 1997; Venkatram et al., 1997).[21] Given that the aqueous phase reaction between ozone and sulfur dioxide is not included, the model's accuracy in predicting sulfate concentrations may be compromised at high pH.[22]

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[14] Sulfuric acid may be in the gaseous phase or liquid phase depending upon the ambient relative humidity. Sulfuric acid has an extremely low vapor pressure so it is likely to exist in the liquid form at relative humidities greater than approximately 5%. The model assumption that sulfuric acid does not exist as a gas is therefore considered appropriate (Seinfeld and Pandis, 1999).

[15] The hydroxyl radical concentrations are determined from multidimensional lookup tables as a function atmospheric altitude, temperature, humidity, and other pertinent chemical species.

[16] Other oxidants species are not mentioned here as they are not considered to play a substantial role in forming sulfuric acid.

[17] pH is defined as $pH=-\log_{10}[H^+]$.

[18] Hydrogen peroxide solubility increases with pH, however, the rate constant of this reaction decreases with pH. In essence, these two properties tend to have opposite but similar effects thereby making the reaction pH independent.

[19] Pure water at a temperature of 298K has a pH of 7.0. The average pH of rainwater varies from 3.8 to 6.3 where a pH of 5.6 is considered 'natural'. Hence, rainwater is naturally acidic.

[20] When ozone reacts with sulfur dioxide, the pH within the cloud then becomes more acidic bringing down the pH.

[21] This method requires knowledge of the amount of cloud fraction that is precipitation. REMSAD does not keep track of non-precipitating versus precipitating clouds, hence, an assumption is made that 50% of cloud coverage is non-precipitating.

[22] In the U.S., NADP 1996 data show pH values determined from measurements at field laboratories for the Eastern U.S. to range from 4.3 to 4.8. The values are higher for other parts of the country (i.e. pH=5.9

Both the aqueous phase and gas phase processes result in the formation of sulfuric acid particles. These particles may then react with ammonia to form an oxidized sulfate: ammonium bisulfate, letovicite, or ammonium sulfate. The level of oxidation depends upon many factors including the ammonia to sulfate molar ratio, relative humidity and temperature. Additionally, the concentration of nitric acid plays an important role as nitric acid may chemically react with ammonia to form particulate nitrate in the absence of sufficient quantities of the preferred reactant, sulfuric acid. When high concentrations of sulfuric acid are present (i.e. when the ratio of ammonia to sulfate is less than two) REMSAD does permit the formation of ammonium nitrate; however a parameterized equilibrium is assumed which strongly favors the formation of ammonium sulfate. REMSAD further assumes that all oxidized sulfate is present as a fully neutralized sulfate species, ammonium sulfate. Nitrate may be considered particulate nitrate or gas phase nitric acid depending upon the availability of ammonia, temperature, relative humidity, and other meteorological parameters.

The REMSAD model separates particle mass by chemical species for model output so that 'pure' sulfate mass is included in one variable while 'pure' ammonium mass that is associated with sulfate is another. A similar procedure is followed for nitrate. This has consequences for visibility calculations which ordinarily would consider differences in the scattering properties of sulfuric acid and ammonium sulfate.

The REMSAD chemistry treatment discussed above has been revised in version 6 of the model with improvements that have been incorporated by the developer, SAI. For example, the ozone and diatomic oxygen reactions with dissolved sulfur dioxide in the aqueous phase have been included in version 6. Similarly, version 6 also contains treatment of liquid water content attributable to clouds. Table 2 outlines current and scheduled changes of the chemical treatment included in REMSAD by version number. NESCAUM is in the process of installing and compiling version 6.2 on a linux workstation for testing.

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measured at a laboratory in Northern Colorado) (information available at ftp site: http://nadp.sws.uiuc.edu/isopleths/maps1996/phfield.gif).

Table 2. REMSAD version comparison

Version	Approx. Release Date	Chemical Treatment			
		Sulfate Aqueous Chemistry	**Nitrate**	**Mercury**	**SOA**
4	Spring, 2000	H_2O_2	Parameterized[23]	Aqueous Phase chemistry (O_3 only)	No[24]
5[25]	Spring, 2001	H_2O_2	Parameterized	Aqueous Phase chemistry (O_3 only)	No
6[26]	Fall, 2001	H_2O_2, O_3, O_2	MARS-A algorithm	Based on Lin and Pehkonen (1999)[27]	No
7	Winter, 2001	H_2O_2, O_3, O_2	MARS-A algorithm	Based on Lin and Pehkonen (1999)	Yes

[23] Nitrate concentrations is determined from estimated amounts of nitric acid versus aerosol nitrate based on the ratio of ammonia to sulfate.
[24] SOA emissions is estimated on a percentage of SOA formed from VOC emissions.
[25] Version 5 hosts a faster numerical solver for μ-CB4.
[26] Version 6 code has been restructured with "time-splitting" processes (each process contributing to the continuity equation is treated at the minimal time step necessary).
[27] This includes 17 Hg reactions (Lin, C.A. and S.O. Pehkonen, The chemistry of atmospheric mercury: a review. *Atmos. Environ*., 33, 2067-2079, 1999).

III. Methodology

RESMAD version 4.0 was installed on a PC running a Windows NT operating system. Three separate simulations were conducted including: a 1996 base year, a 2020 projected base year and a 2030 projected base year. Each simulation extends from January to July of the respective years. The 1996 base year was modeled using USEPA 1996 emissions information and the 1996 MM5 meteorological data available on USEPA modeling center ftp site.[28] Simulations for 2020 and 2030 use the same 1996 MM5 meteorological data with projected emission information for the respective years provided by USEPA. Results from USEPA's simulations produced on a Sun Workstation running REMSAD version 4.1 and using the same inputs were also available on this site and are used for comparison purposes.[29] It should be noted, however, that limited information was available on initialization files and post-processing routines that were used to produce these results and that a slightly different version of the model was used. While a qualitative comparison of data produced on the two different platforms lends confidence that REMSAD was successfully installed and executed, perfect agreement should not be expected.

There are two types of input emission files accepted by REMSAD: low level emission files and point emission files. Low level emission files include surface emissions from area and mobile sources. Point emission files include information regarding the emissions and locations of specific point sources, as well as stack parameters. Sources whose effective plume height (as calculated using stack parameters and meteorological parameters) is lower than the first vertical layer are treated as low-level sources. The emissions files are further delineated into weekday, Saturday and Sunday emissions. The 2020 and 2030 simulations employ emission files where growth and control assumptions have been applied to the 1996 emission files (USEPA, 2000a).

Table 3. REMSAD parameters used in this study.

Parameter	Description
Domain	126 degrees West and 24 degrees North to 66 degrees West and 52 degrees North
Number of Grid Cells	120 (longitude breaks) by 84 (latitude breaks)
Grid Cell Size	0.5 degree longitude by 0.3333 degree latitude
Vertical Layers	8 (surface to 100mb)
Chemistry Scheme	micro-CB4
Meteorological Input	Fifth-Generation NCAR/Penn State Mesoscale Model, MM5

[28]This data can be found at: ftp://ftp.epa.gov/modelingcenter/Heavy_Duty_Diesel/REMSAD/
Missing meteorological files prevented NESCAUM from conducting full-year runs. (See footnote 2).
[29] See footnote 3.

Table 3 outlines the model parameters employed for our runs. These simulations invoked the micro CB-IV chemistry. Each simulation was run with 8 vertical layers (the first vertical layer extends to approximately 153 meters or 982 mb) and at a spatial resolution of approximately 36 km.[30]

[30] Runs were conducted on a 850 MHz PC with a Pentium III processor. A PGI Fortran compiler was used. A one month run requires approximately 15 hours of processing time and approximately 3 Gb of data storage for all output information. Hence, a one-year run is expected to require a little more than a week of processing time and roughly 35 Gb of storage. Run times on the Sun Workstation are estimated at 1 hour of computation time per simulation day. Linux workstations may be able to run the model four times faster than the Sun (Tom Braverman, USEPA, personal communication).

IV. Results

A. Platform Comparisons

Aside from minor differences in initialization files used, the major difference between NESCAUM simulations and those performed for USEPA by their contractor is the different version of REMSAD (version 4.0 versus 4.1) and the difference in computer platform used by each group. The USEPA contractor runs were performed on a Sun Workstation and the NESCAUM simulations were performed on a Pentium III equipped PC. By comparing a portion of the 1996 output (available through the USEPA ftp site) to our in-house 1996 simulation, NESCAUM is able to verify the successful installation, compilation and execution of REMSAD code on the PC platform.

Given the strong connection between visibility conditions and ambient concentrations of $PM_{2.5}$, we chose to compare daily average concentrations of the PM2.5 variable[31] calculated for the first layer[32] and extinction coefficients calculated using the REMSAD visibility post-processing routine (XYVIS). Specific methodologies for calculating daily average PM2.5 values and daily average 'reconstructed' extinction values, as well as the individual component specific concentrations, were not provided on the USEPA ftp site containing results for their simulations. Therefore, some concern remains whether differences due to variations in averaging methods exist and whether the comparisons presented here are truly representative of platform differences alone.

We initially compared one day of model output – January 6th. We summed the daily average concentrations of all particle species contributing to $PM_{2.5}$. These species include particulate nitrate (PNO3), primary organic aerosol (POA), secondary organic aerosol (SOA), primary elemental carbon (PEC), ammonium that is considered to have reacted with sulfate to form ammonium sulfate (NH4S), ammonium that is considered to have reacted with nitrate to form ammonium nitrate (NH4N), aqueous sulfate (ASO4)[33], gaseous sulfate (GSO4)[34] and other primary $PM_{2.5}$ (PMFINE)[35]:

[31] PM2.5 represents all particle species with diameter less than 2.5 μm: sulfate, nitrate, organics, primary PM, ammonium and elemental carbon. This is not the same variable as REMSAD output variable PMFINE, that consists of certain types of primary PM. See footnote 13.
[32] Ideally, a comparison of each individual haze-related particle species would be performed at the highest temporal resolution (3-hourly); however, this was not possible as the output files provided on the USEPA ftp site contain only daily PM2.5 concentration, PMCoarse concentration and visibility parameters.
[33] Aqueous Phase formed sulfate.
[34] Gaseous Phase formed sulfate.
[35] See footnote 13.

$$\left\{\overline{PM_{2.5}}\right\}_{daily} = \left\{\overline{PNO3} + \overline{POA} + \overline{SOA} + \overline{PEC} + \overline{NH4S} + \overline{NH4N} + \overline{ASO4} + \overline{GSO4} + \overline{PMFINE}\right\}$$

where the bar signifies a daily average concentration for that species. These daily average concentrations for each species were determined by first extracting the species from the average output file and then converting the extracted binary file to an ascii file for analysis.

Figure 1 illustrates the daily average PM2.5 determined by each model run. The pattern and magnitude of PM2.5 is very similar between runs. However, without individual species concentrations available on the USEPA ftp site, it is not possible to further analyze the reason for any discrepancies.

Comparisons of USEPA to NESCAUM monthly average PM2.5 concentrations for January,[36] April, and July were then performed. To obtain NESCAUM monthly average PM2.5 values, the daily average of each particle species concentration was summed to obtain a total PM2.5 daily value. These daily values were then averaged to obtain a monthly PM2.5 value:

$$\left\{\overline{PM_{2.5}}\right\}_{monthly} = \frac{1}{n}\sum_{j=1}^{n}\left\{\overline{PNO3}_j + \overline{POA}_j + \overline{SOA}_j + \overline{PEC}_j + \overline{NH4S}_j + \overline{NH4N}_j + \overline{ASO4}_j + \overline{GSO4}_j + \overline{PMFine}_j\right\}$$

where the bar above each species represents a daily average and the summation index, j, represents each day of the specified month.

Figures 2 – 4 compare monthly PM2.5 average concentrations between REMSAD version 4.0 conducted in-house on a Windows NT platform versus those conducted for USEPA with version 4.1 on a Sun Workstation. Spatially, the patterns of PM2.5 concentration compare well between platforms with differences averaging no more than 1 – 2 $\mu g/m^3$. The Sun workstation calculates higher levels of PM2.5 along the East Coast particularly for July. It is also interesting to note the significantly lower amounts of PM2.5 in January for a few grid cells in California's central valley, though, this pattern is not repeated for the April or July comparisons. Though these differences are non-negligible and warrant further investigation, it is difficult to conduct a comprehensive comparison with the limited data available through the USEPA ftp site.

Despite the potential for small differences due to rounding error and compiler differences across platforms, the more significant differences which are observed in Figures 2 – 4 are not entirely surprising given the differences in model version and that the potential for other differences. Different initialization files may have been used and no information is available on individual species calculations or summation and

[36] The days from January 1st to January 5th are considered to represent the necessary model 'spin-up' time and thus, have been removed each simulation.

averaging algorithms used to produce the USEPA results. While we assume that similar techniques were followed, a detailed explanation for the observed platform differences can only be obtained through a controlled experiment. This will require dedicated time on a Sun workstation for NESCAUM staff to re-run the simulations using exactly the same inputs and model code. The calculated results for each species, individually, at each timestep must be monitored until the source of the observed differences can be identified. Such a detailed comparison was not intended here and is discussed in the *Future Work* section at the conclusion of this memorandum.

Extinction coefficients are calculated using the XYVIS post-processing tool provided with the REMSAD modeling system. The XYVIS routine used in the current analysis was designed for a previous version of REMSAD and may require updating for a fair comparison with the USEPA runs. The differences were anticipated to be very slight; however, revisions to the XYVIS routine that may have been included in the version used by USEPA could result in significant discrepancies between runs. Nevertheless, we are confident that the extinction coefficients generated using different versions of XYVIS should still provide a basis for judging successful intallation and execution of the model code. The NESCAUM calculated 'reconstructed' extinction coefficient was produced as follows:

$$b_{ext} = 3.0*f(rh)*(GSO4 + ASO4 + NH4S + PNO3 + NH4N) + 4.0*(SOA + POA) + 10*PEC + PMFINE + 0.6*PMCOARSE$$

where f(rh), the relative humidity adjustment factor, provides for enhanced scattering due to the growth in particles with increasing relative humidity. This algorithm is used by the Interagency Monitoring of Protected Visual Environments (IMPROVE) program (Malm et. al., 1996) and is further advocated by the Federal Land Managers Air Quality Workgroup (FLAG, 2000). A number of assumptions and approximations have been applied in quantifying this optical property.[37] The XYVIS routine provides the daily extinction coefficient in binary format. These values were then converted from binary to ascii and an average monthly value was obtained by averaging the daily values in ascii format. The XYVIS routine does not include 10 Mm^{-1} for Rayleigh scattering suggested by IMPROVE and FLAG. This amount was added to the monthly average assuming that the USEPA runs includes Rayleigh scattering.

Figures 5-7 compare the extinction coefficient (Mm^{-1}). As expected, the extinction coefficient patterns are similar, spatially, to the patterns produced for PM2.5 concentrations. The greatest variation between platforms can be seen in the Eastern United States. Along the East Coast, significant regions exist where the NESCAUM runs have substantially under-predicted atmospheric extinction relative to the USEPA runs

[37] Such assumptions include: spherical particles, log-normal distribution of particles with a standard deviation of 2.0 and a mean radius of 0.05 μm, all sulfate is assumed to be pure ammonium sulfate, growth for sulfate and nitrates particles with relative humidity simulated in REMSAD can be represented by ammonium sulfate. See NESCAUM report (2001) outlines such assumptions in greater detail.

performed on a Sun workstation. These regions are centered in the Southeast in January and along the Northeast coast in July.

In order to better understand the potential cause of these and other discrepancies, NESCAUM obtained sulfate specific results from USEPA for a run conducted by their contractor, SAI. This simulation was also for the 1996 base year, however, SAI used REMSAD version 5 and incorporated a greater number of vertical layers. Given these important differences, a comparison between these data and results produced by NESCAUM using REMSAD version 4.1 cannot be expected to yield identical results. However, it is interesting to note that despite the differences, we find qualitative agreement between the contractor results using REMSAD version 5 and sulfate concentrations produced by our REMSAD version 4 run on the Windows NT.[38] We compared the sulfate concentrations for January, April and July. Figure 8 displays an overall seasonal similarity in sulfate concentrations between the two runs. This agreement in this qualitative comparison lends no additional insight as to potential causes of the observed differences between the USEPA version 4.1 and the NESCAUM version 4.0 1996 results shown in Figures 1-7 or any indication as to which set of results may be more valid.

B. Simulation Analysis

The comparisons described in the preceding section were intended to insure consistent results between simulations performed on the Windows NT platform at NESCAUM and other platforms used by USEPA and their contractors. In this section we present limited analysis of the NESCAUM results. Figures 9 – 17 show 7 month average concentrations for the individual PM2.5 components as calculated for the first layer of the model during the 1996 base year. Additionally, these figures show how the species average concentration is projected to change between the 1996 base year and the two years for which we obtained projected emission inventory information, 2020 and 2030. These projected emission estimates reflect emissions controls arising from Title IV, Tier II, NOx SIP Call, and MACT. The percent change is simply the increase (decrease) in a species' concentration relative to the 1996 base year:

(projected year – 1996 base year) /1996 base year * 100

The figures display red for increases in species concentrations and blue for decreases in species concentrations. An important first step in developing control

[38] We performed a similar comparison for nitrate, but after determining the newer version of REMSAD exhibited substantially higher winter nitrate concentrations, we discovered the nitrogen treatment within the model had been significantly modified in version 5 and hence, the comparison is not included.

strategies for regional haze will be to understand the changes that are predicted to occur in the absence of further regulations specifically designed to address visibility concerns.

As shown in Figure 9, the highest concentrations of sulfur dioxide occur in the eastern half of the United States where emissions are greatest. This is particularly important as sulfur dioxide is a precursor species required for the formation of sulfate aerosol, which plays a predominant role in contributing to regional haze in the Northeast and Mid-Atlantic States. Control measures and growth accounted for in the USEPA projections indicate that the concentration of sulfur dioxide is anticipated to decrease over much of the East while increases are noted across the Western half of the country. Of course, as the figure displays percent change, the decrease over the Eastern U.S. is substantially greater in magnitude than the increase expected to be experienced in the Western U.S.

Figure 10 illustrates comparative results for sulfate particulate. Consistent with the results for sulfur dioxide, the eastern half of the U.S. displays a decrease in sulfate concentrations while the western half of the country shows substantial increases relative to the 1996 baseline.

Figures 11-17 show projected increases (decreases) in 7-month ambient average concentrations of other haze related chemical species. Ground level nitrogen dioxide concentrations (Figure 11) are expected to decrease considerably by 2020 with further reductions by 2030. Interestingly, we note that increases are expected along the Gulf Coast and regions of Arkansas, Idaho and Southern California. Nitrogen chemistry has been enhanced in subsequent versions of REMSAD and incorporating these improvements may affect the findings presented here.

Ammonia emissions are anticipated to increase substantially along the East Coast in future years leading to greater modeled concentrations as shown in Figure 12. While ammonia is a key component of most particulate measured in the East, it is not likely that ammonia-focused control strategies would be effective. In the absence of ammonia, the sulfur dioxide emitted into the atmosphere will still ultimately form sulfate particulate matter; however, it is likely to be in the form of sulfuric acid which is a much more acidic form of sulfate.

Figures 13 through 17 show projected behavior of various constituent components of particulate matter including primary $PM_{2.5}$ (PMFINE), coarse PM (PMCOARSE), particulate nitrate, organics and elemental carbon. Only organic particles are projected to decrease over the next 30 years in the MANE-VU region.

The XYVIS post-processing routine (included with the REMSAD software distribution) offers a choice of method for calculating total atmospheric extinction. For the present analysis, option number 2 was selected which follows the IMPROVE protocol and uses hourly averaged relative humidity data to derive adjustment factors. Figure 18 illustrates the monthly average extinction calculated for January and July of 1996, 2020, and 2030. A greater degree of extinction is present in January compared to July in the eastern U.S. which may be explained, in part, by the significant seasonal differences in

atmospheric mixed layer heights (depth of the boundary layer). The same phenomenon is evident in runs performed by USEPA and is consistent with monitoring data from 1996, suggesting that 1996 may have been an anomalous year from a visibility perspective. A more significant finding illustrated by this figure is the lack of visibility improvement between 1996 and 2030 as a result of currently projected control programs. This suggests that current federal regulations are not anticipated to achieve desired visibility goals in terms of remedying existing visibility impairment.

While the results of these initial REMSAD exercises offer several potential insights into the causes and extent of the regional haze problem across the country, the model must be thoroughly evaluated prior to drawing any conclusions from the data presented. This topic is addressed in the next section.

V. Future Work

REMSAD has been successfully installed, compiled and executed for a series of 7-month simulations using 1996 base year meteorology and emissions data provided by USEPA. Results of this exercise show some differences between computer platforms which may be due to differences in numerical precision and rounding errors or, potentially, differences in initialization files and post-processing techniques applied to the two platforms. Nevertheless, these exercises have advanced MANE-VU's ability to examine detailed fine particle simulation results on a species-specific basis. Next steps for use of this model will include upgrading to the latest version of REMSAD (version 6.2) with improved nitrate chemistry. Future versions (version 7 is due out imminently) are expected to include a more detailed treatment of organics.

A. Platform Comparisons

Current plans include a comparison between version 6.2 modeling results obtained on a Sun workstation, a PC running a Linux based operating system, and a PC running a Windows NT operating system. This comparison will need to be carefully controlled to ensure that the same model code and same post-processing routines are used in each case. These results will identify any platform specific biases that need to be accounted for between different installations of REMSAD. With USEPA, WRAP, CENRAP and the Midwest RPO all utilizing REMSAD for some aspects of their modeling workplan, it is critical that any potential biases between modeling platforms be identified. The present analysis suggests that such biases are small but significant (<1.5 $\mu g/m^3$ on a nationally averaged basis). Given the observed differences between the runs described in this memorandum, a more detailed investigation using the latest version of REMSAD is warranted.

B. Performance Evaluation

In order develop further confidence in the results of REMSAD simulations a detailed performance evaluation needs to be conducted including comparisons between calculated and monitored ambient pollutant concentrations. To some extent, this has already been investigated by comparing REMSAD output with concentrations collected by U.S. monitoring stations (Wayland, 1999; USEPA, 2000b). In addition, a scientific peer review was conducted for version 4.1 of REMSAD that led to important changes in subsequent versions of the model (Seigneur et al., 1999). By comparing our installation of the most recent version of the model with monitoring data, we can determine model bias, mean error, and other statistics in order to ensure the accuracy of modeled results and identify potential deficiencies that may need to be addressed.

C. Sensitivity Analyses

It is often useful to identify those model properties (e.g. initial conditions, chemical species or reactions, parameterizations, spatial and temporal resolution of input data, etc..) to which results are most sensitive. Analysis of REMSAD results in response to small changes in individual model properties will identify those properties which require the most care and warrant increased attention in the model's handling of that property. For example, small but uncertain elements in the emission inventory can be varied between minimum and maximum emission levels to see if the uncertainty has any effect on resulting visibility calculations. If not, then MANE-VU may be able to save considerable time and energy in preparing inventories for haze work.

Ultimately, REMSAD will be used in conjunction with other regional air quality models to explore various control strategies designed to combat visibility impairment. Each of these models contains approximations, assumptions and parameterizations that determine the accuracy of their results. Understanding the accuracy to which any model is able to simulate visibility conditions is a high priority for regional organizations as they prepare to develop comprehensive plans for combating regional haze.

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Figure 1. January 6th comparison of total $PM_{2.5}$ predicted by REMSAD simulations operating on Sun Workstation (version 4.1) versus Windows NT (version 4.0).

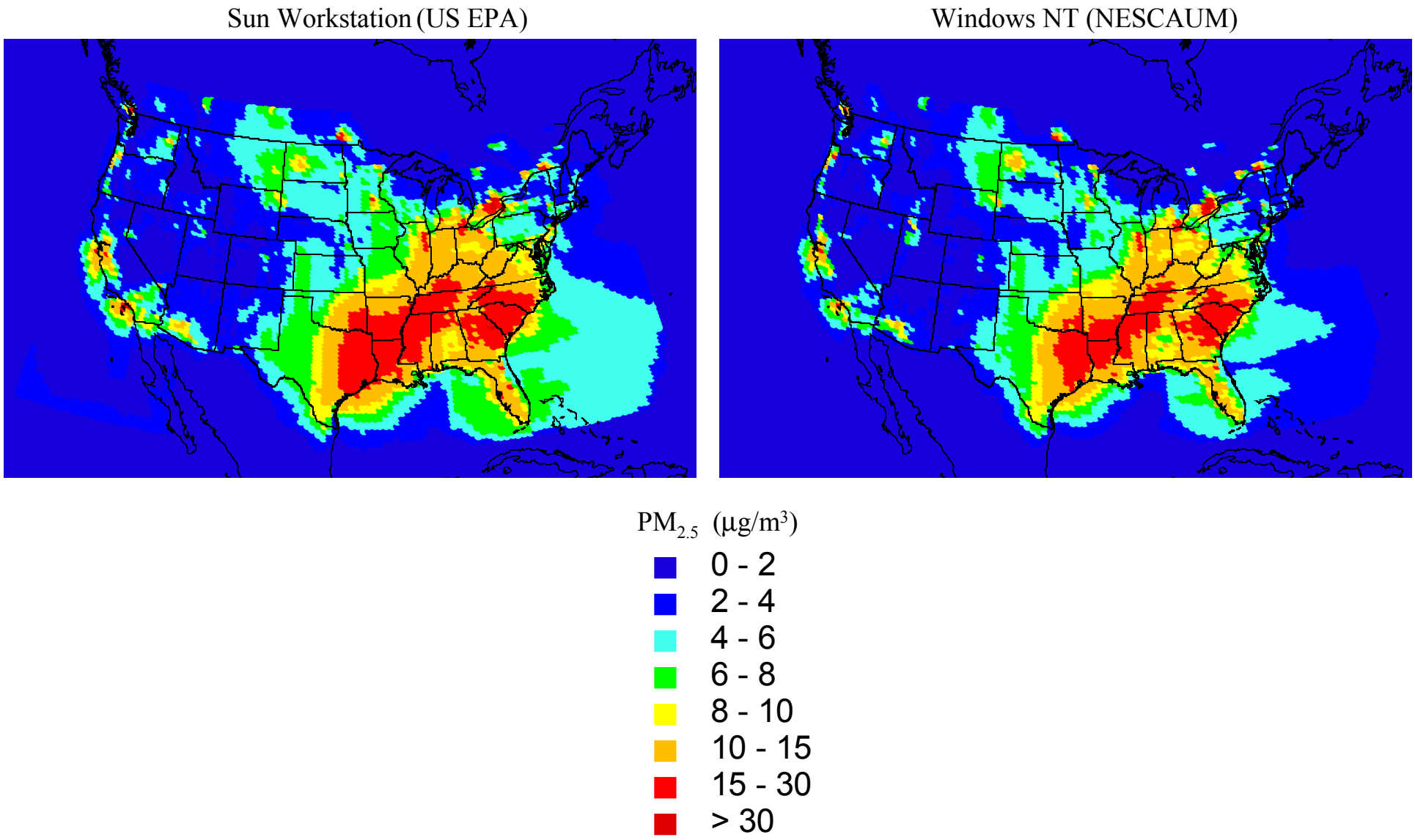


Figure 2. January PM2.5 Comparison

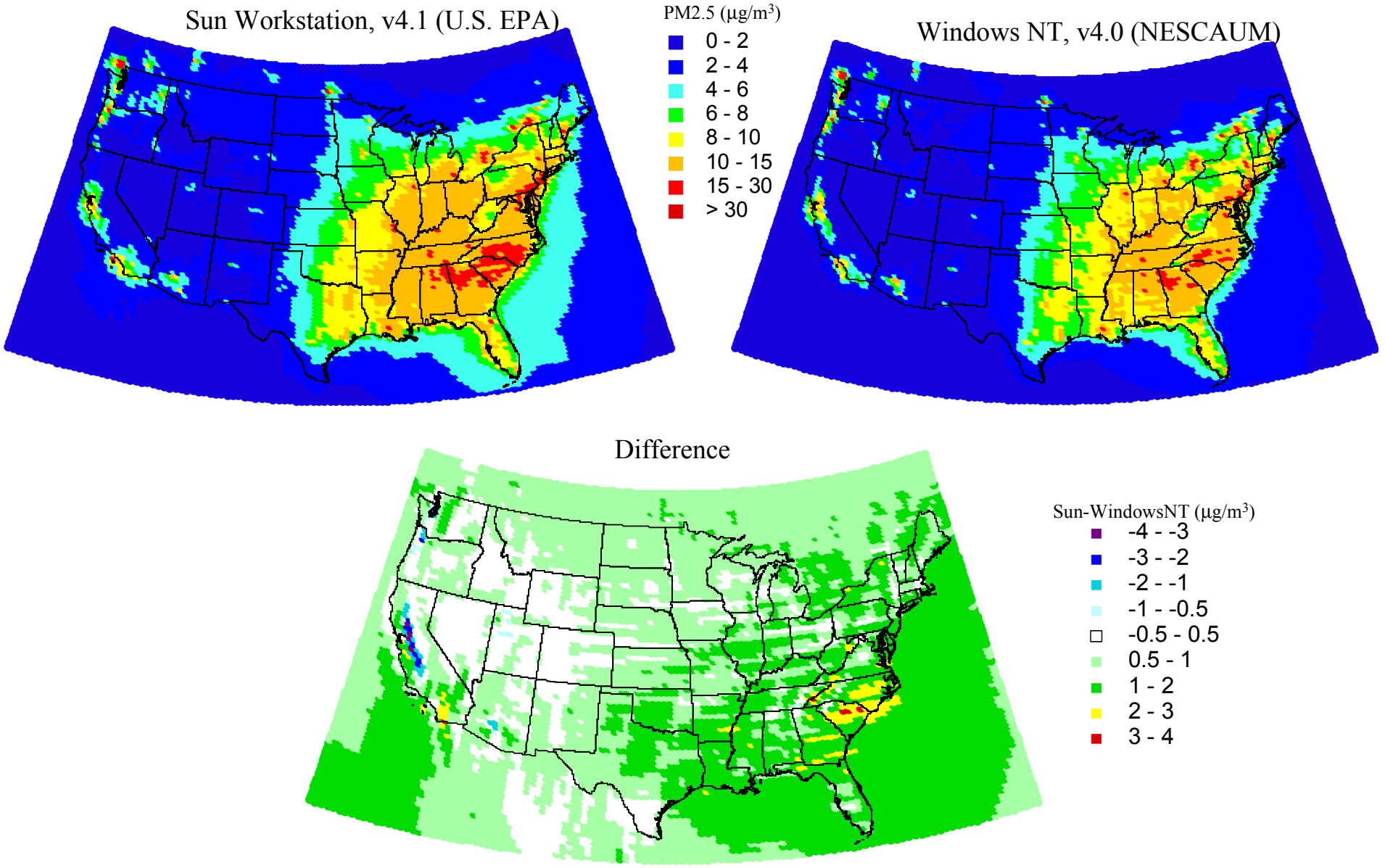


Figure 3. April PM2.5 Comparison

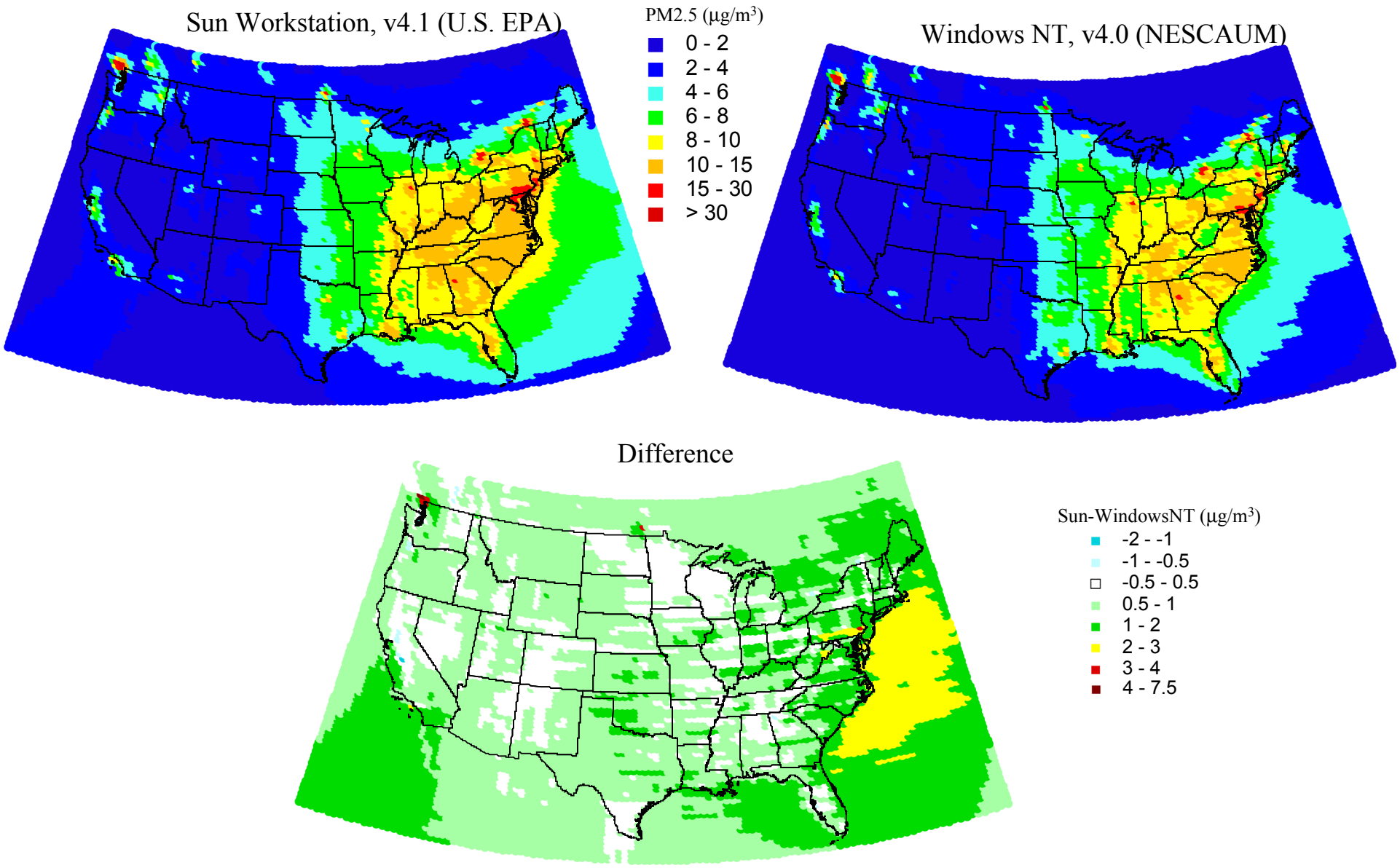


Figure 4. July PM2.5 Comparison

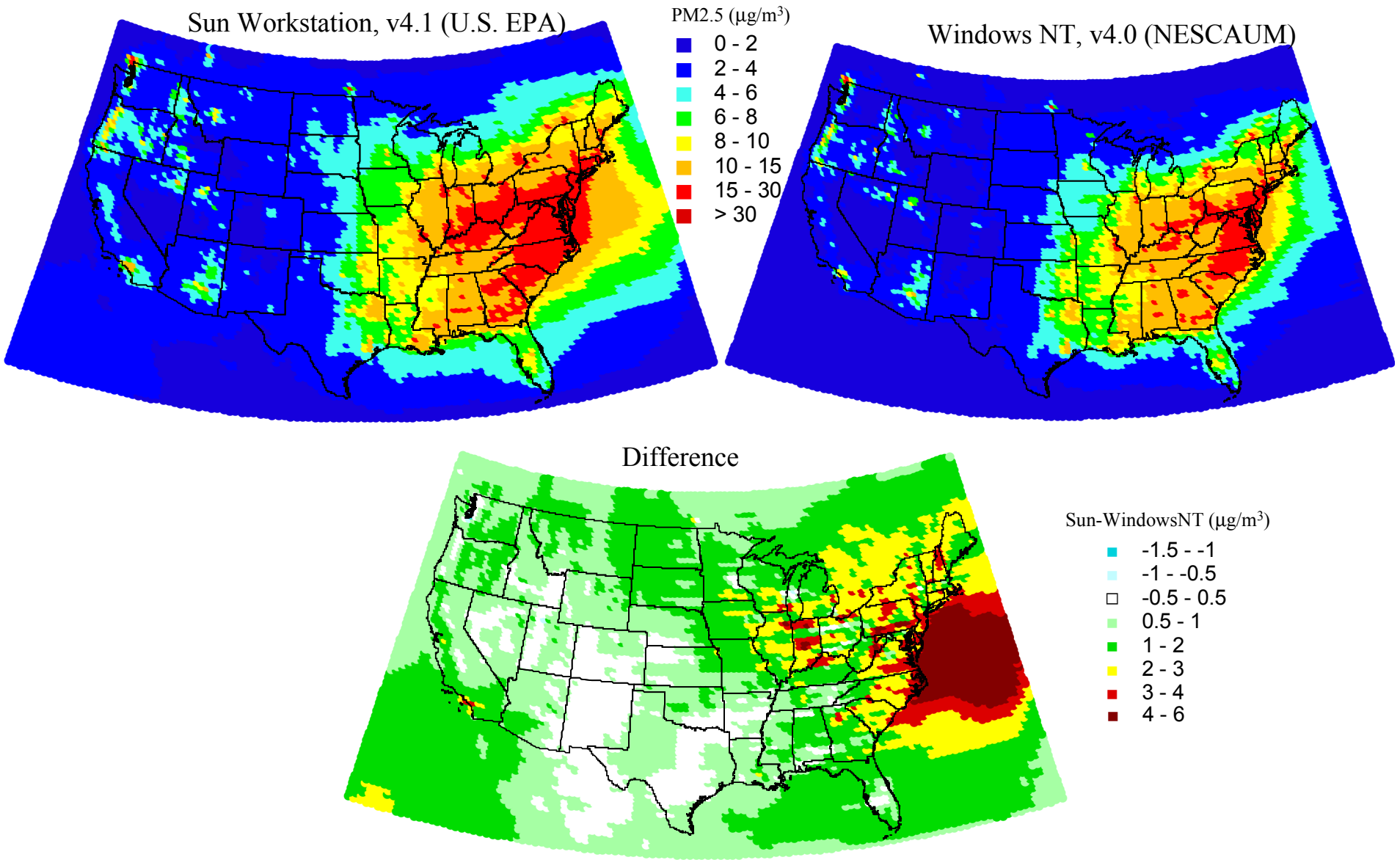


Figure 5. January - Visibility Comparison

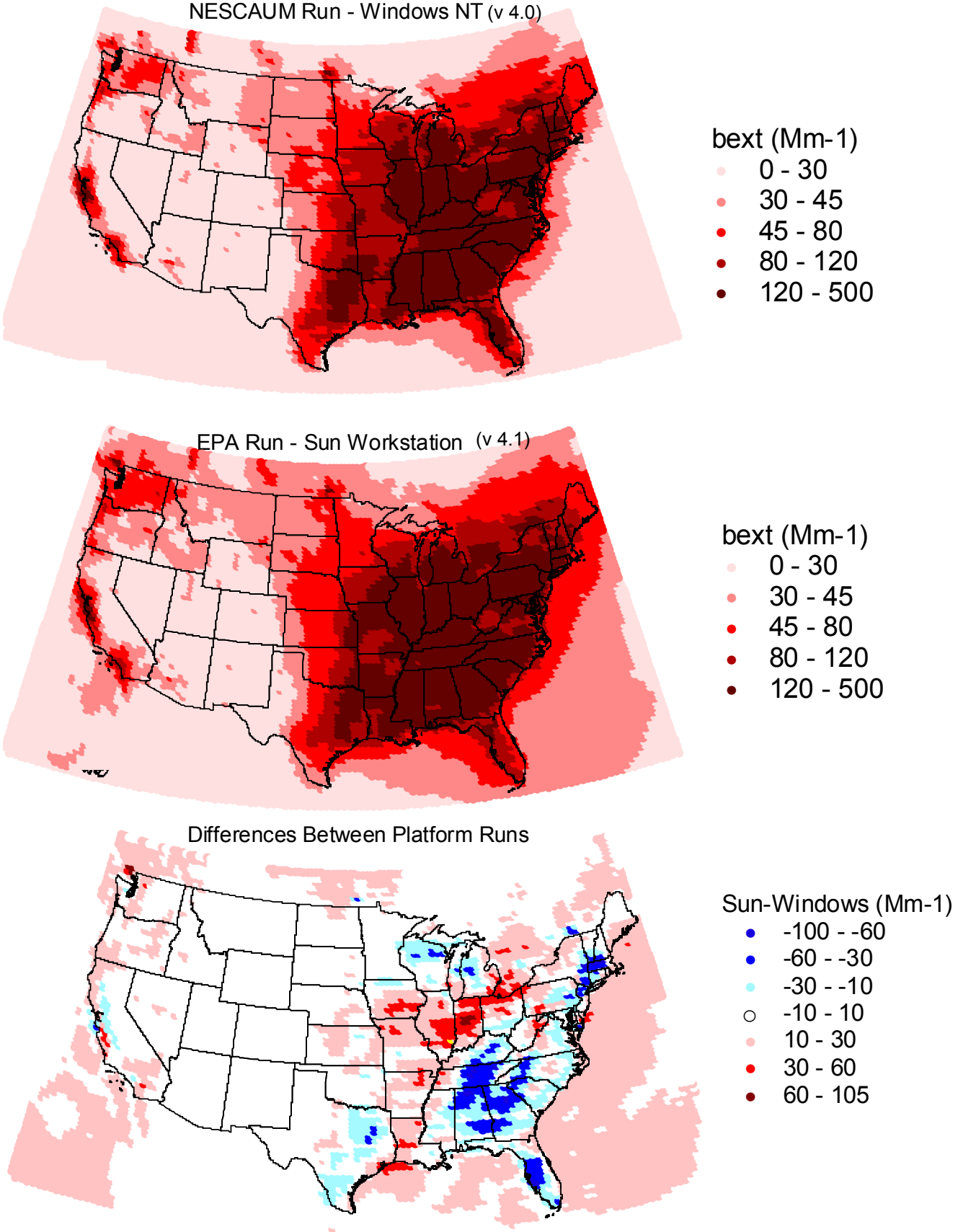


Figure 6. April - Visibility Comparison

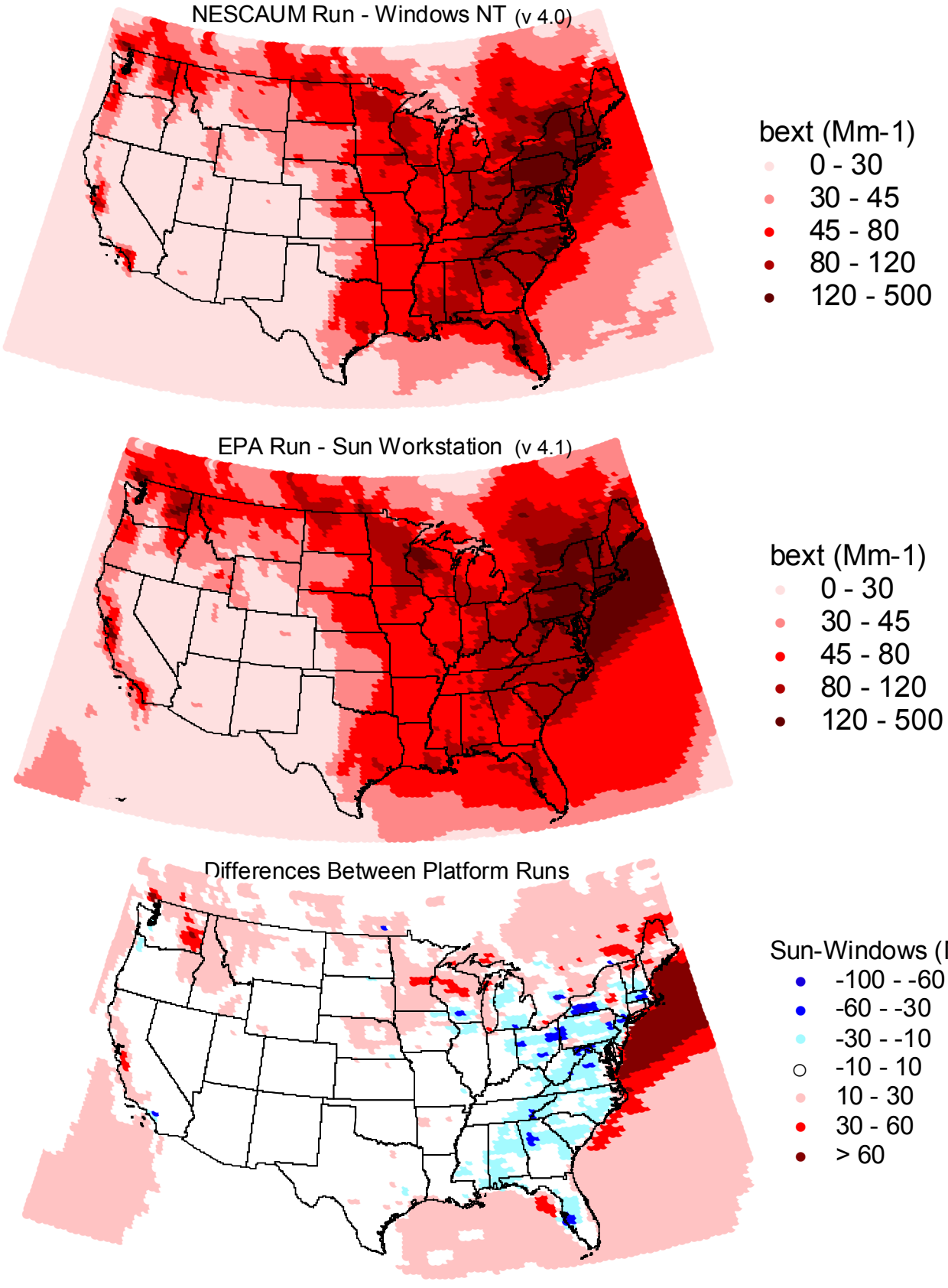


Figure 7. July - Visibility Comparison

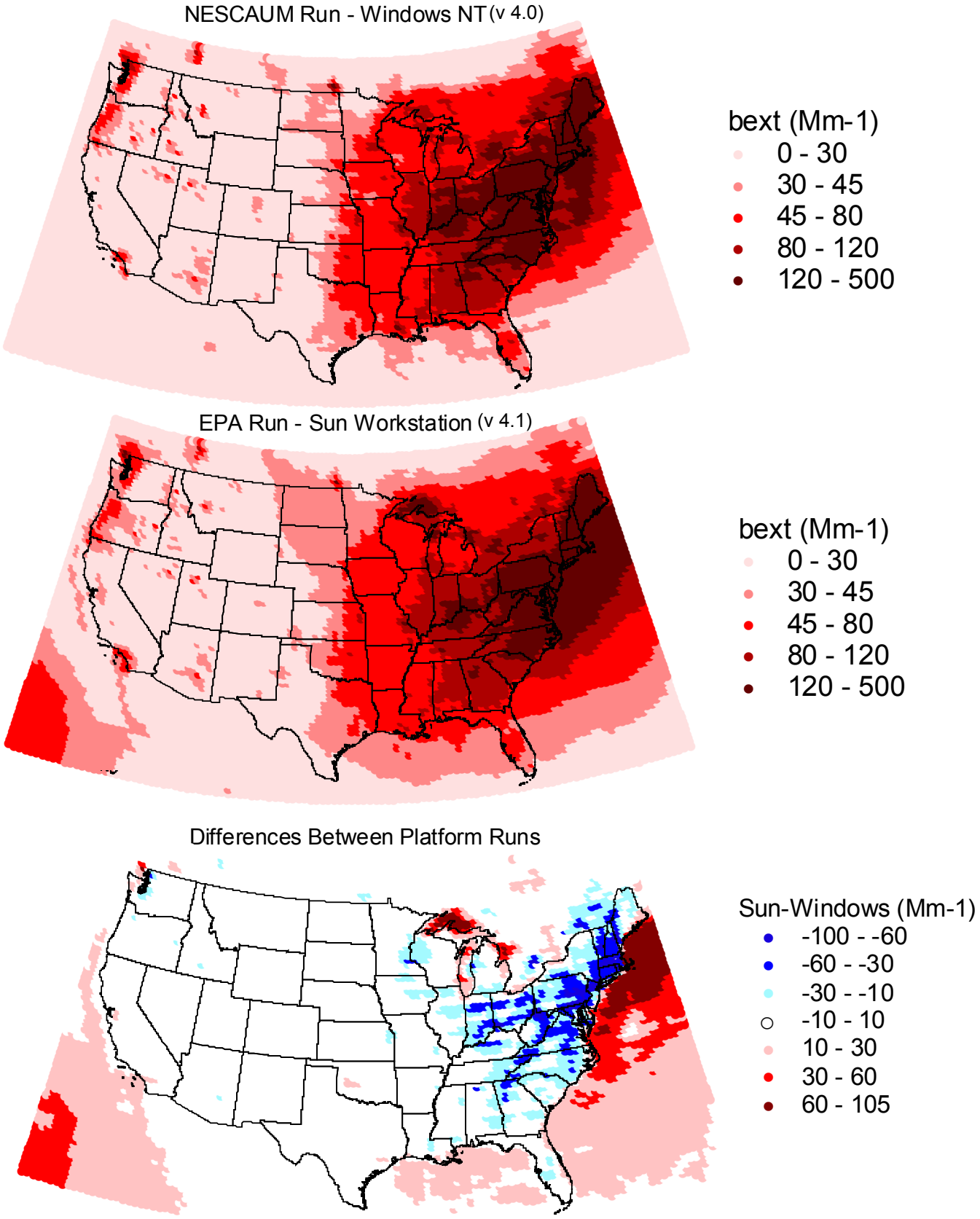
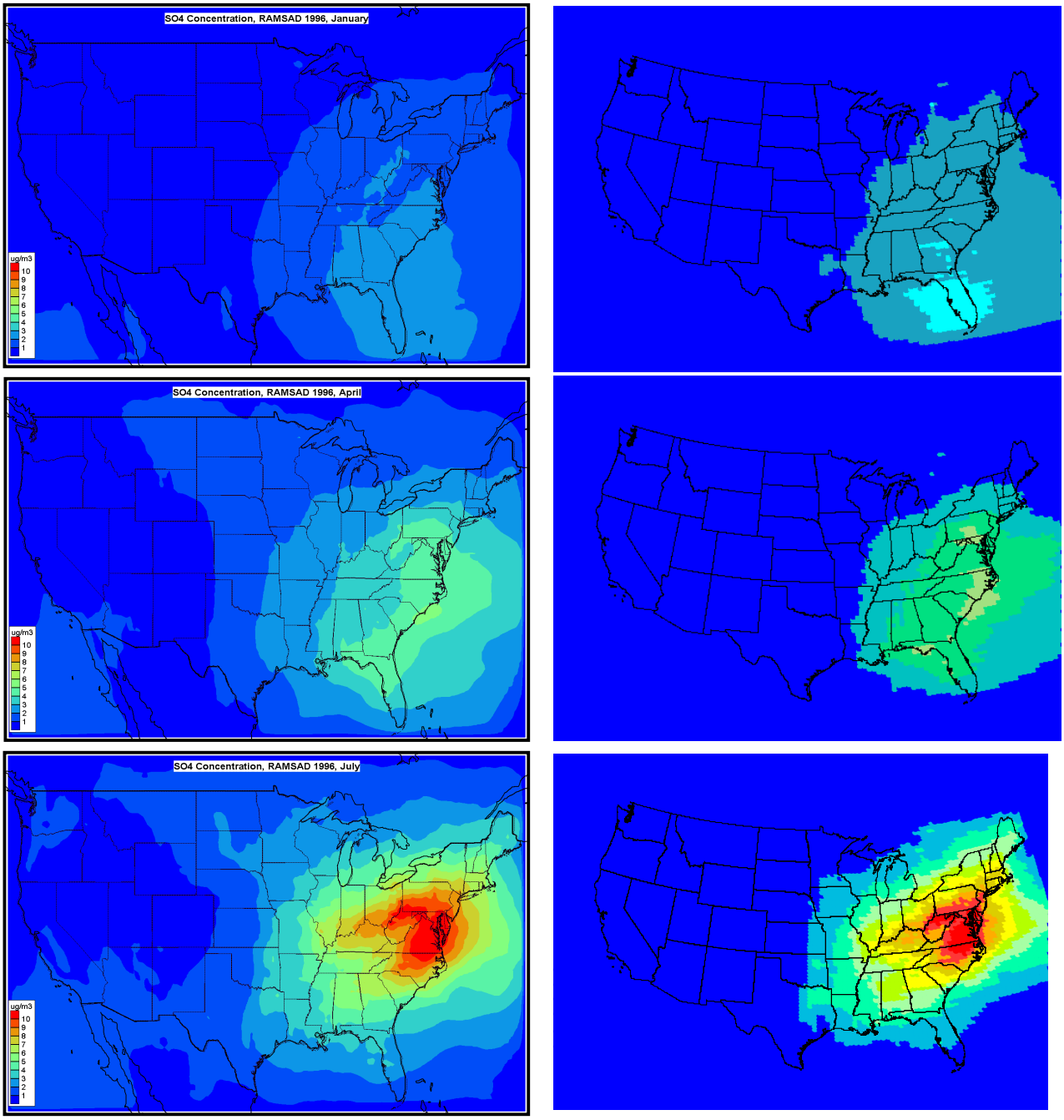


Figure 8. Comparison of Sulfate Results.

REMSAD v5.0[1]

REMSAD v4.0[2]

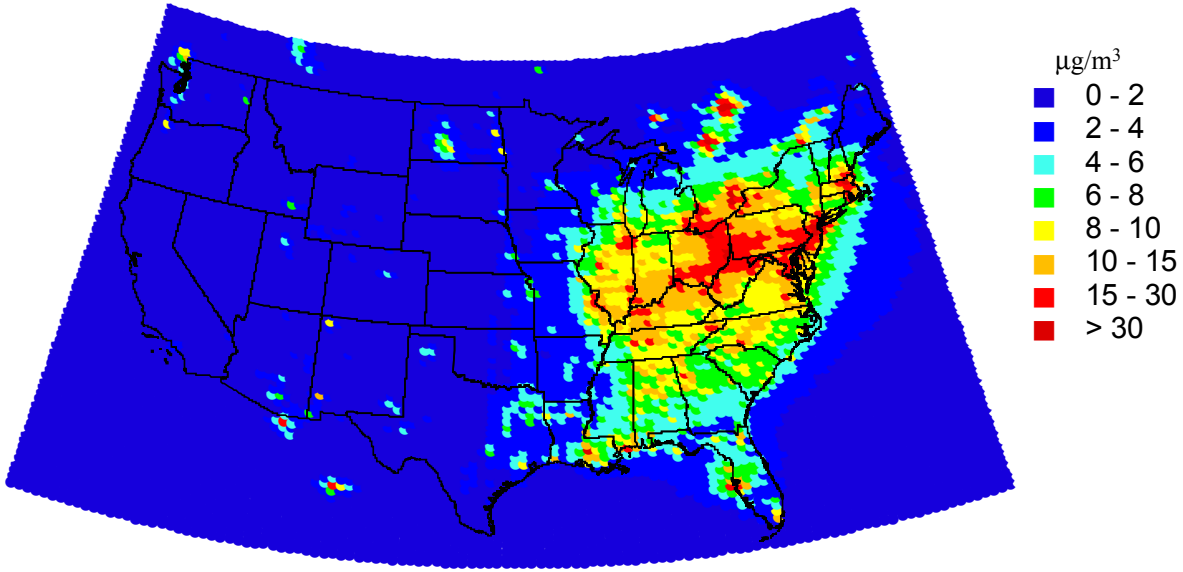


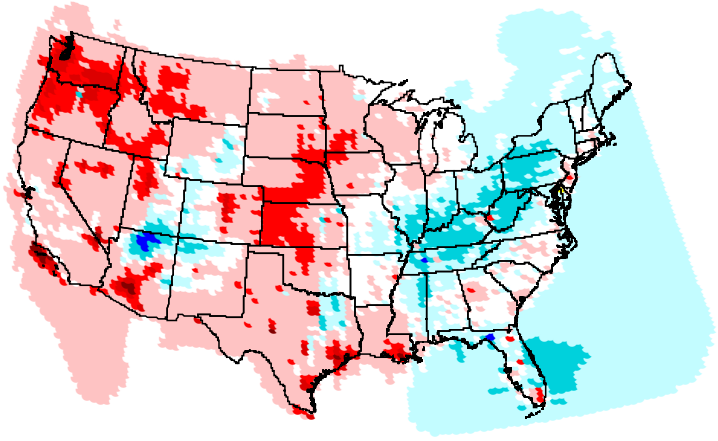
[1]Source: Run conducted by SAI for U.S. EPA. Figures created by Capita

[2]Source: Run conducted by NESCAUM.

Figure 9. Average Sulfur Dioxide

January - July, 1996 REMSAD v4.0





Percent Change
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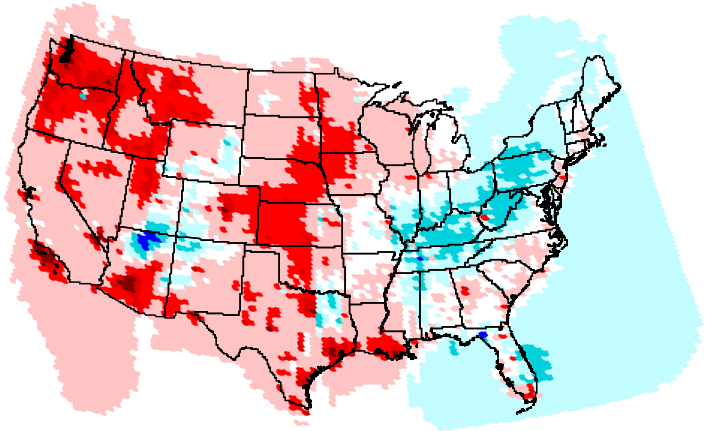
- -100% -to -50%
- -50% to -30%
- -30% to -5%
- - 5% to 5%
- 5% to 20%
- 20% to 50%
- 50% to 100%
- > 100%

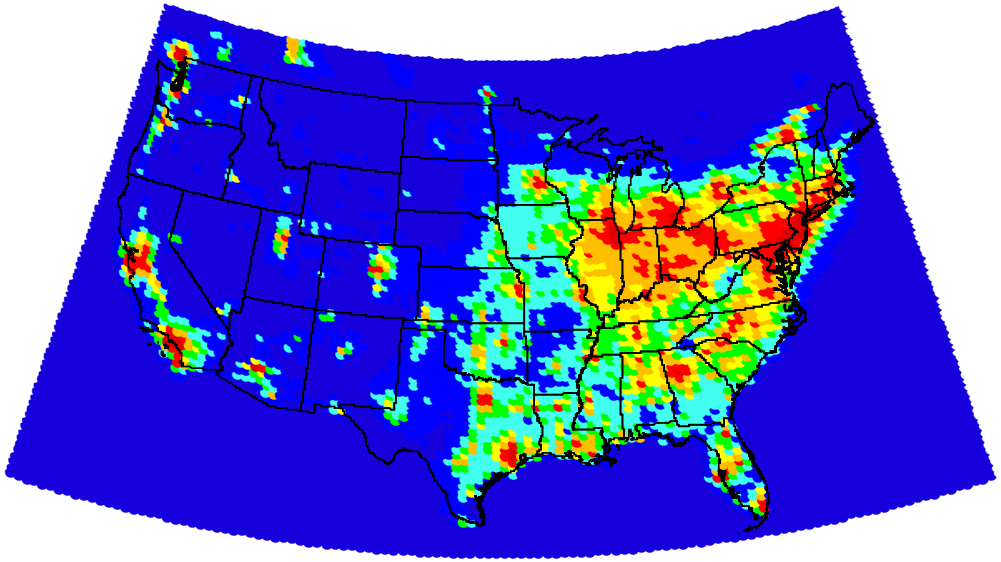


Compared to 2020 projected REMSAD run

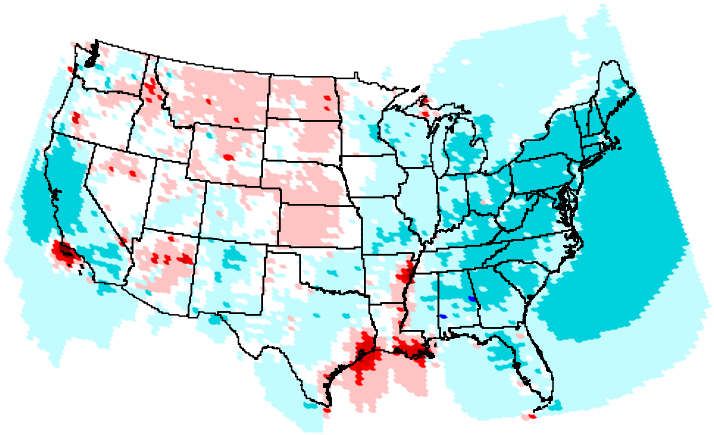
Compared to 2030 projected REMSAD run

Figure 10. Average Nitrogen Dioxide

January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 2
- 2 - 4
- 4 - 6
- 6 - 8
- 8 - 10
- 10 - 15
- 15 - 30
- > 30



Percent Change
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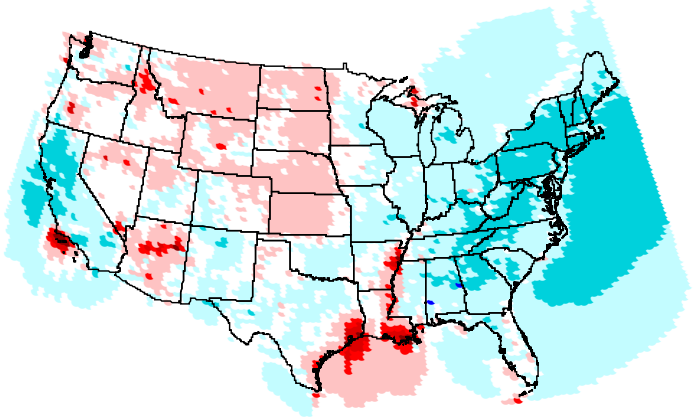
- -30% to -5%
- - 5% to 5%
- 5% to 20%
- 20% to 50%
- 50% to 100%
- > 100%

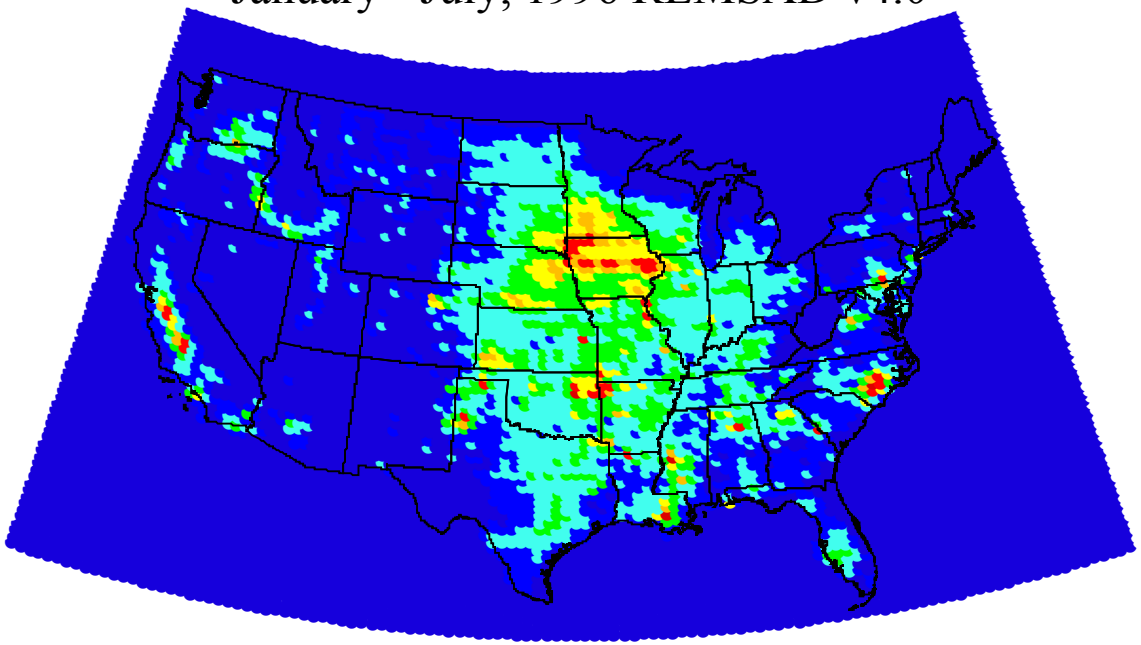


Compared to 2020 projected REMSAD run

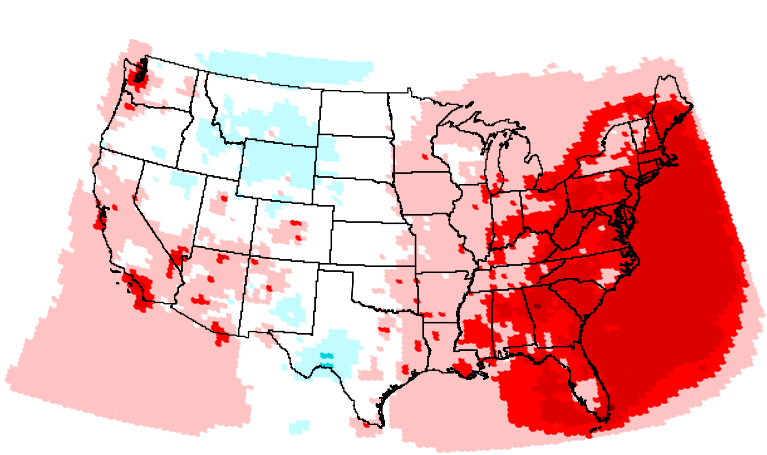
Compared to 2030 projected REMSAD run

Figure 11. Average Ammonia

January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
- 2 - 3
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
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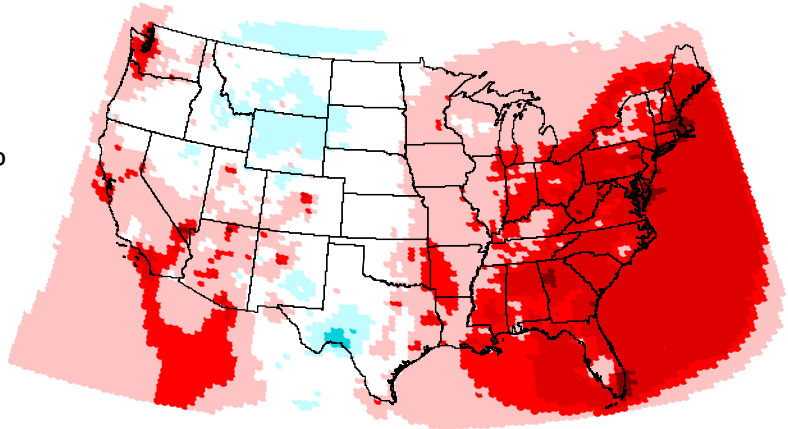
- -30% to -5%
- - 5% to 5%
- 5% to 20%
- 20% to 50%
- 50% to 100%
- > 100%

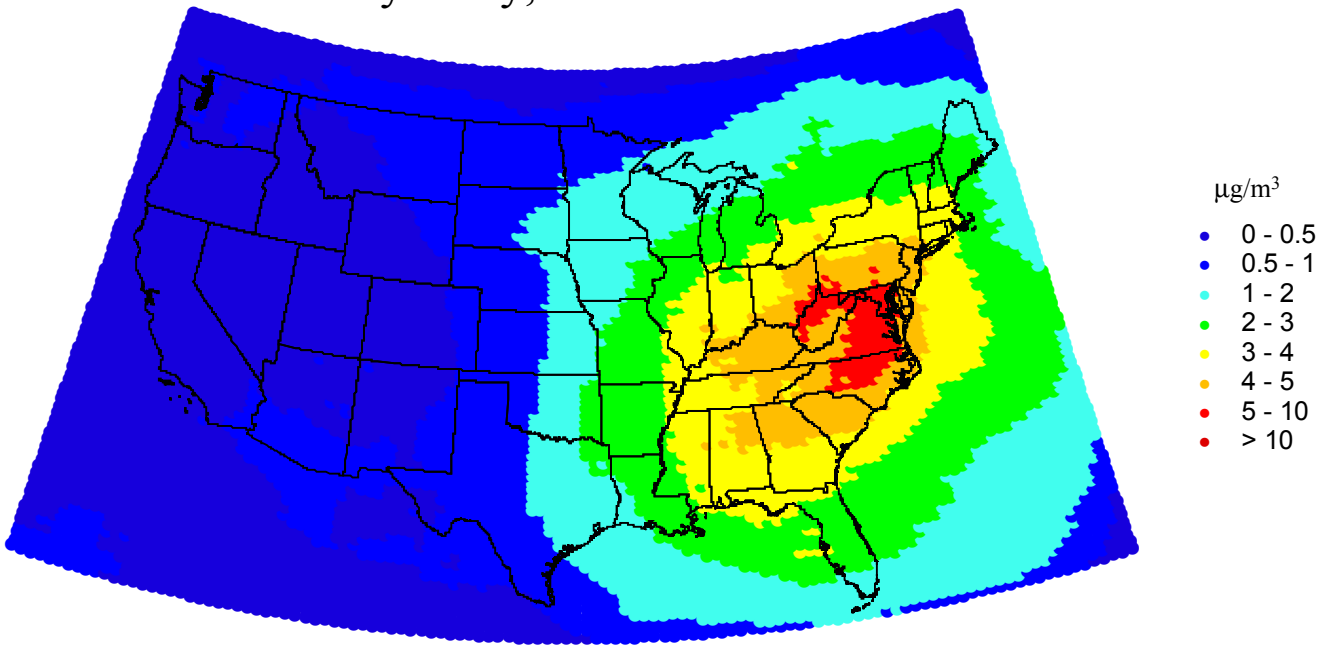


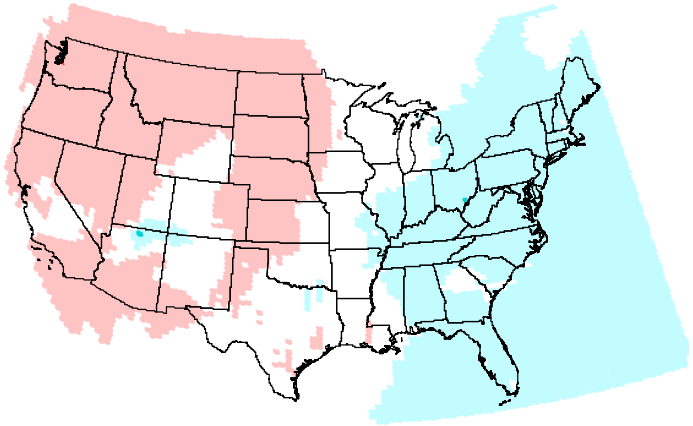
Compared to 2020 projected REMSAD run

Compared to 2030 projected REMSAD run

Figure 12. Average Sulfate Particles

January - July, 1996 REMSAD v4.0





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- 20% to 50%
- 50% to 100%
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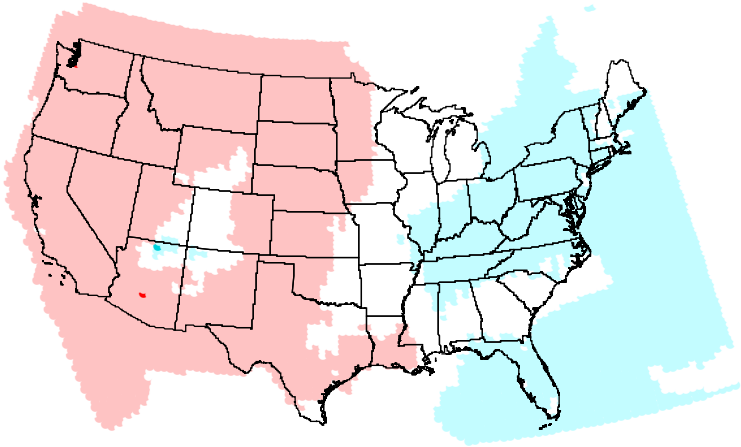
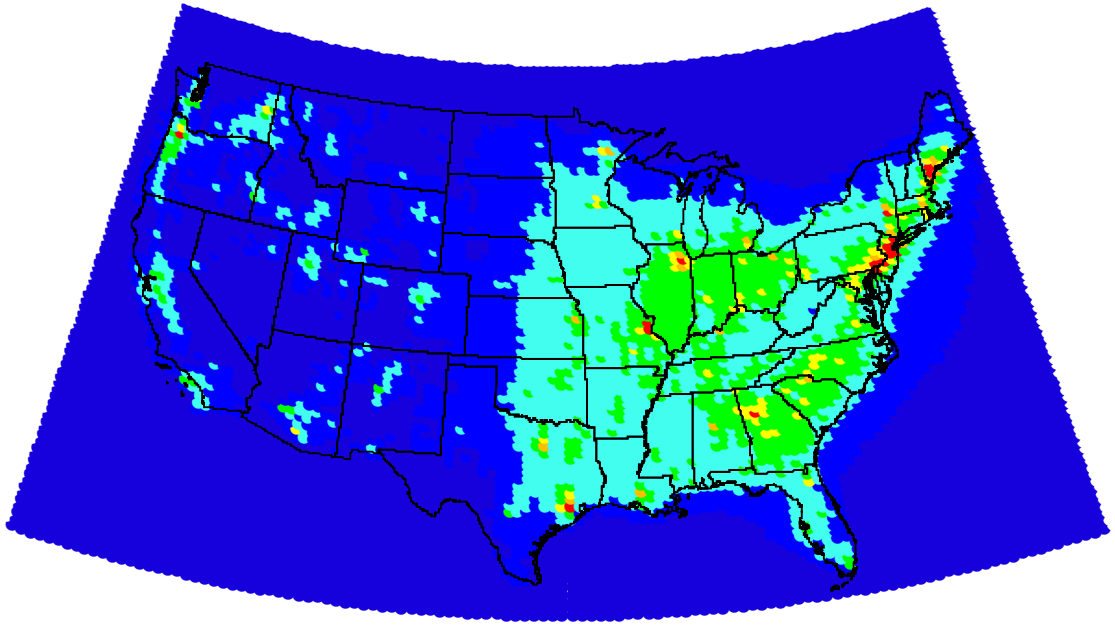


Compared to 2020 projected REMSAD run

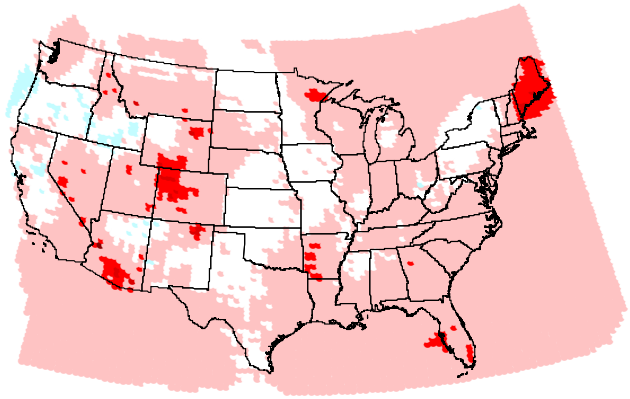
Compared to 2030 projected REMSAD run

Figure 13. Average PMFine Particles

January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
- 2 - 3
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
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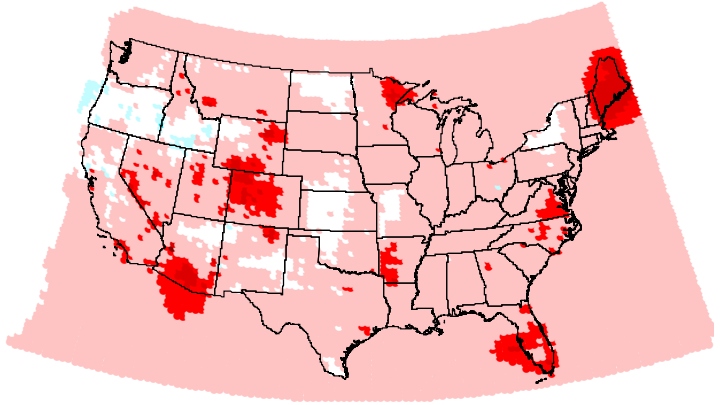
- 50% to 100%
- > 100%

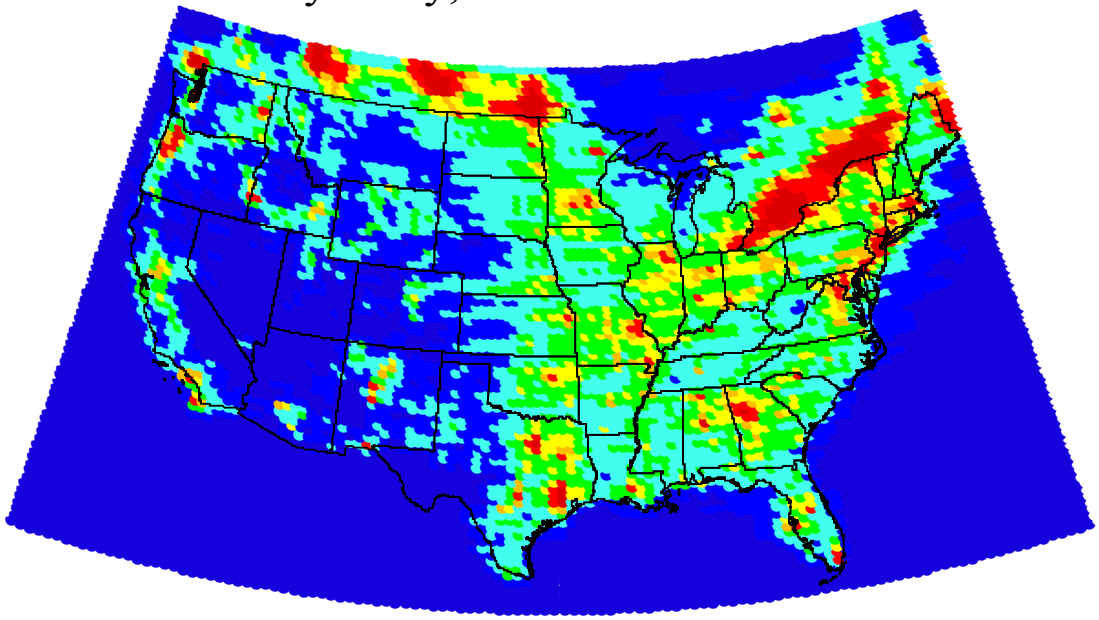


Compared to 2020 projected REMSAD run

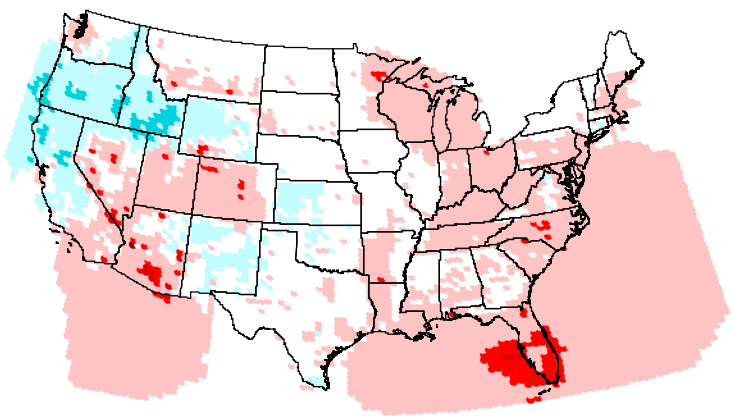
Compared to 2030 projected REMSAD run

Figure 14. Average PMCoarse Particles

January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
- 2 - 3
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
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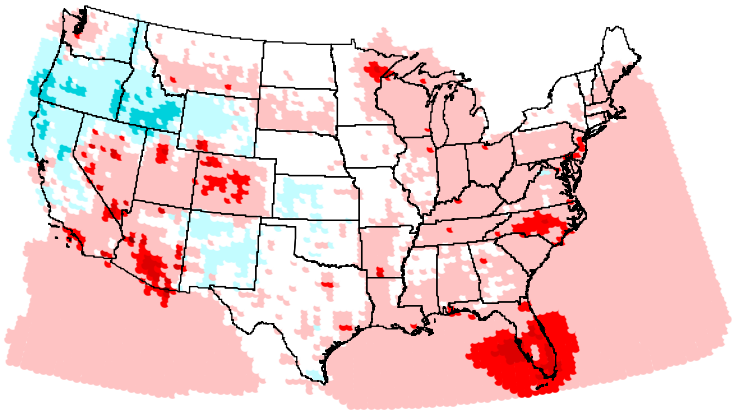
- 50% to 100%
- > 100%

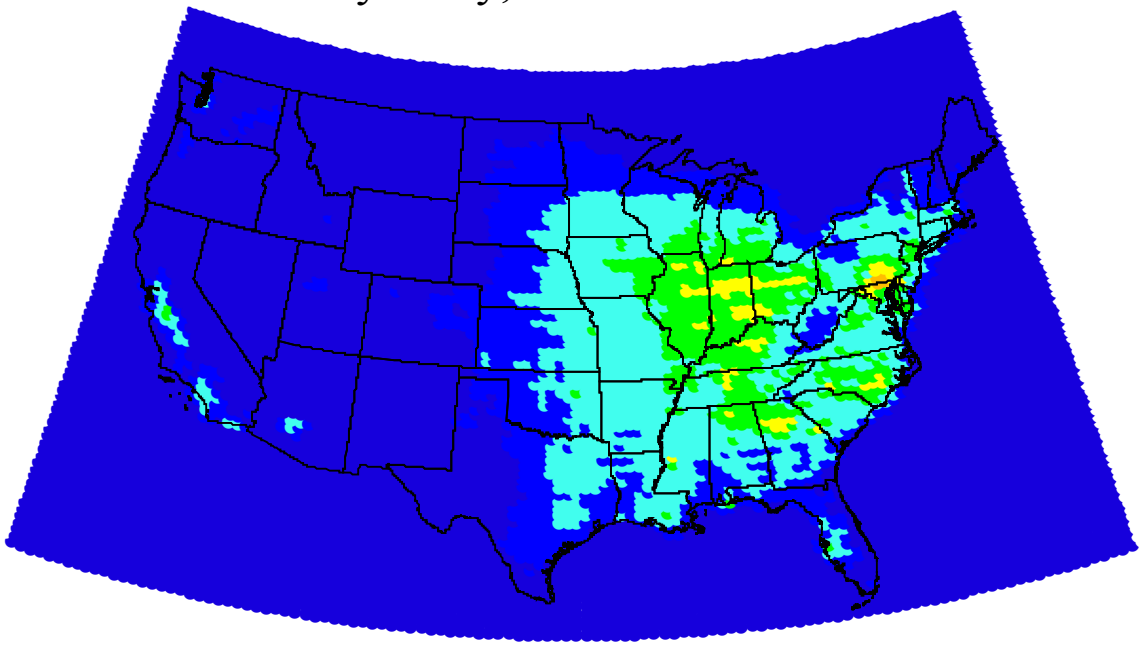


Compared to 2020 projected REMSAD run

Compared to 2030 projected REMSAD run

Figure 15. Average Particulate Nitrate

January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
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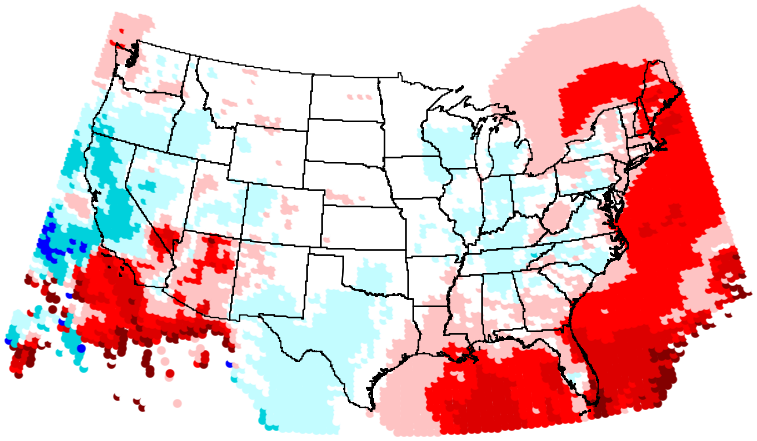
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
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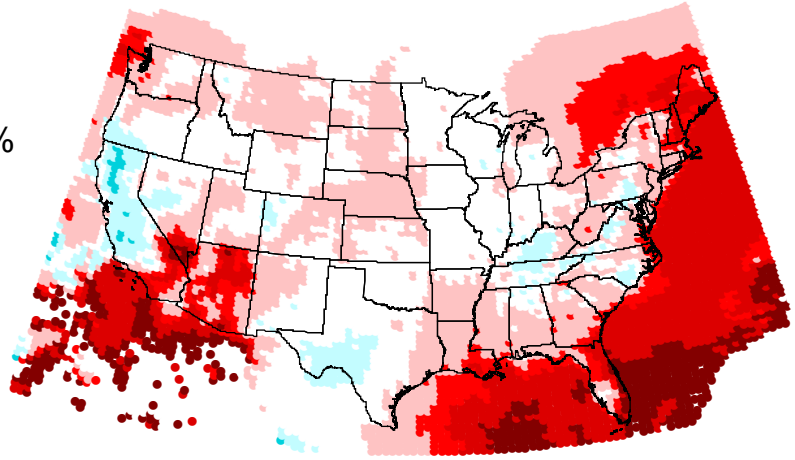
- > 100%

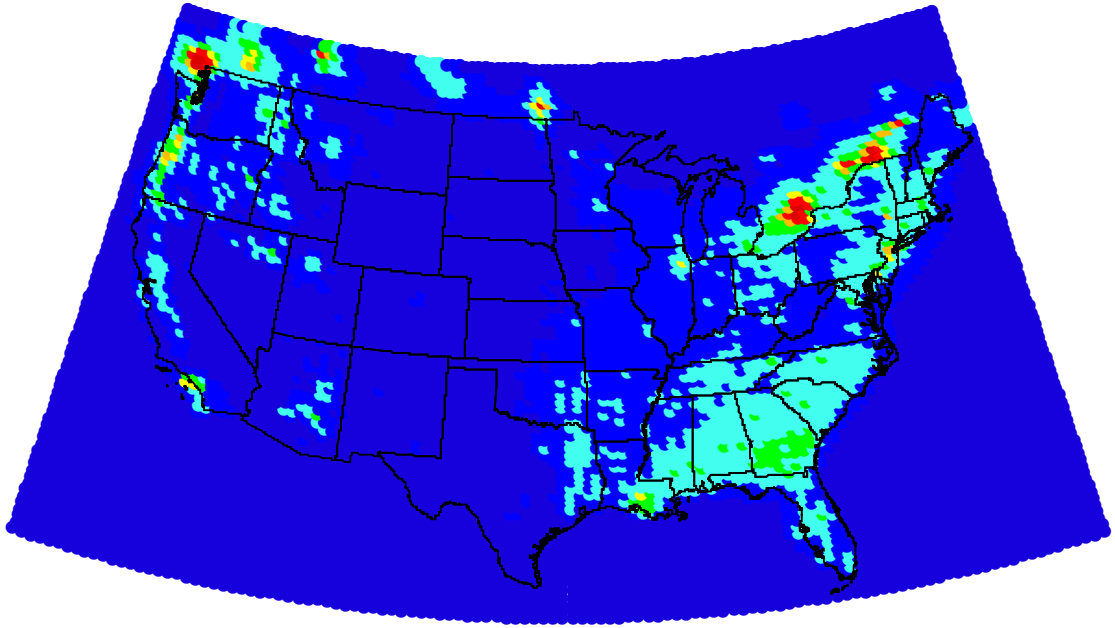


Compared to 2020 projected REMSAD run

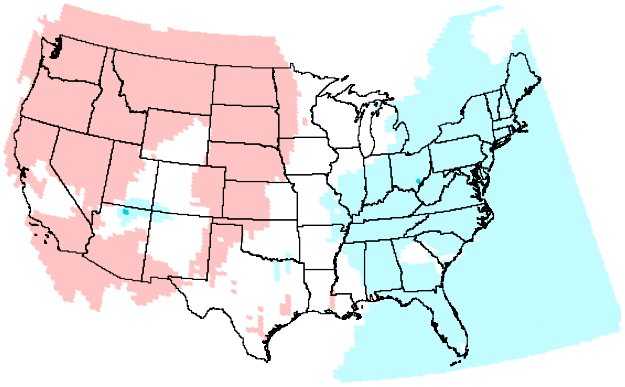
Compared to 2030 projected REMSAD run

Figure 16. Average Organic Particles

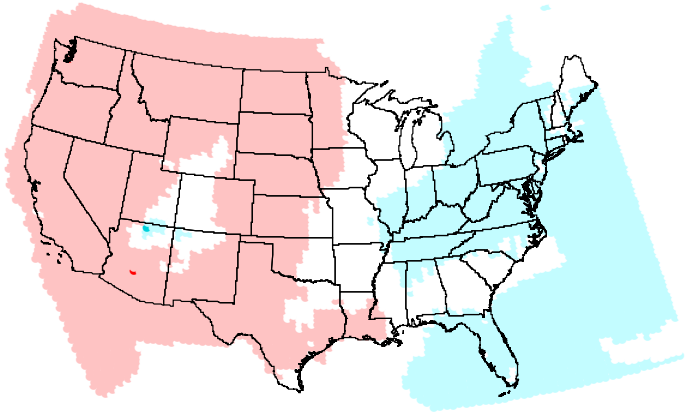
January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
- 2 - 3
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
- > -100 %
- -100% -to -50%
- -50% to -30%
- -30% to -5%
- - 5% to 5%
- 5% to 20%
- 20% to 50%
- 50% to 100%
- > 100%

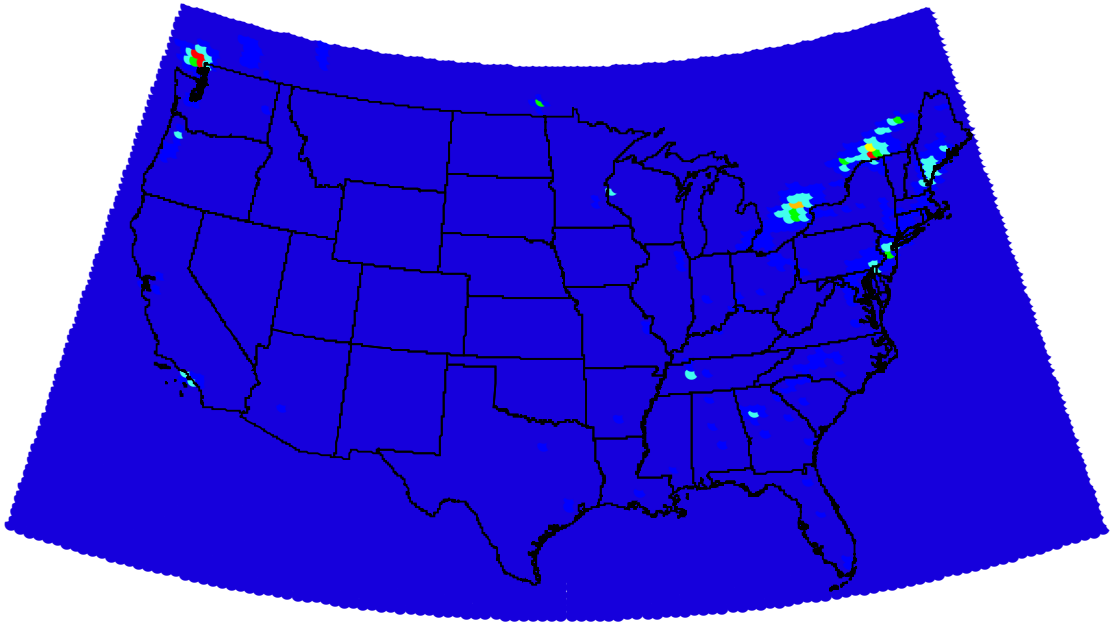


Compared to 2020 projected REMSAD run

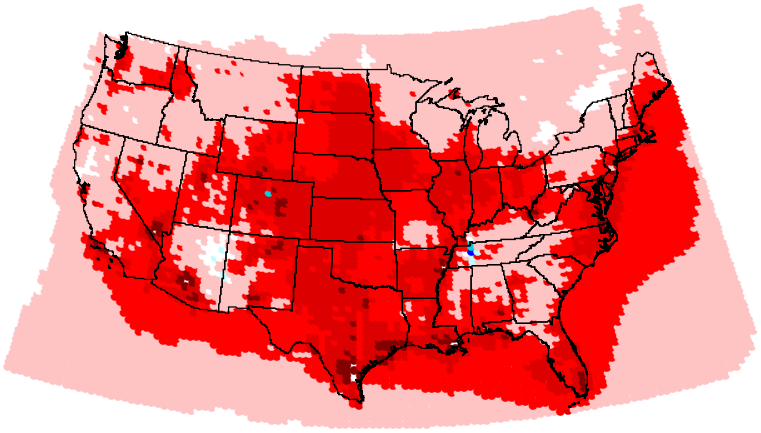
Compared to 2030 projected REMSAD run

Figure 17. Average PEC

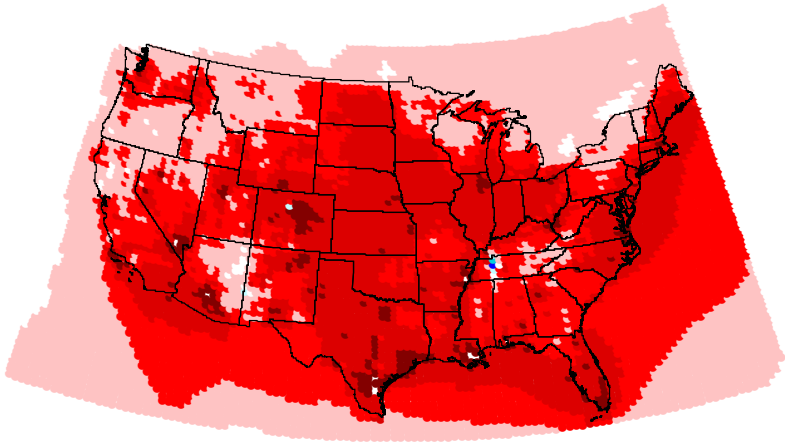
January - July, 1996 REMSAD v4.0



µg/m³
- 0 - 0.5
- 0.5 - 1
- 1 - 2
- 2 - 3
- 3 - 4
- 4 - 5
- 5 - 10
- > 10



Percent Change
- > -100 %
- -100% -to -50%
- -50% to -30%
- -30% to -5%
- - 5% to 5%
- 5% to 20%
- 20% to 50%
- 50% to 100%
- > 100%



Compared to 2020 projected REMSAD run

Compared to 2030 projected REMSAD run

Figure 18. REMSAD v4.0 model results of the Extinction Coefficient.