

IMPROVE data analyses: Trends and Urban Excess

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1990

Acadia, ME

2010

($t = -1.3\% \text{ yr}^{-1}$, $p < 0.01$)



Outline

Trends

- Visualization and trends in worst haze conditions
- Sulfate and SO₂ emissions
- Nitrate, soil

Urban excess

- Sulfate, nitrate, POM, and LAC
- Regional and monthly mean ratios

1990

Great Smoky Mountains, TN

2010

($t = -0.8\% \text{ yr}^{-1}$, $p < 0.01$)

Trends

- Theil regression
- 1989–2010; 2000–2010
- Seasonal and regional
- Urban and rural (sulfate)

1994

Dolly Sods

($t = -1.3\% \text{ yr}^{-1}$, $p = 0.02$)

2010

Trends in Worst Haze Days

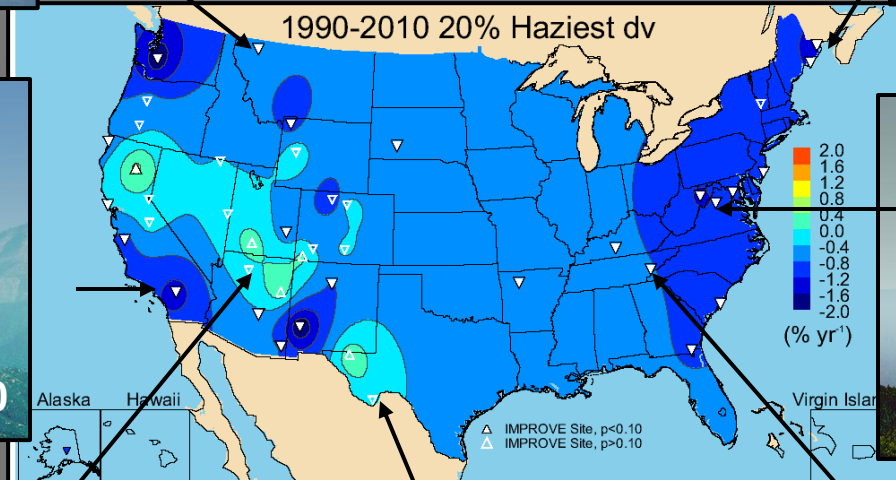
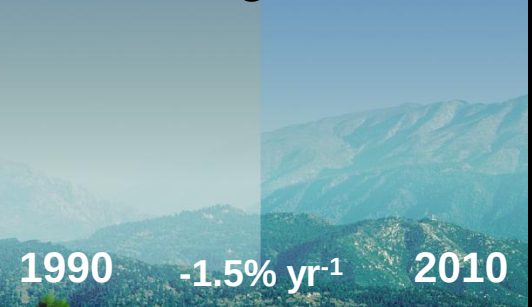
Glacier, MT



Acadia ME



San Geronio, CA



Dolly Sods



Grand Canyon, AZ



Big Bend, TX

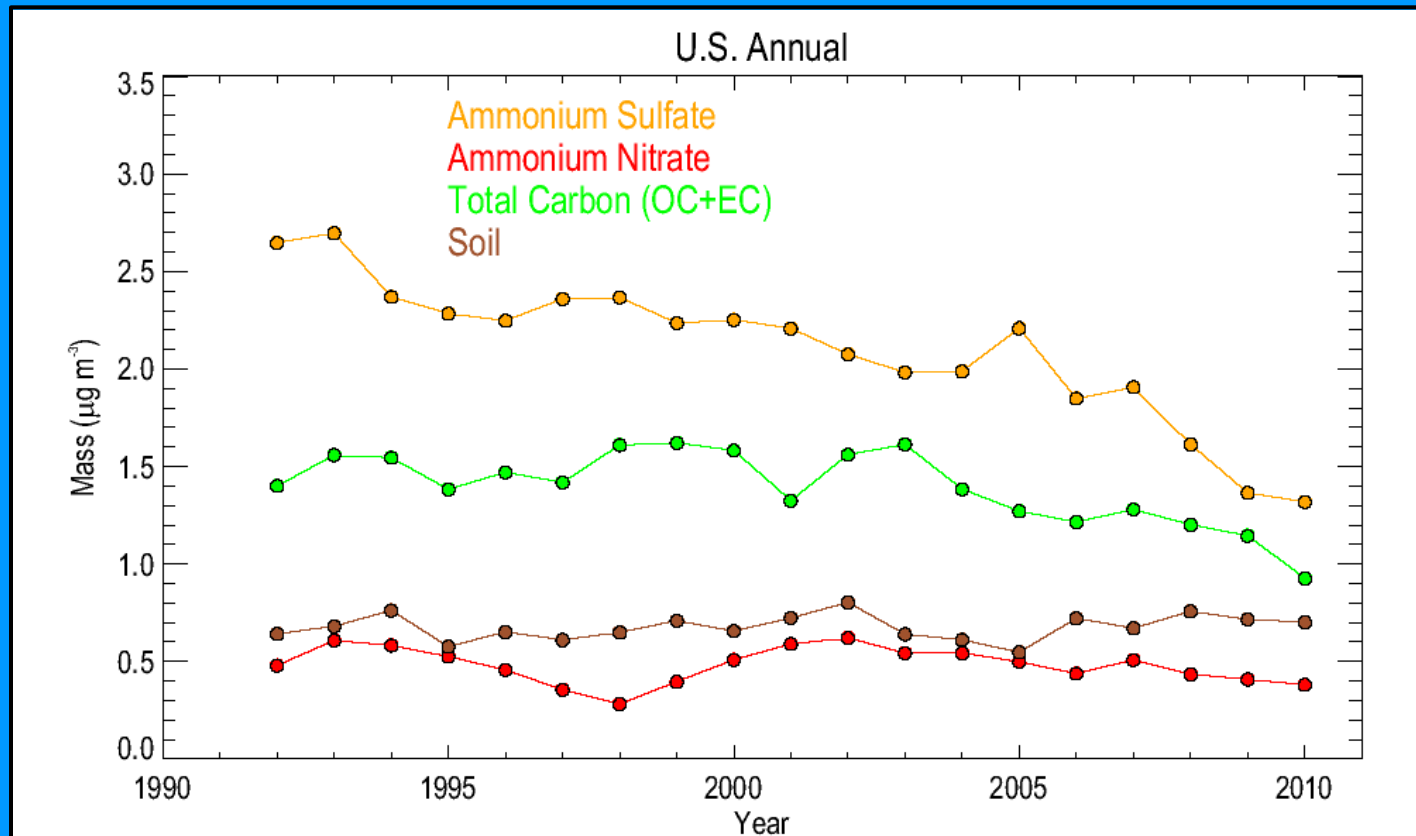


Great Smoky Mountains, TN



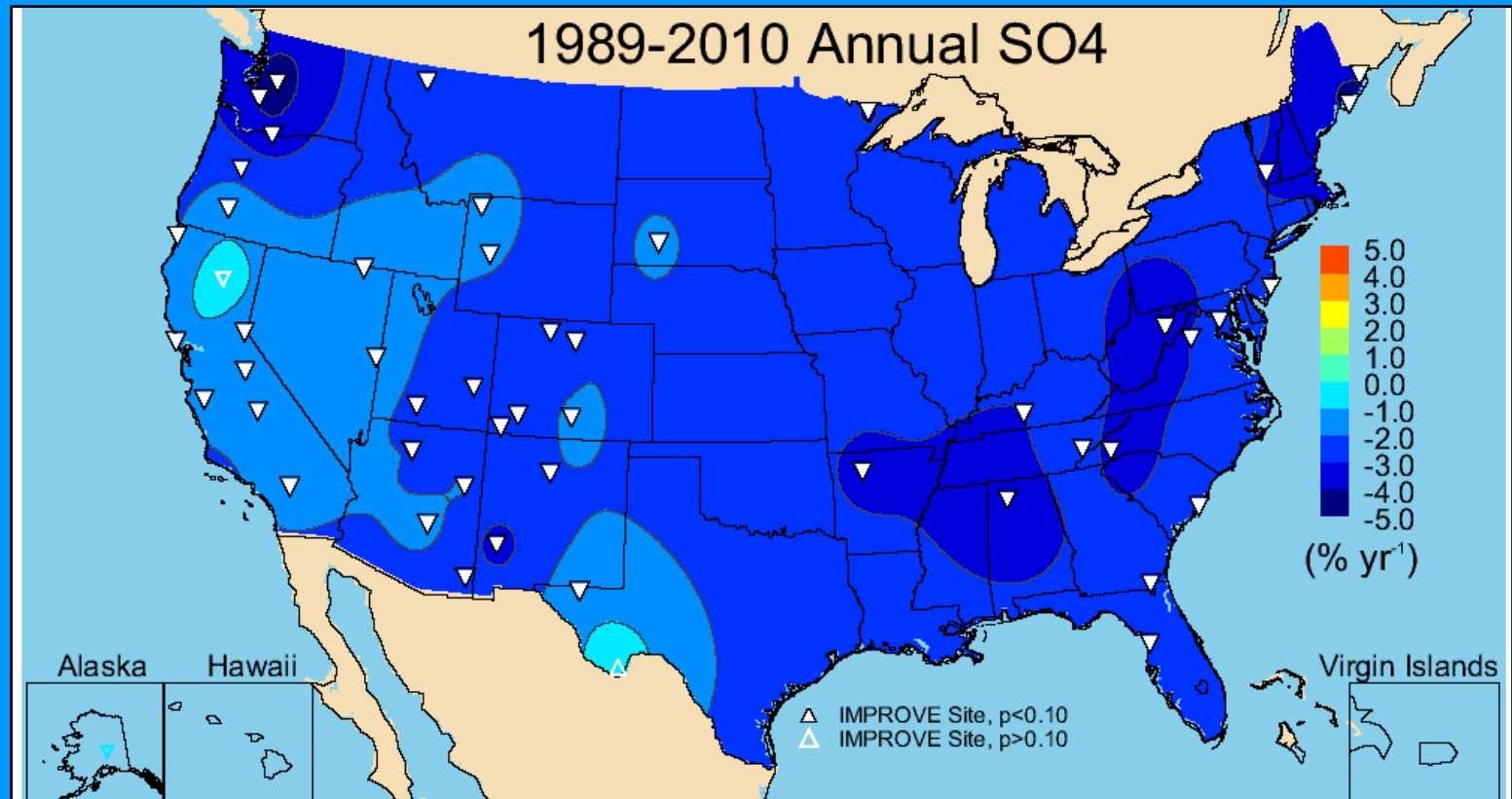
IMPROVE Major Species

U.S. Annual Mean



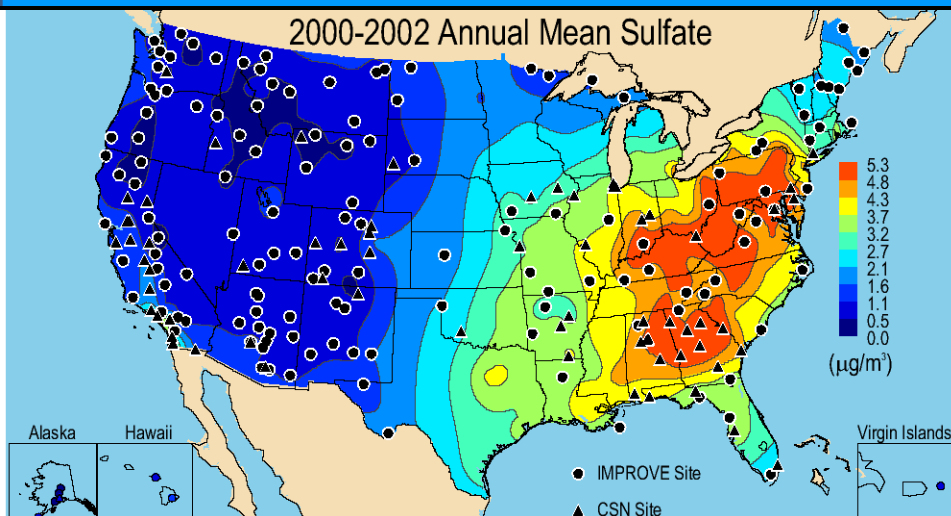
1989-2010 Trends in Annual Mean Sulfate

Widespread decrease in sulfate mass

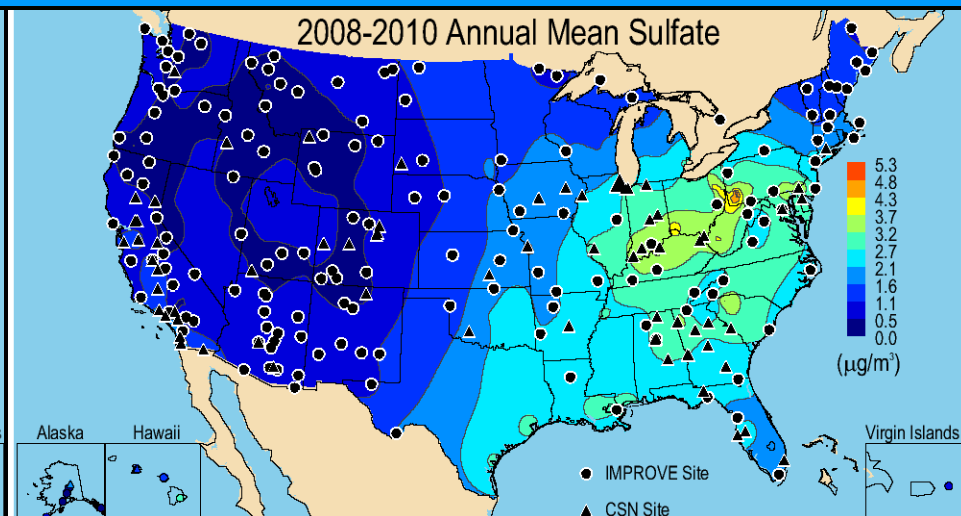


2000-2010 Trends in Annual Mean Sulfate

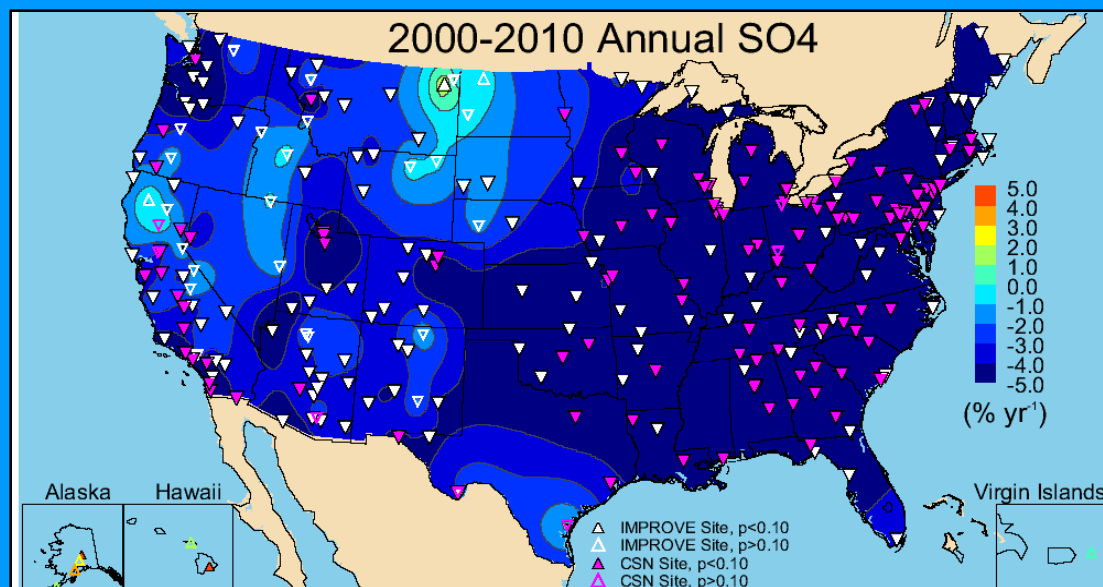
2000-2002 Sulfate Mass



2008-2010 Sulfate Mass

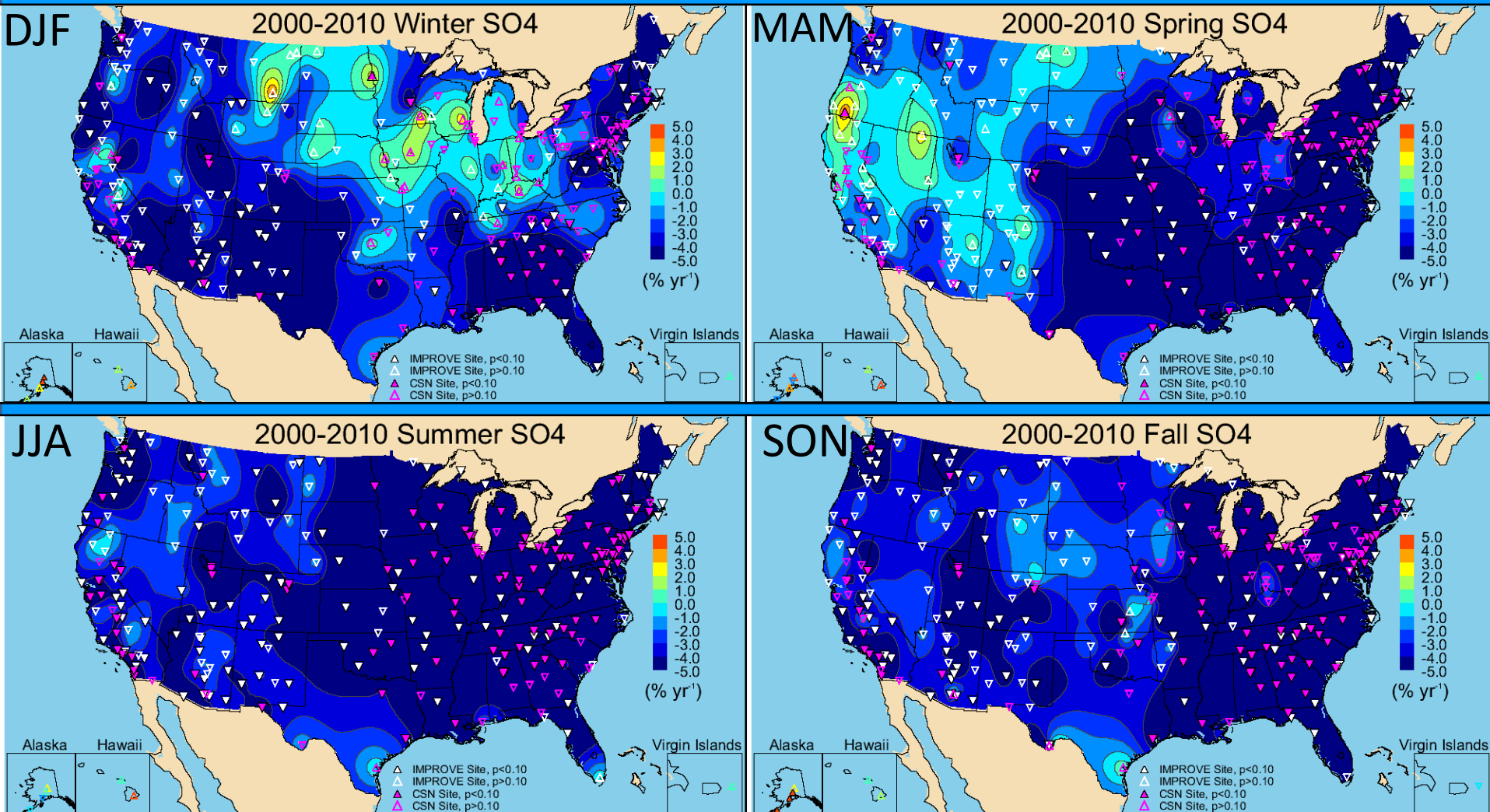


2000-2010 Trends in Annual Mean Sulfate



Hand et al. (2012), accepted, ACP

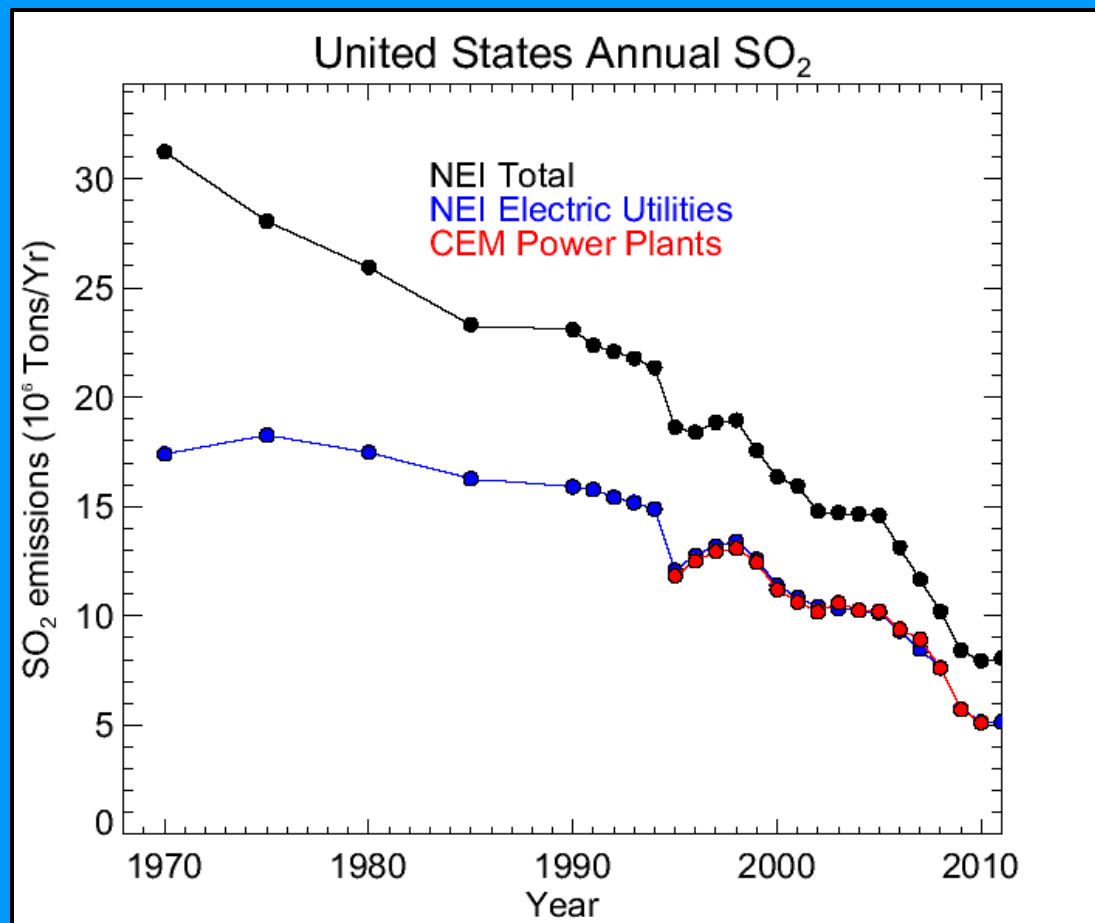
2000-2010 Trends in Seasonal Mean Sulfate



SO₂ Emissions

- National Emissions Inventory (NEI) - Annual Total Emissions (1990-2011) (www.epa.gov/ttnchie1/trends/)
 - All SO₂ sources for annual totals and entire U.S.
- EPA's Air Markets Program–Acid Rain Program (1995-2011) (ampd.epa.gov/ampd/)
 - Continuous emissions monitoring (CEM)- aggregated to monthly, annual, and state level. Power plants only (~65% of total in 2010 according to NEI).

SO₂ Emissions



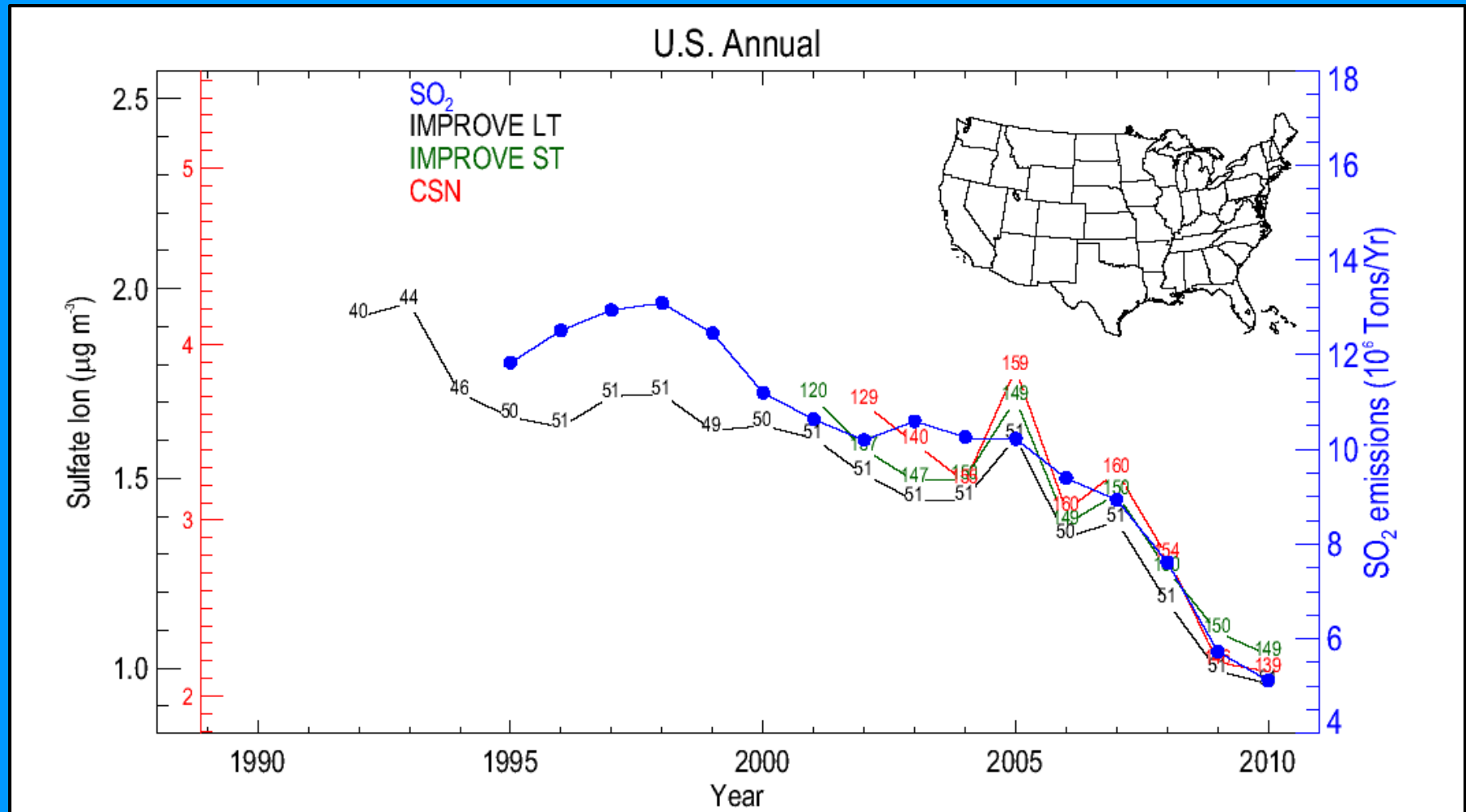
2000-2010 Trends in Total Annual Emissions

NEI: -5.0 % yr⁻¹ (p<0.01)

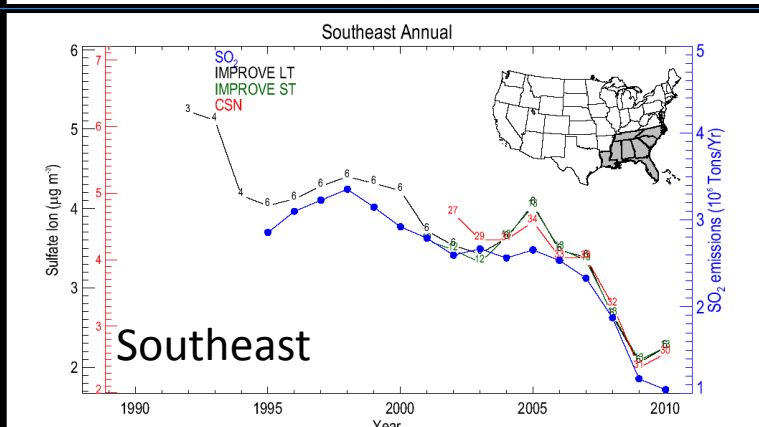
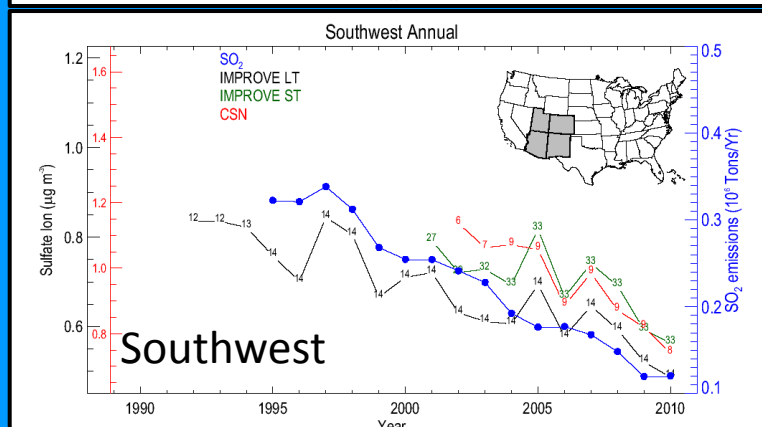
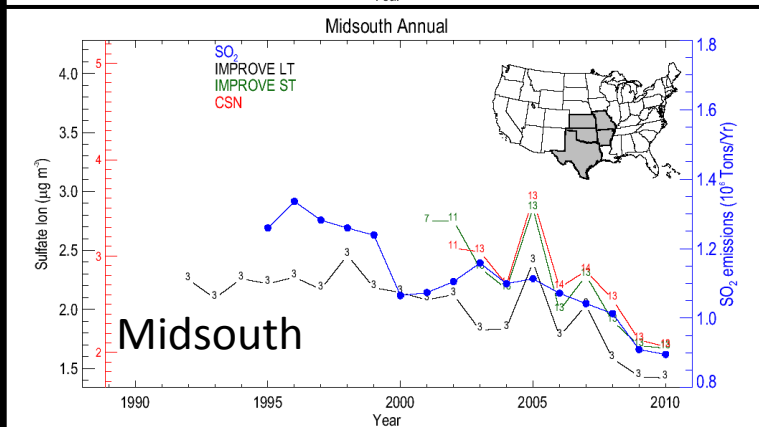
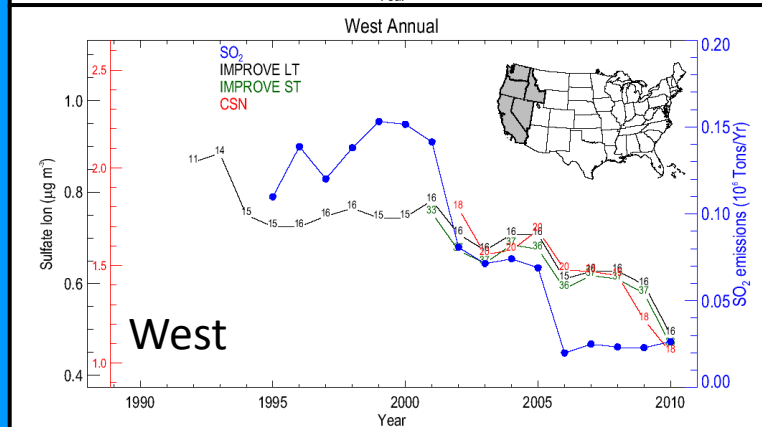
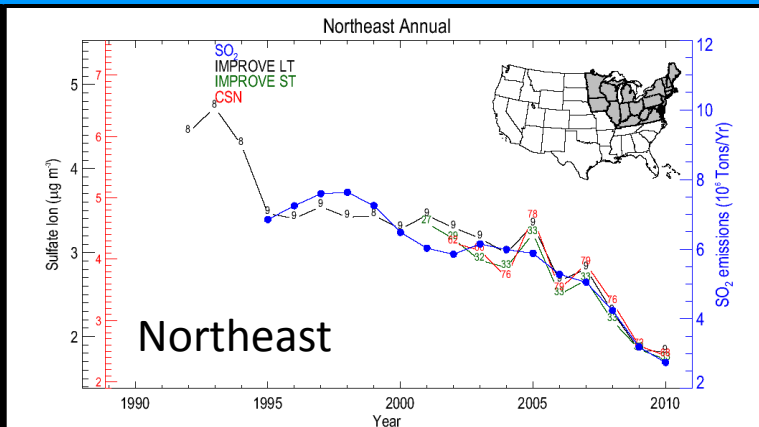
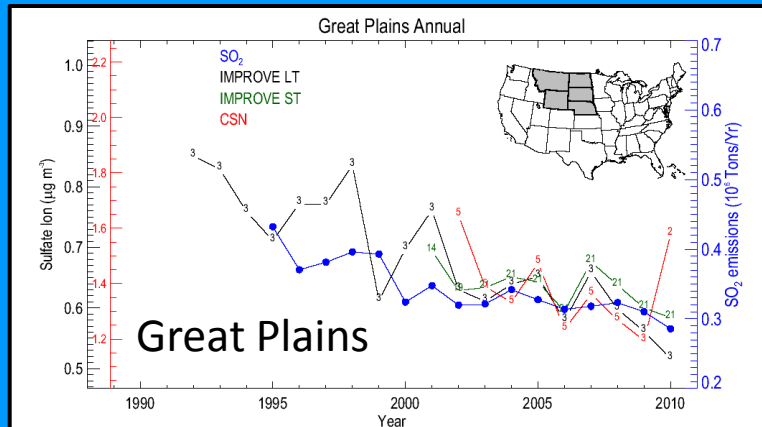
CEM: -4.9 % yr⁻¹ (p<0.01)

CEM SO₂ Emissions and Sulfate Concentrations

U.S. Annual

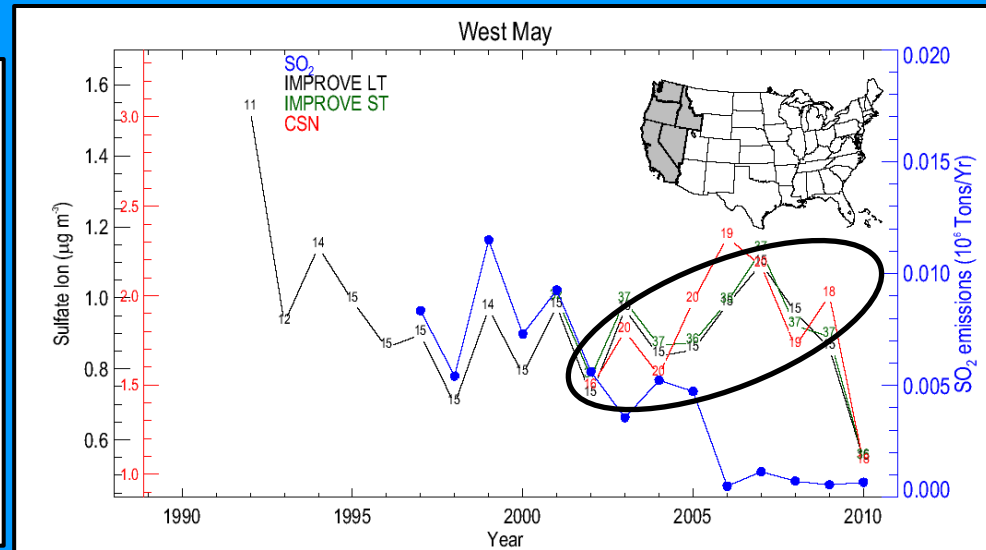
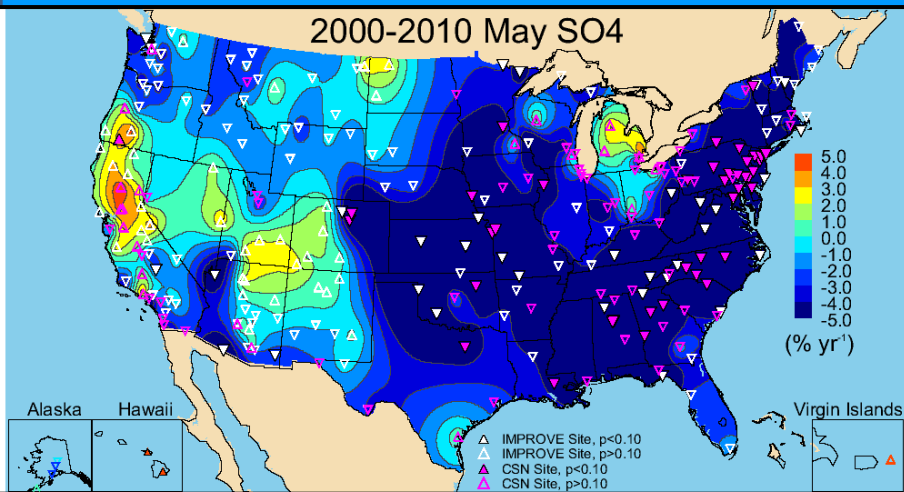


Regional Annual Sulfate and SO₂ emissions

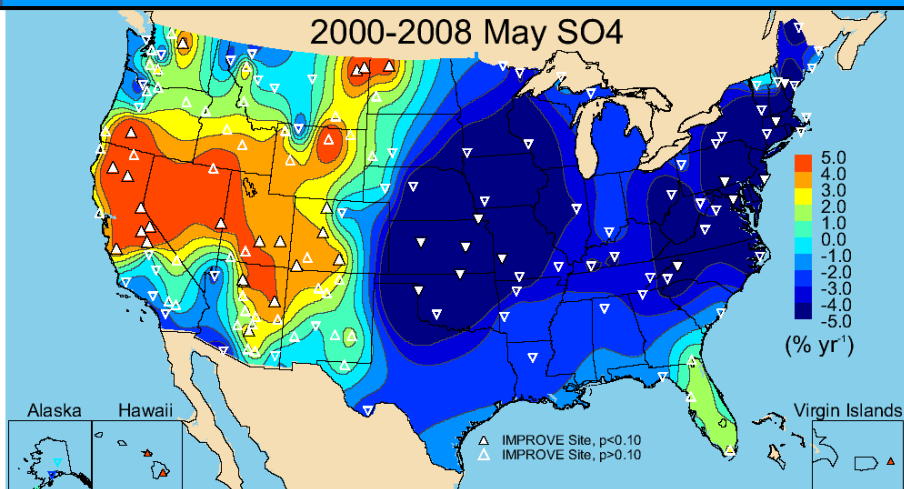


May: Trend Behavior in the West

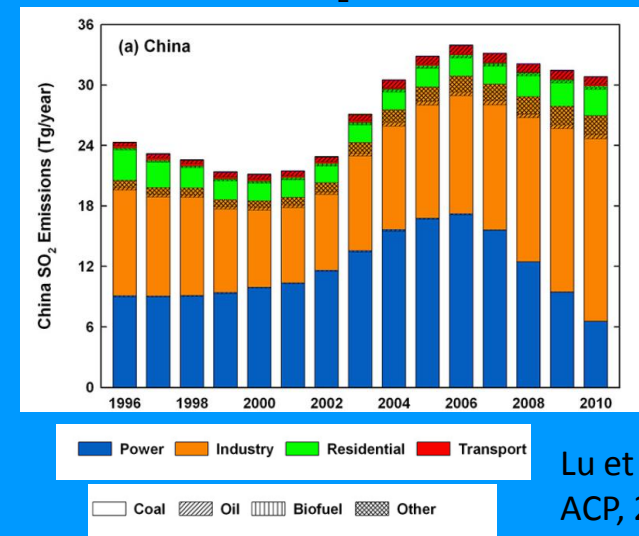
2000-2010 Trend in May Monthly Mean



2000-2008 Trend in May Monthly Mean



Chinese SO₂ Emissions

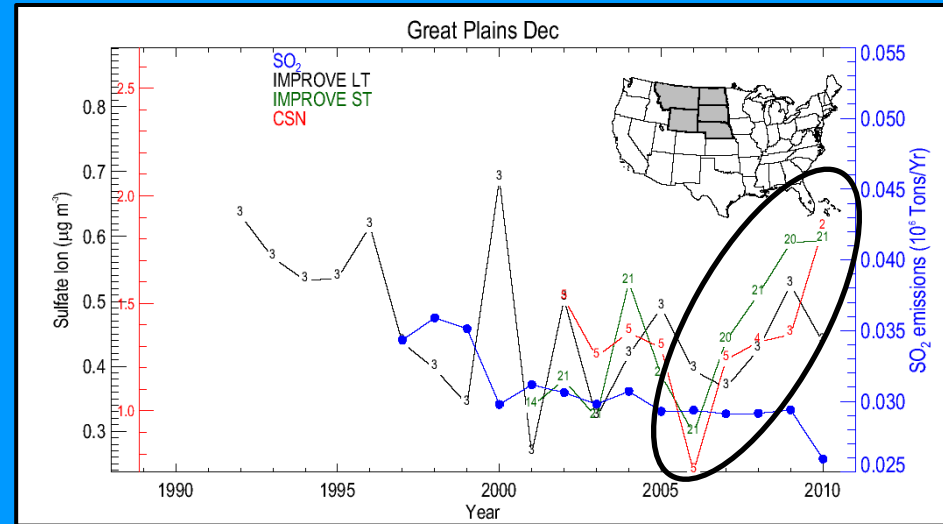
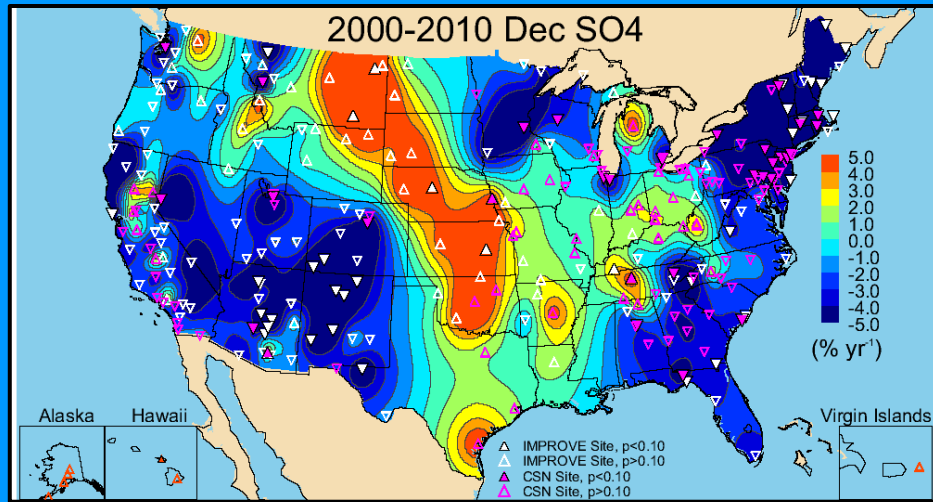


Lu et al.,
ACP, 2011

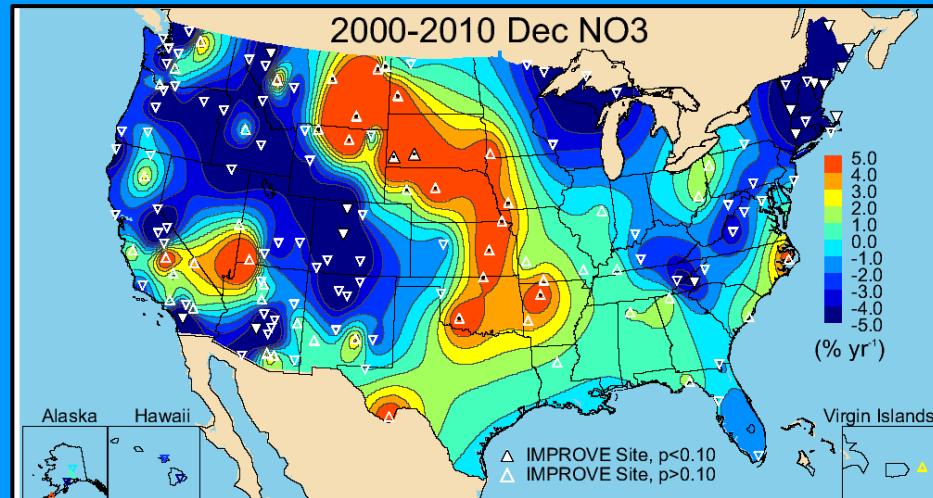
December: Trend Behavior in the Great Plains

December mean concentrations started increasing in 2006

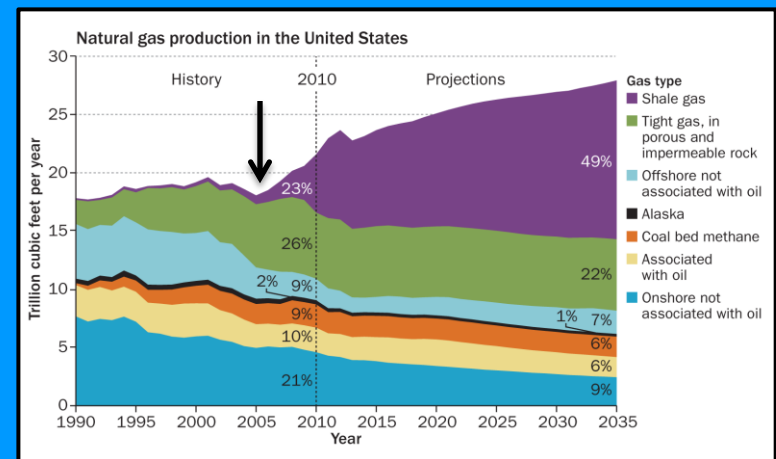
Sulfate December Trends



Nitrate December Trends

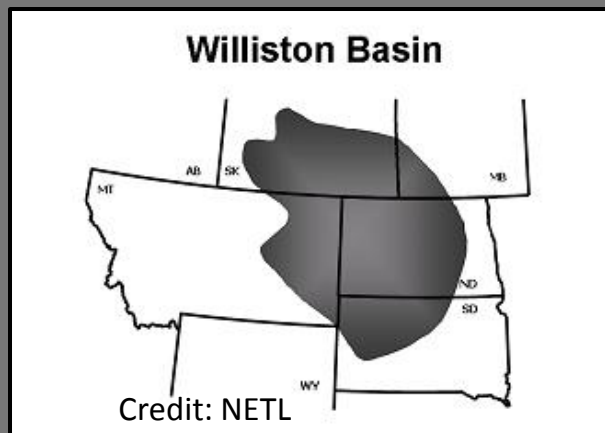


Impacts of oil and gas development?



Source: U.S. Energy Information Administration's Annual Energy Outlook 2012, Science News Sept 8, 2012

Williston Basin, ND

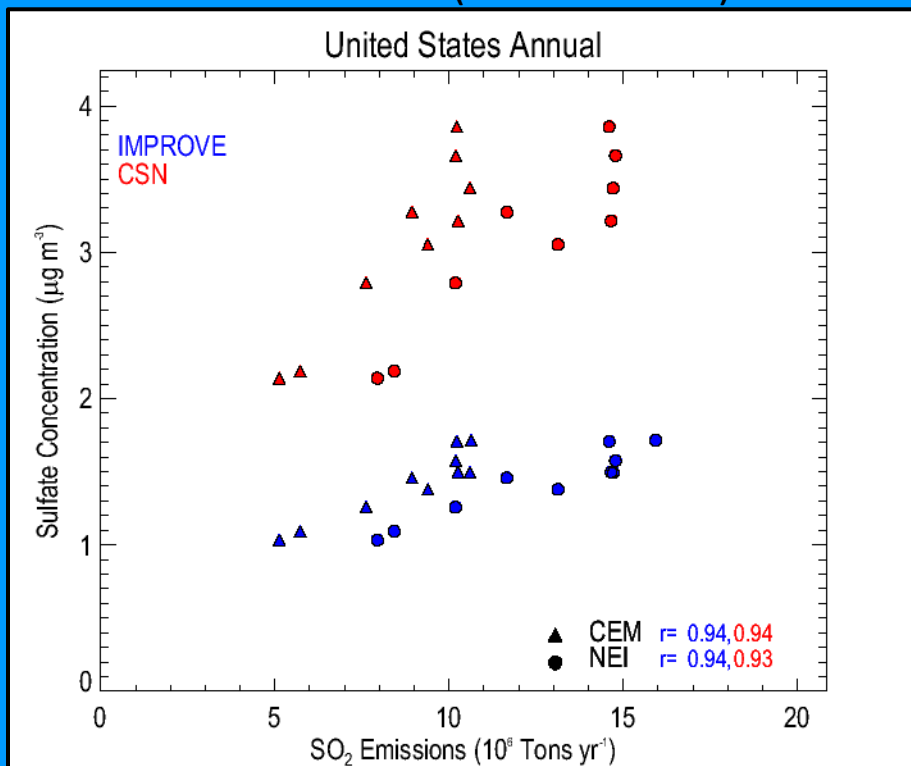


Credit: Bill Malm

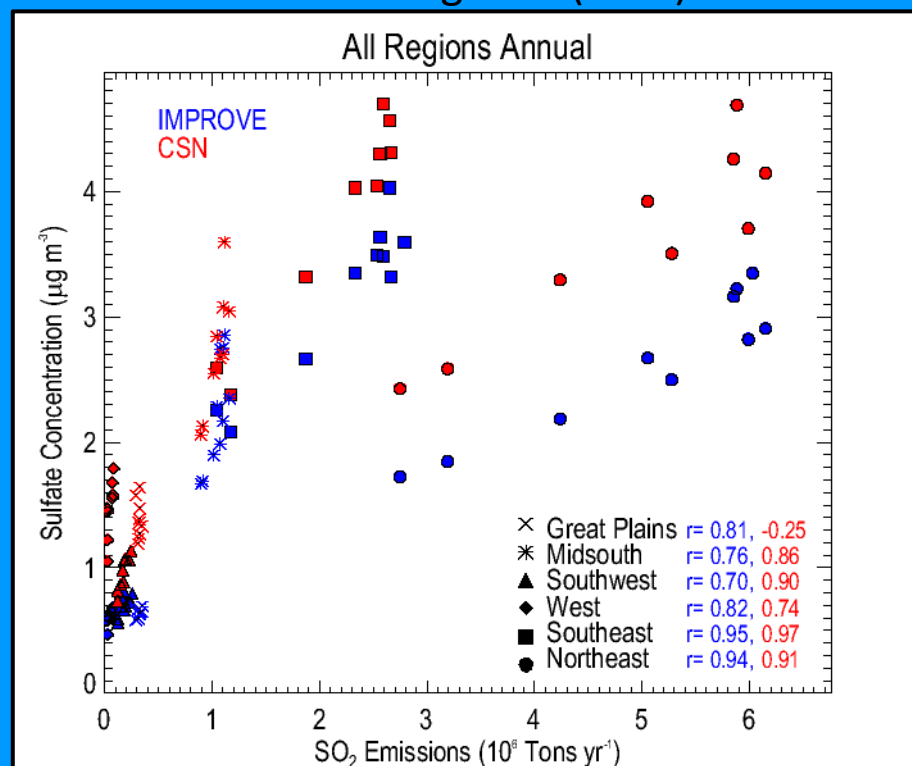
October, 2012

Implications

Annual U.S. (NEI and CEM)



Annual Regional (CEM)



RHR Natural Background levels for

sulfate ion concentrations :

East : $0.17 \mu\text{g m}^{-3}$

West: $0.09 \mu\text{g m}^{-3}$

Based on Theil regression for U.S. using NEI :

IMPROVE: $0.39 \pm 0.12 \mu\text{g m}^{-3}$

CSN: $0.66 \pm 0.24 \mu\text{g m}^{-3}$

Implications

Sulfate:

- On an annual basis, strategies for reducing SO_2 from power plants have been successful in lowering particulate sulfate in the atmosphere, especially for the eastern U.S.
- For certain regions and seasons, factors other than known local emissions have had significant impacts on sulfate concentrations.

1989

Point Reyes, CA

($t = -0.4\% \text{ yr}^{-1}$, $p = 0.02$)

2010

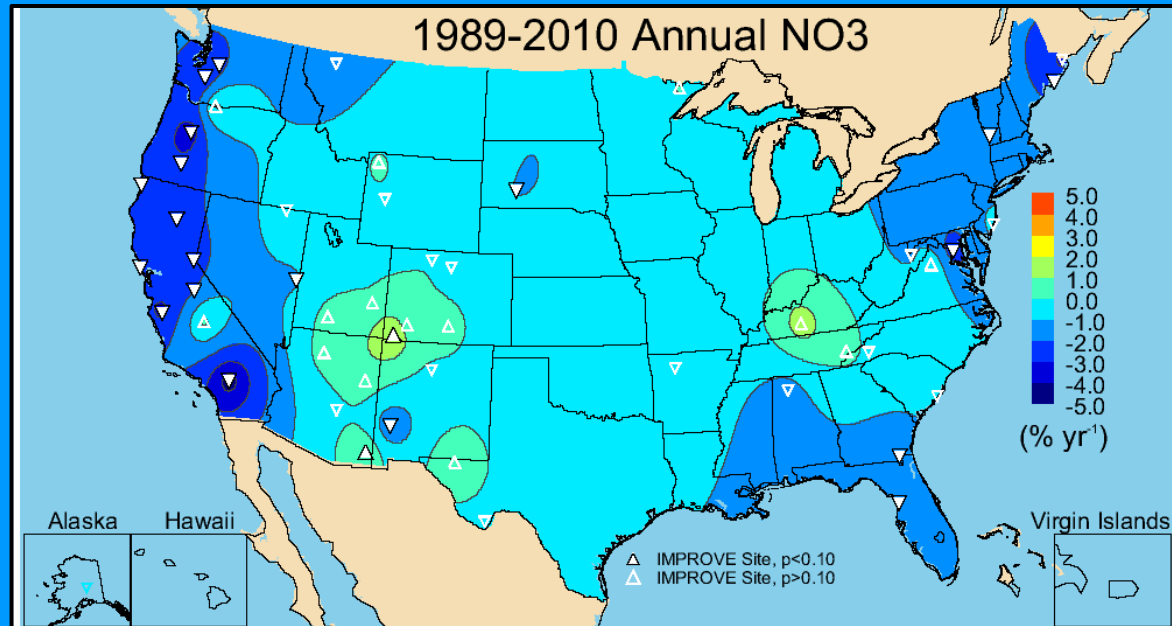
NITRATE

Williston Basin, ND

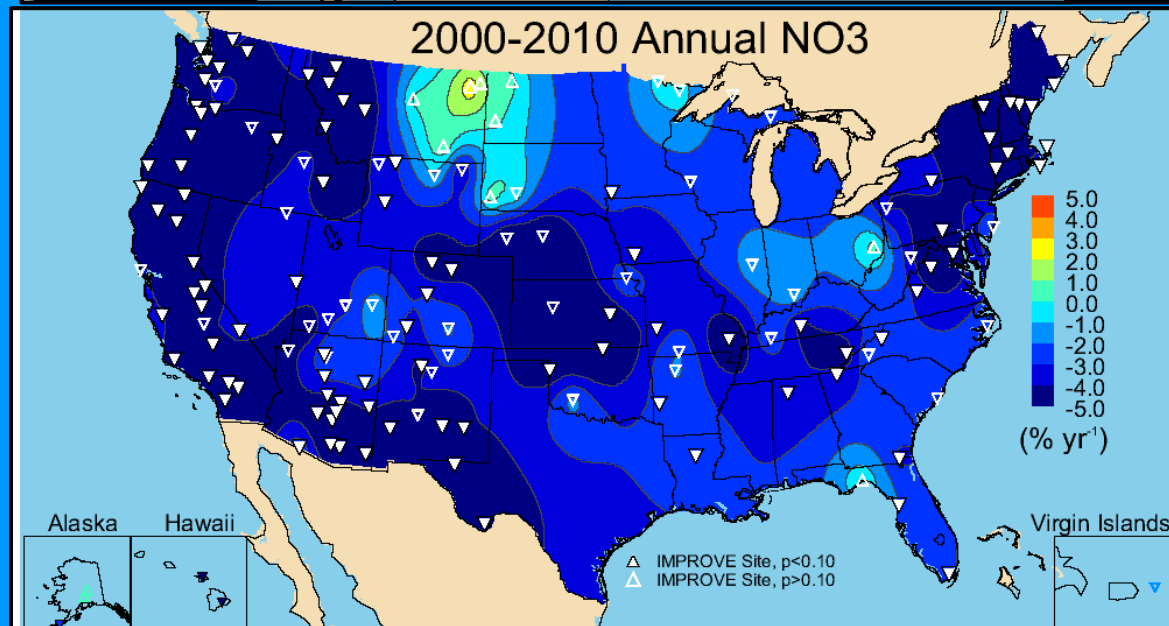


Trends in Annual Mean Nitrate

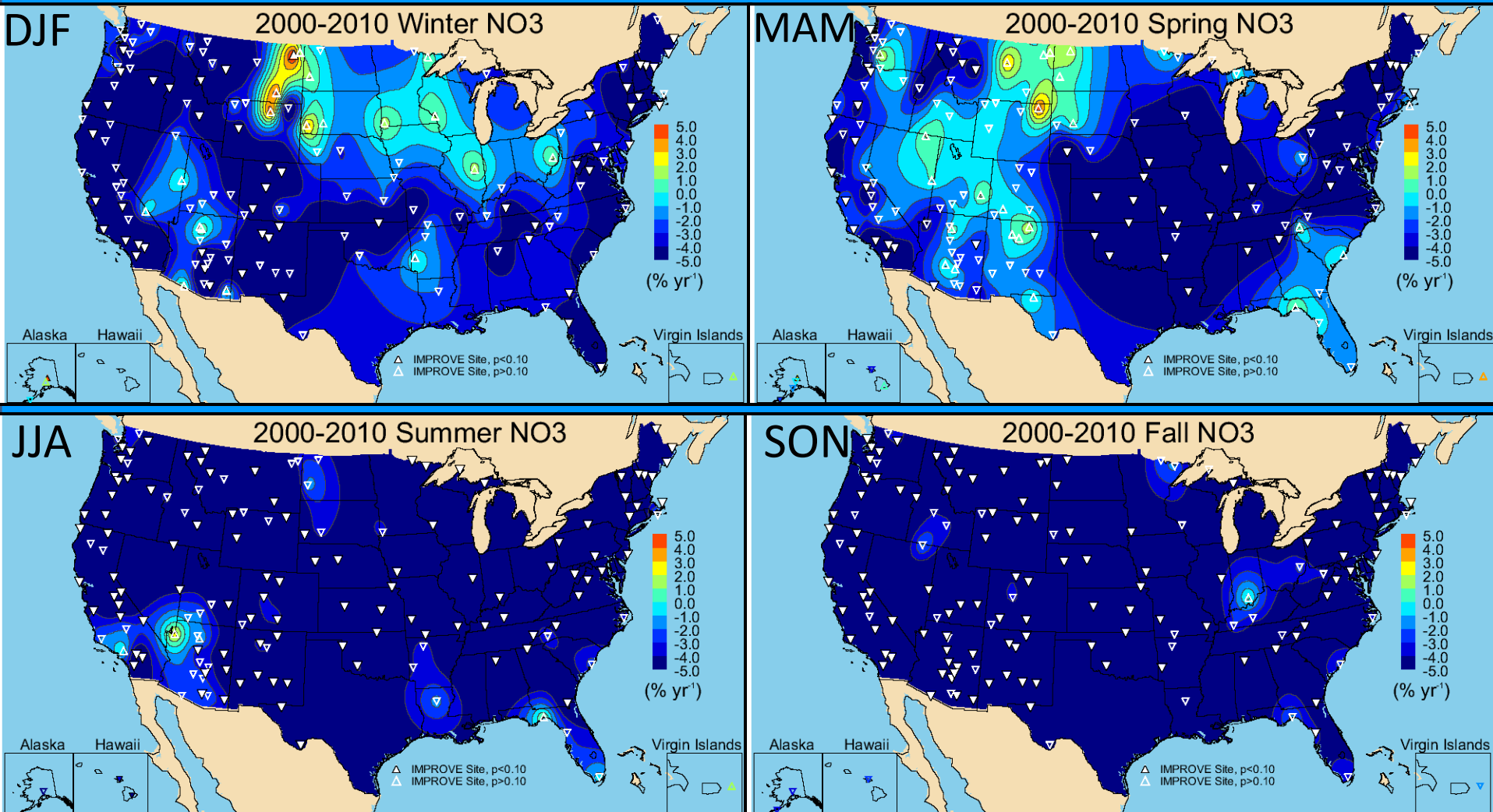
IMPROVE
1989-2010



IMPROVE
2000-2010



2000-2010 Trends in Seasonal Mean Nitrate



SOIL

March, 2012, El Paso, TX
($\text{PM}_{10} > 5000 \mu\text{g m}^{-3}$)

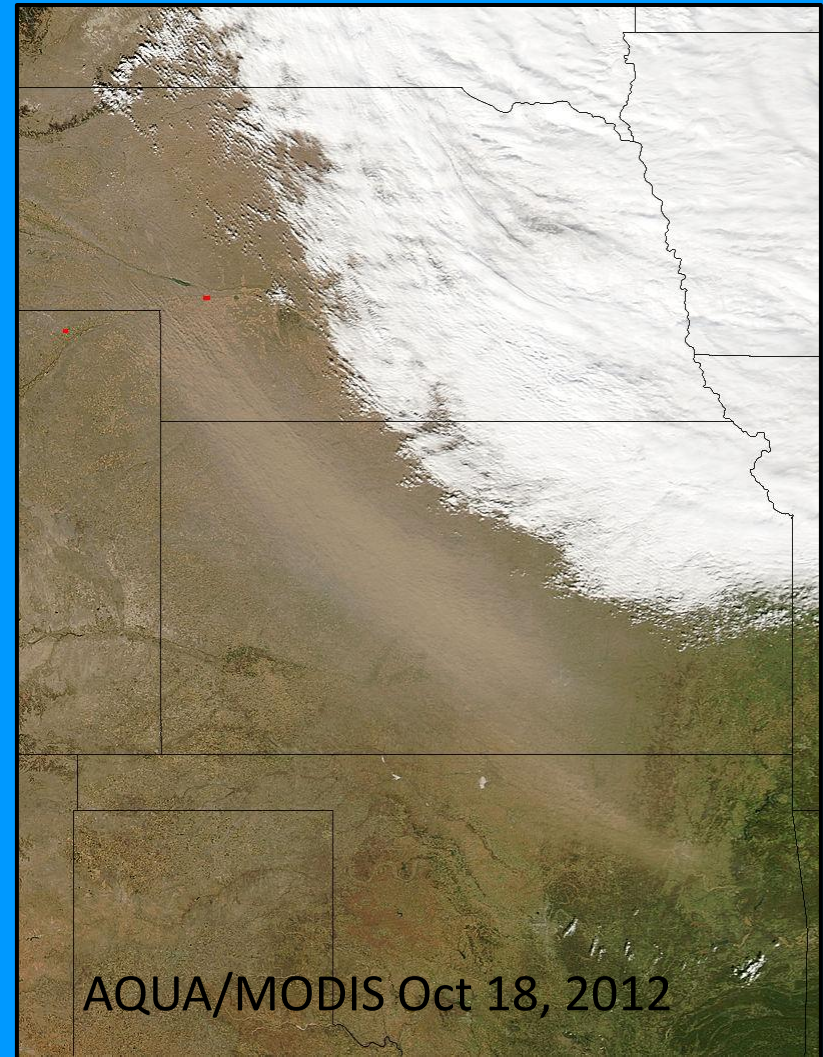
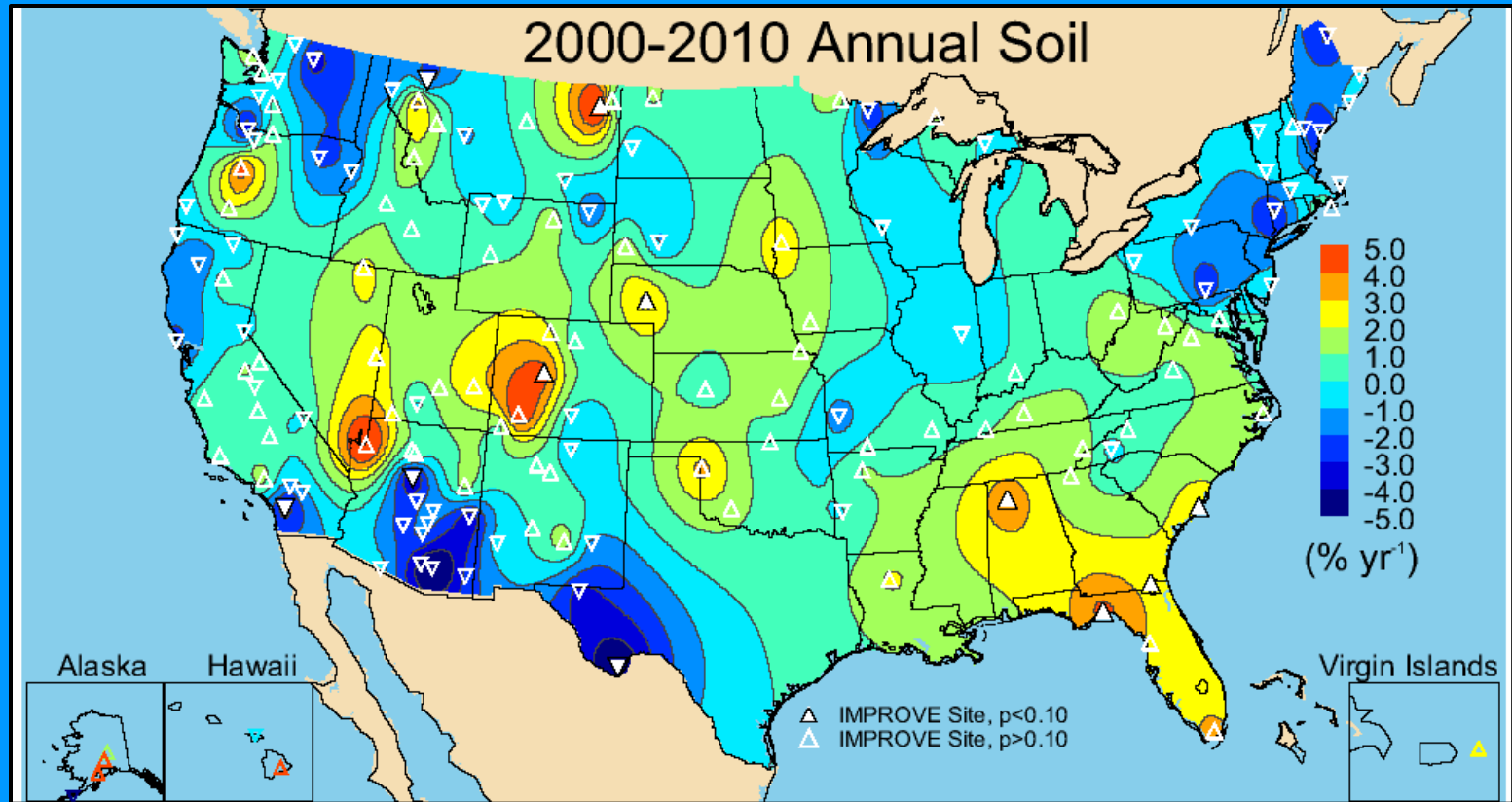


Photo by Joseph Rogash, NOAA-NWS, courtesy of Tom Gill, UTEP

2000-2010 Trends in Annual Mean Soil



Trends in soil are much more variable relative to other species

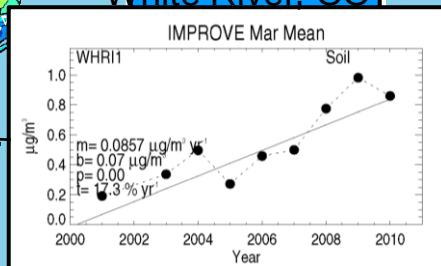
$$\text{Soil} = 2.20[\text{Al}] + 2.49[\text{Si}] + 1.63[\text{Ca}] + 2.42[\text{Fe}] + 1.94[\text{Ti}]$$

SOIL

IRON

2000-2010 Mar Soil

White River, CO

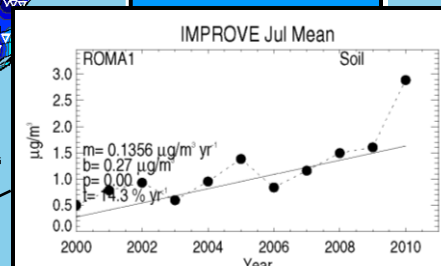


2000-2010 Mar Fe

5.0
4.0
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
-4.0
-5.0
(% yr⁻¹)

2000-2010 Jul Soil

Cape Romain, SC

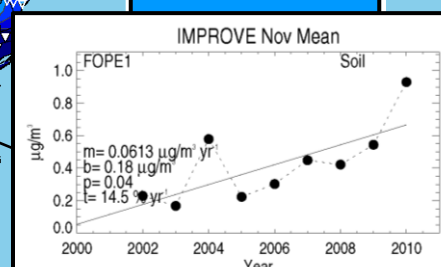


2000-2010 Jul Fe

5.0
4.0
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
-4.0
-5.0
(% yr⁻¹)

2000-2010 Nov Soil

Fort Peck, MT

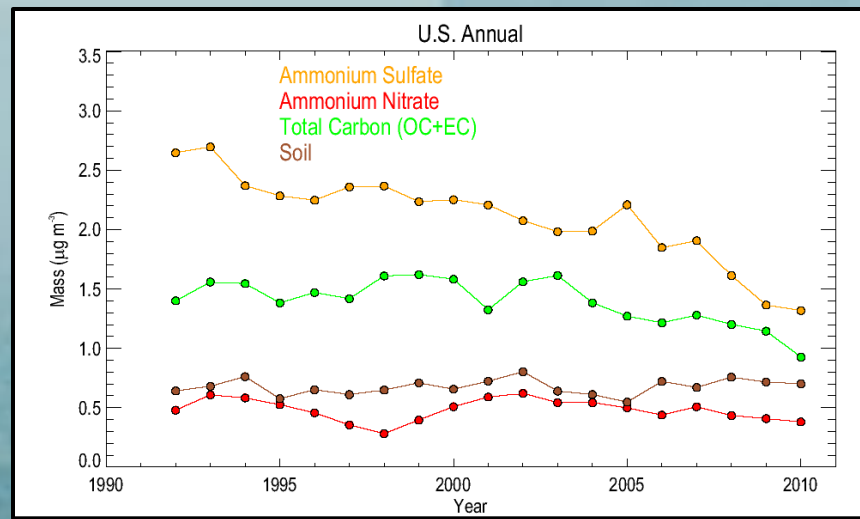


2000-2010 Nov Fe

5.0
4.0
3.0
2.0
1.0
0.0
-1.0
-2.0
-3.0
-4.0
-5.0
(% yr⁻¹)

Implications

- Nitrate- Potential contributions from oil and gas development and international transport
- Soil- Drought, land use change, anthropogenic sources, long-range transport



Linville Gorge, NC

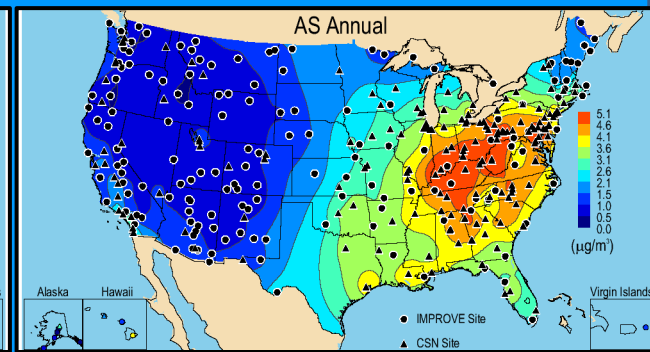
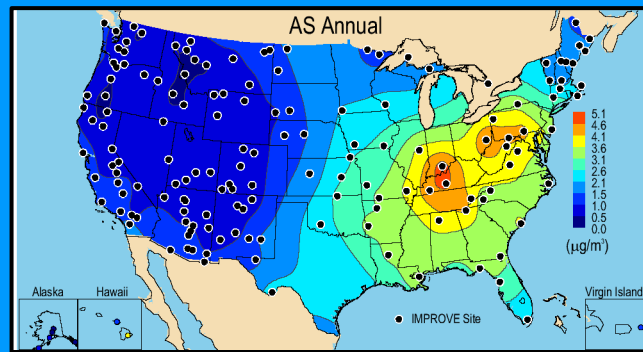
2001

2010

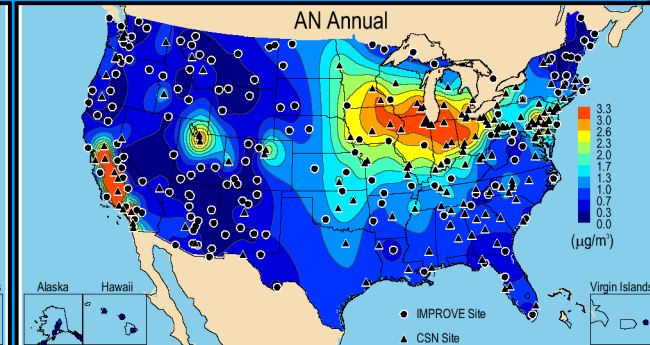
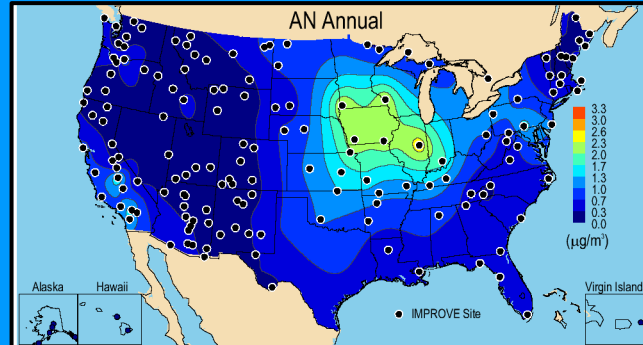
($t = -2.5\% \text{ yr}^{-1}$, $p = 0.095$)

Urban Excess:

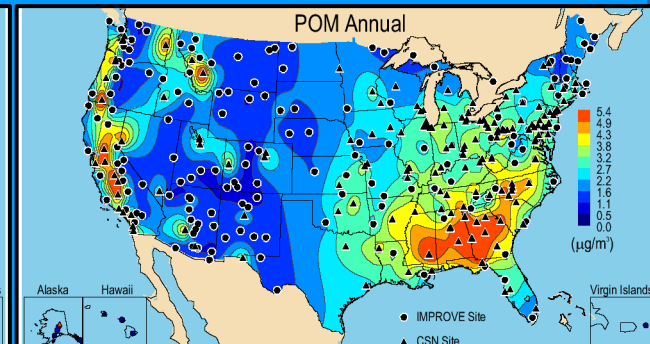
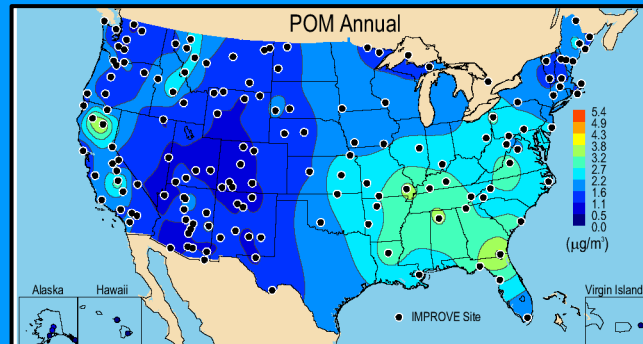
Ammonium Sulfate



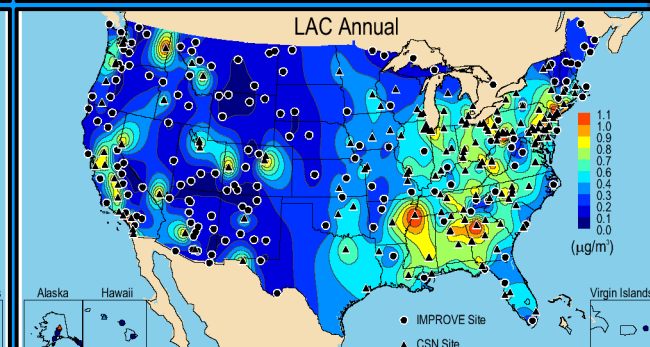
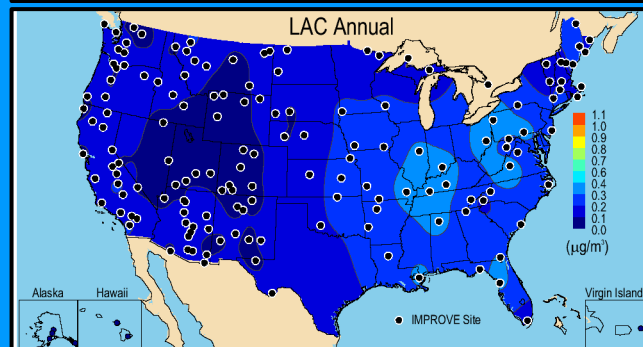
Ammonium Nitrate



Particulate Organic Matter



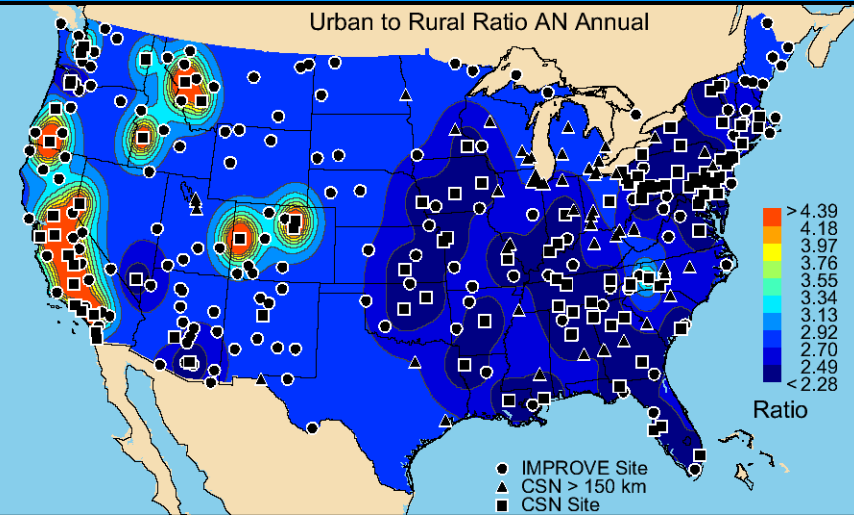
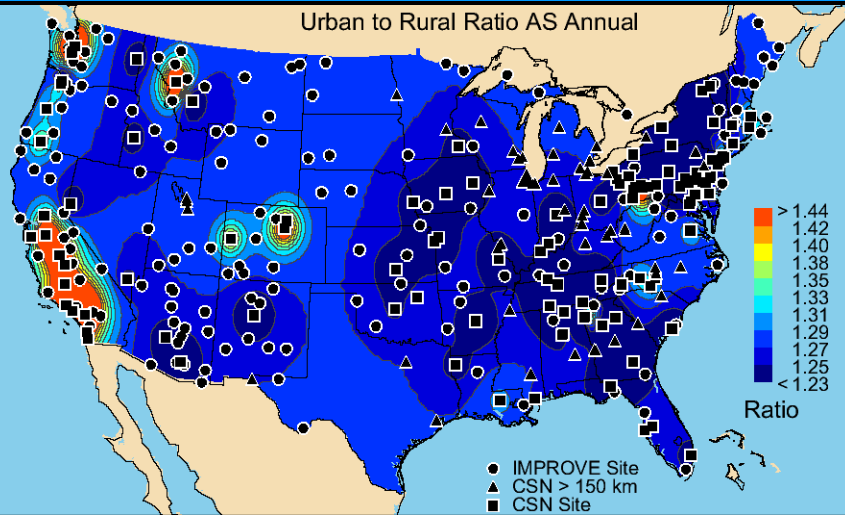
Light Absorbing Carbon



Urban Excess: Ratio of Urban to Rural Concentrations

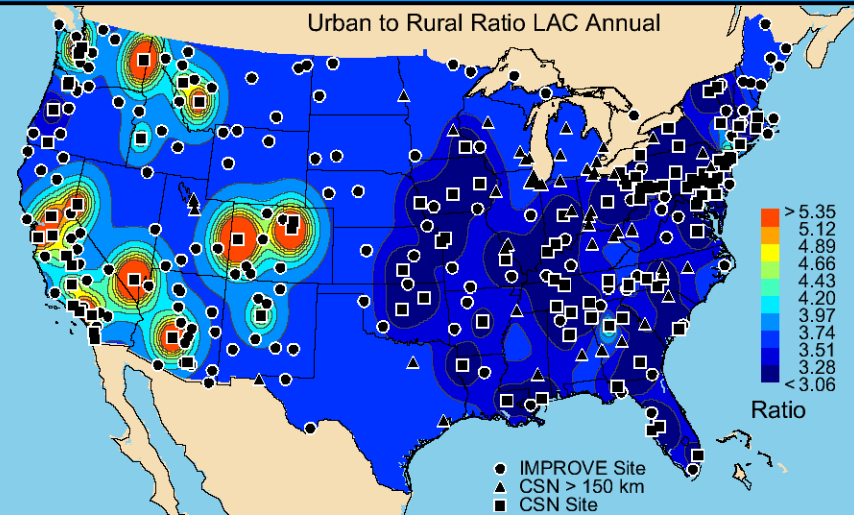
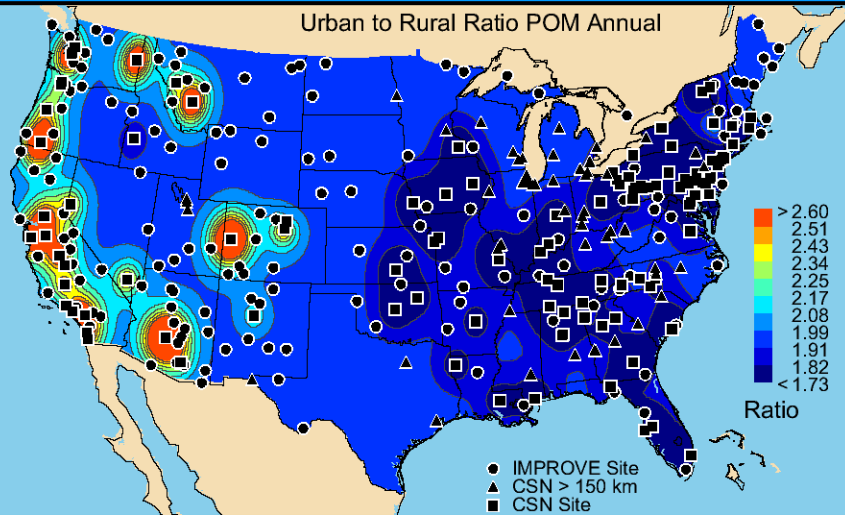
Ammonium Sulfate

Ammonium Nitrate



Particulate Organic Matter

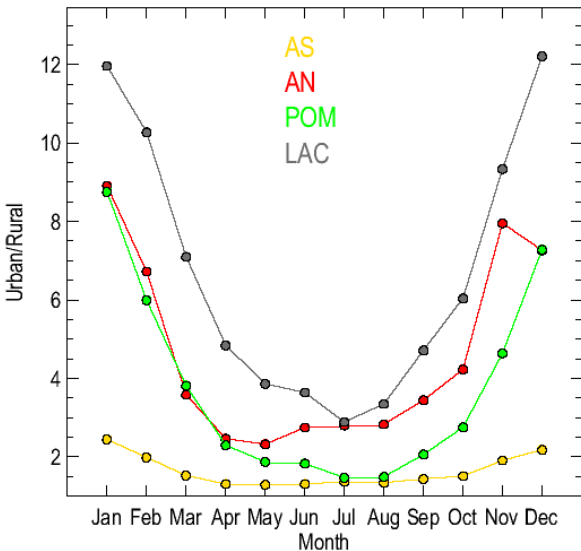
Light Absorbing Carbon



Monthly Mean Urban Excess

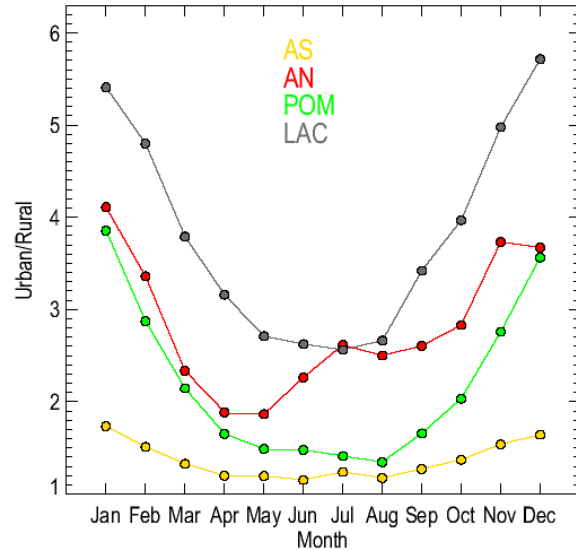
WEST

West Urban/Rural Ratio



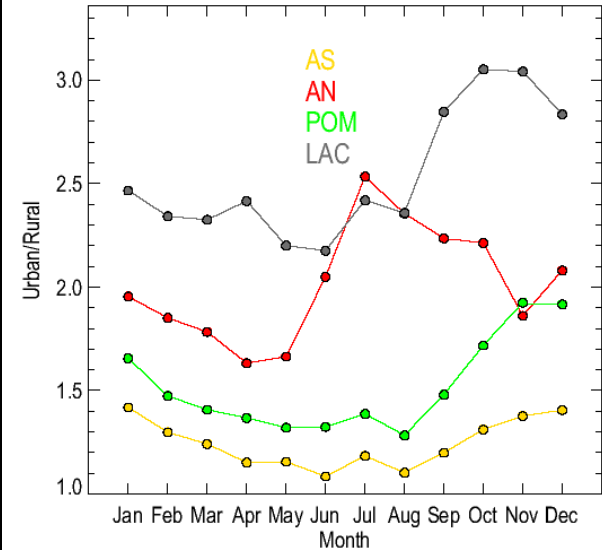
U.S.

US Urban/Rural Ratio



EAST

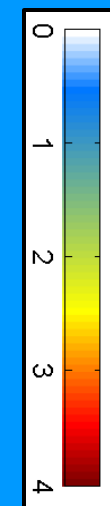
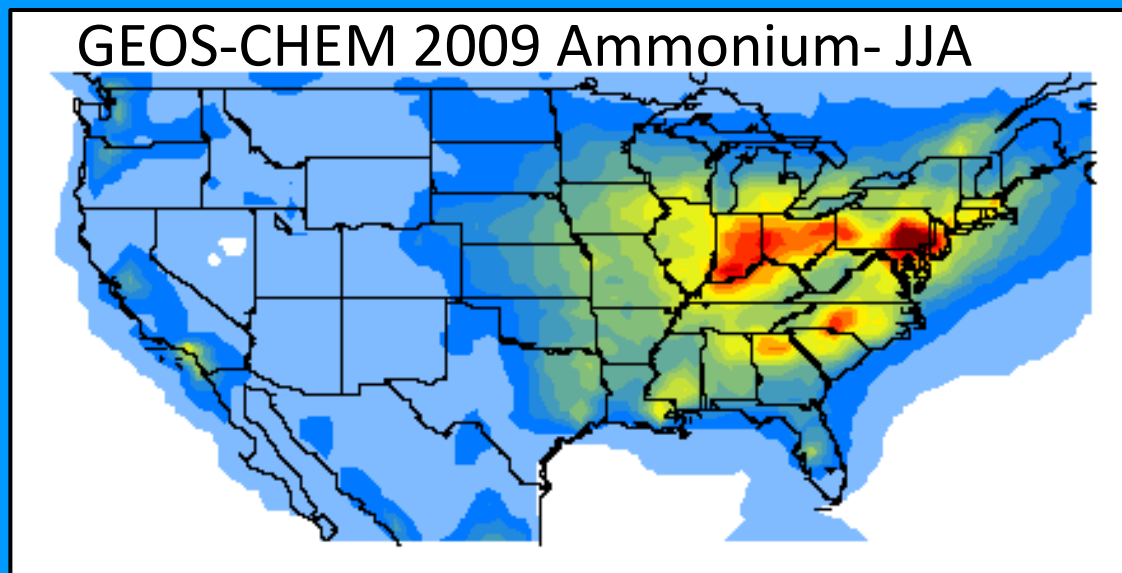
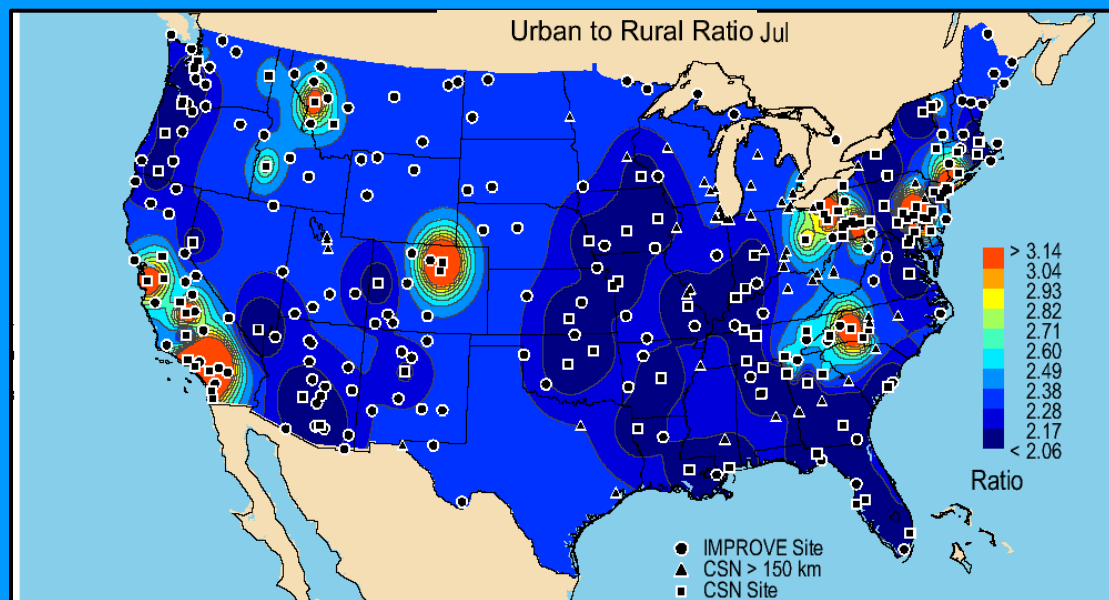
East Urban/Rural Ratio



Annual Mean Ratio

Species	US	East	West
AS	1.3±0.3	1.20±0.14	1.5±0.4
AN	2.7±1.8	1.9±0.7	4.4±2.2
POM	1.9±0.8	1.5±0.3	2.8±0.9
LAC	3.5±2.3	2.5±1.1	5.8±2.8

Urban Excess- Ammonium Nitrate- July



$\mu\text{g m}^{-3}$

Future Work

- Seasonal sulfate trends
- Nitrate and soil trends
- Urban excess

1989

Glacier, MT

($t = -0.7\% \text{ yr}^{-1}$, $p = 0.05$)

2010

Acknowledgements

National Park Service

IMPROVE

WinHaze

(www.air-resource.com/resources/downloads.html)

jlhand@colostate.edu



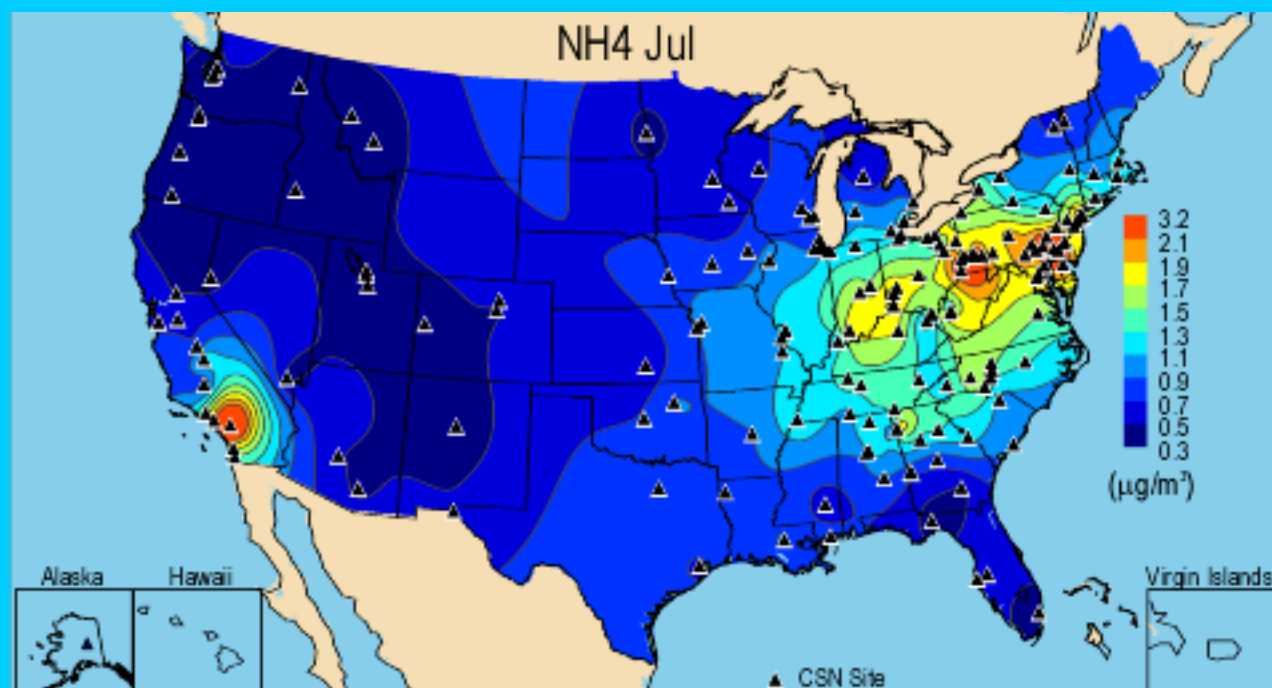
1990

San Geronio, CA

($t = -1.5\% \text{ yr}^{-1}$, $p < 0.01$)

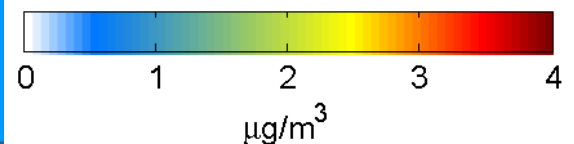
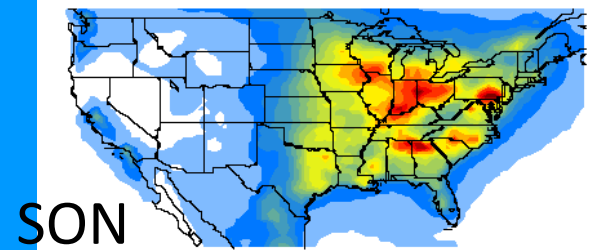
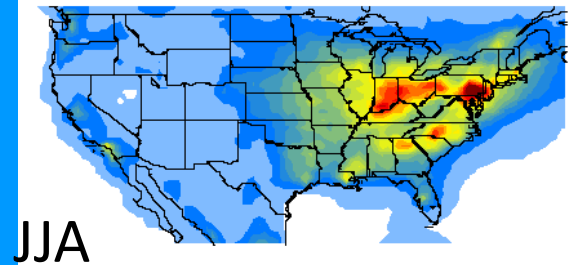
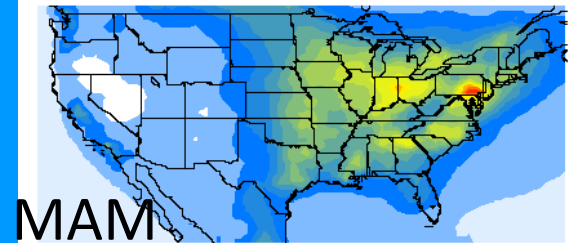
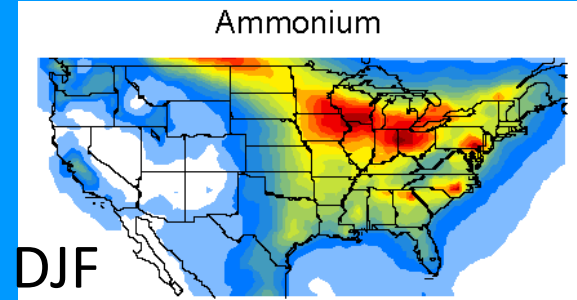
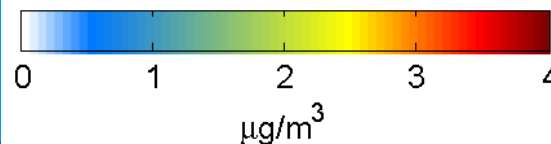
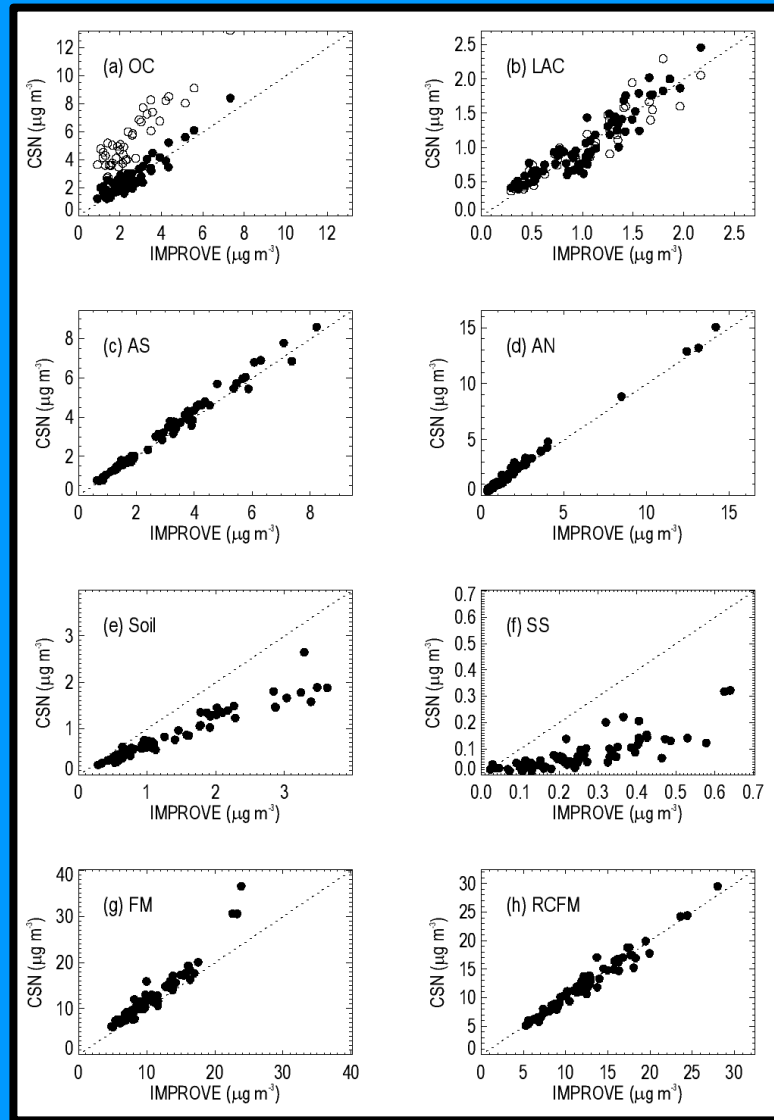
2010



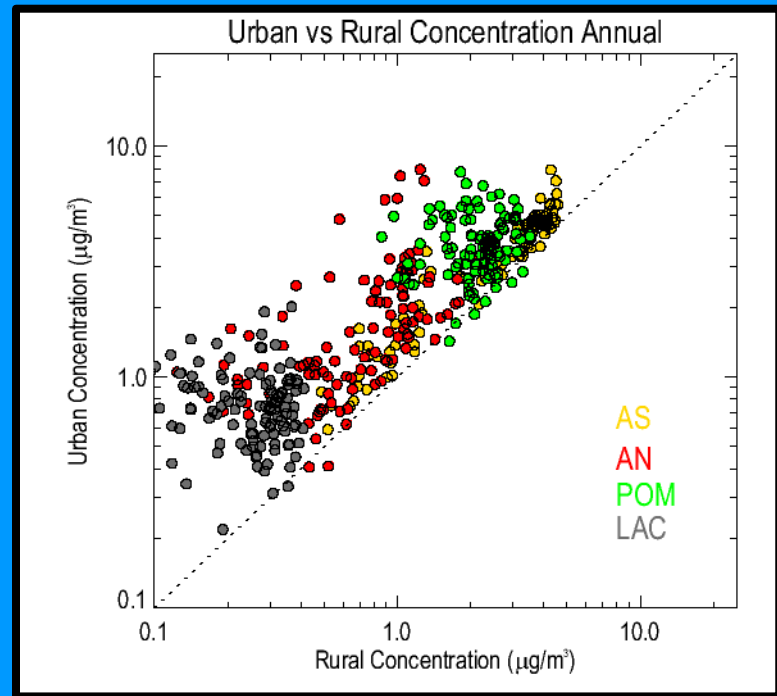
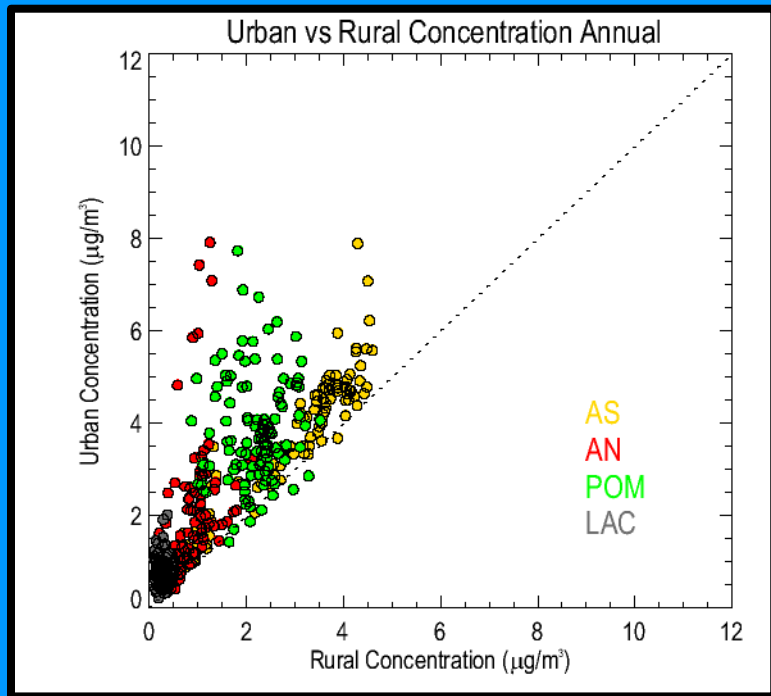


Urban Excess

2009 Modeled Ammonium

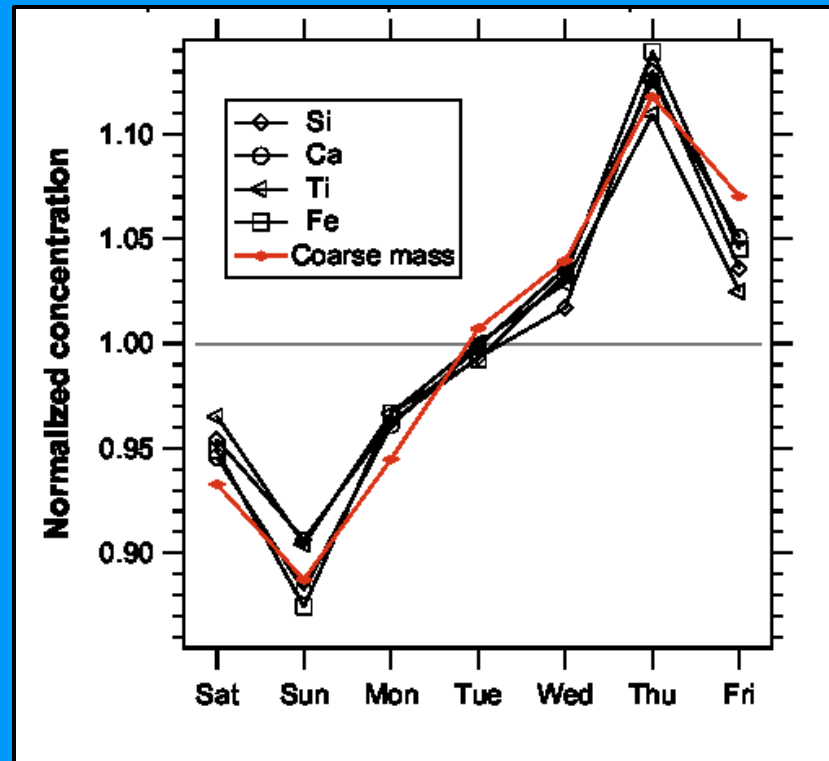


Urban Excess



Weekly cycle suggests anthropogenic soil source

Murphy et al., 2008



Sept 1, 2000- Aug 31, 2006

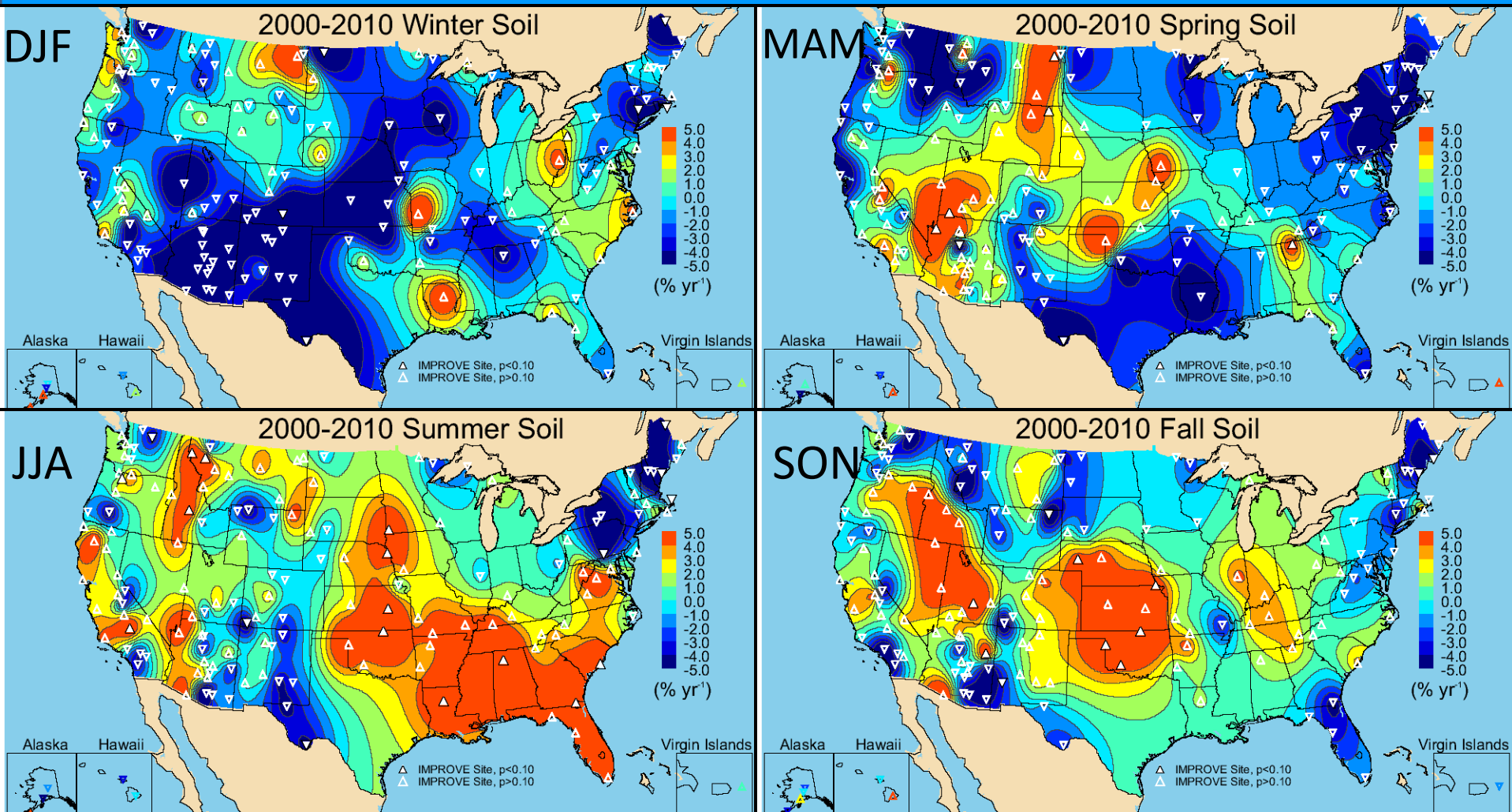
TOTAL CARBON

High Park Fire, CO, June 2012



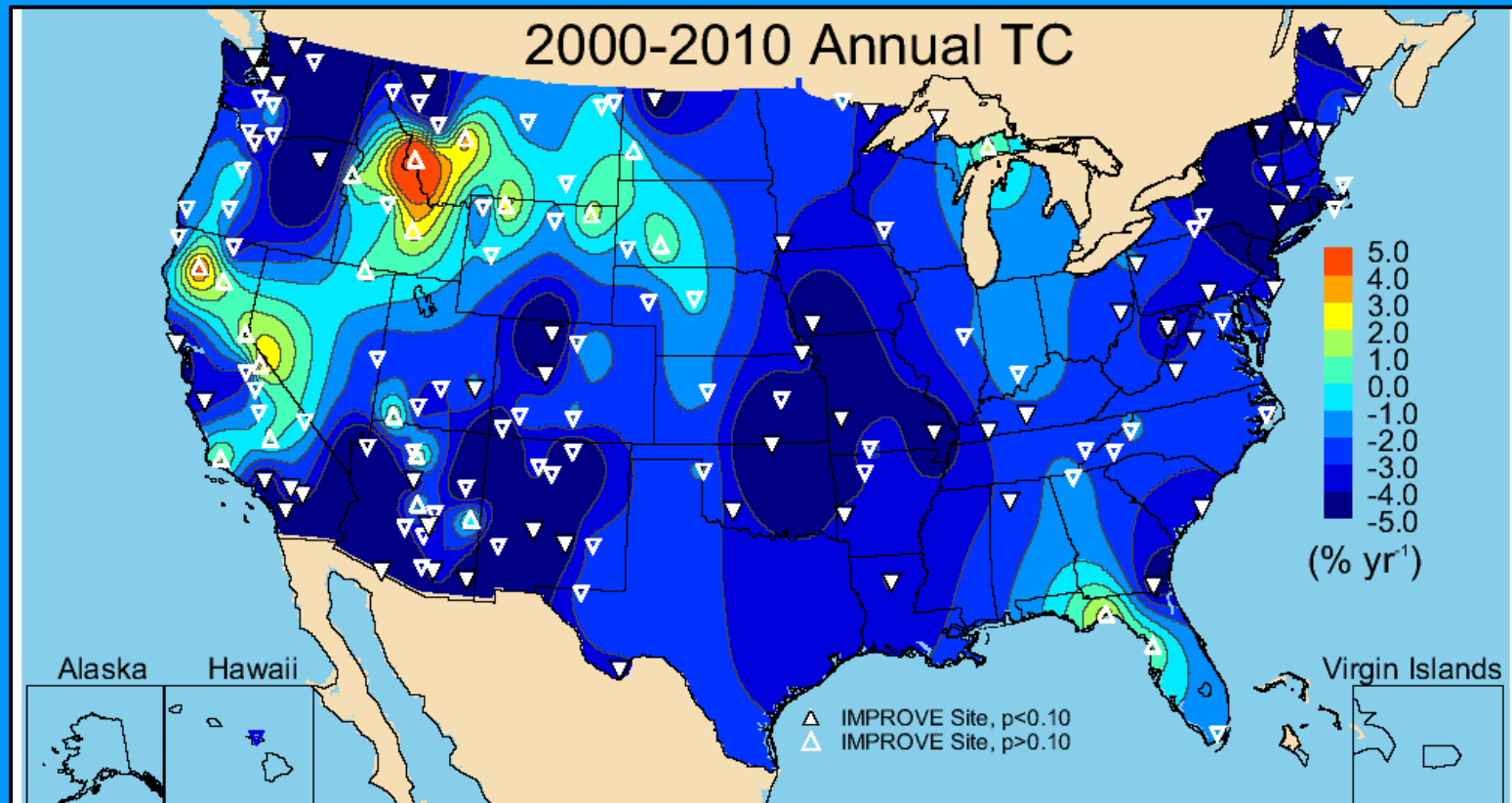
Photos courtesy of Kristi Gebhart

2000-2010 Trends in Seasonal Mean Soil

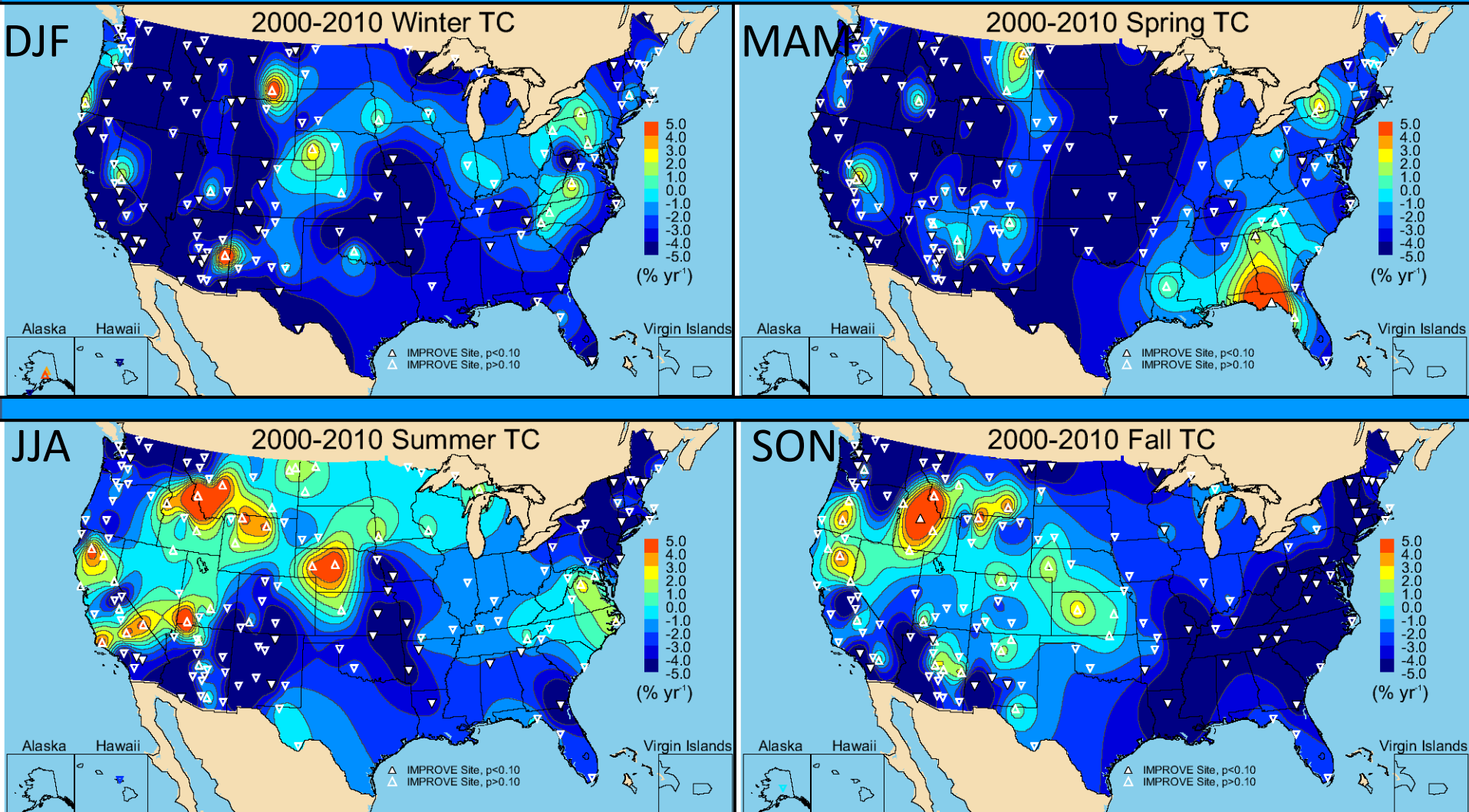


Spring: Impacts from local, Asian and Mexican transport
Summer: Impacts from North Africa

2000-2010 Trends in Annual Mean Total Carbon (TC=OC+EC)



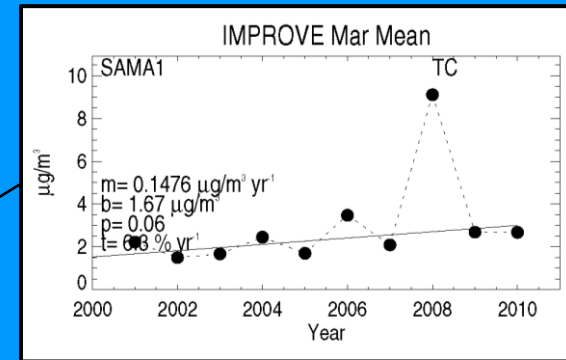
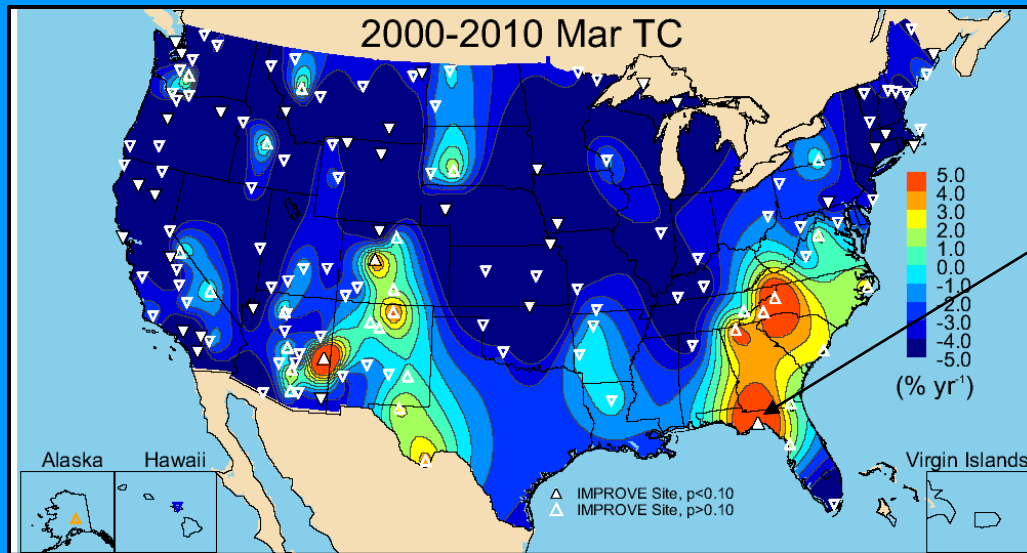
2000-2010 Trends in Seasonal Mean Total Carbon



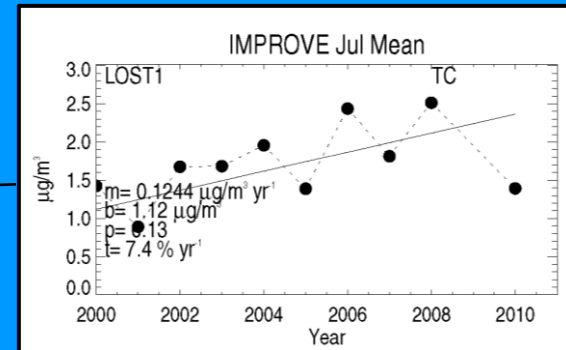
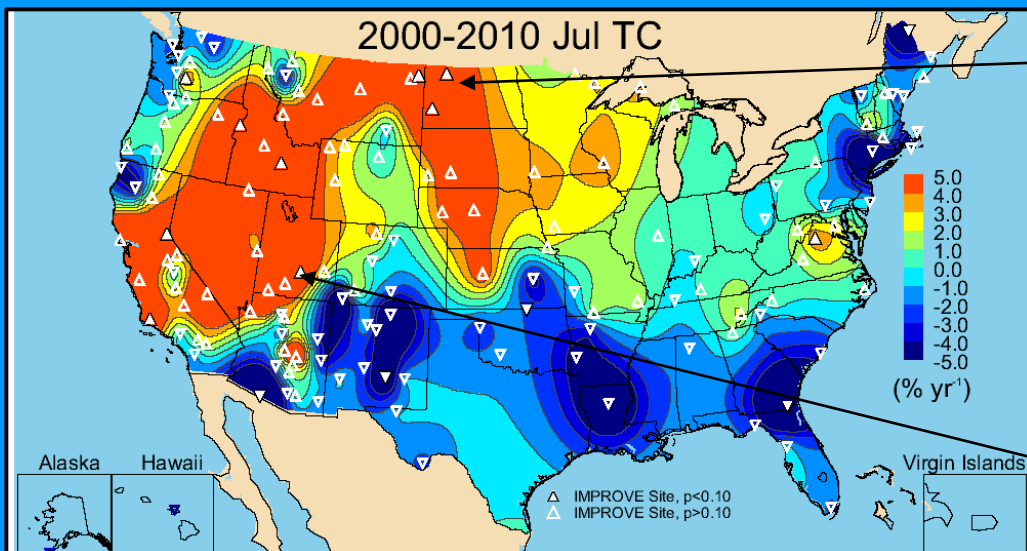
Summer and fall seasonal means are increasing at sites in the West

Impacts from biomass smoke?

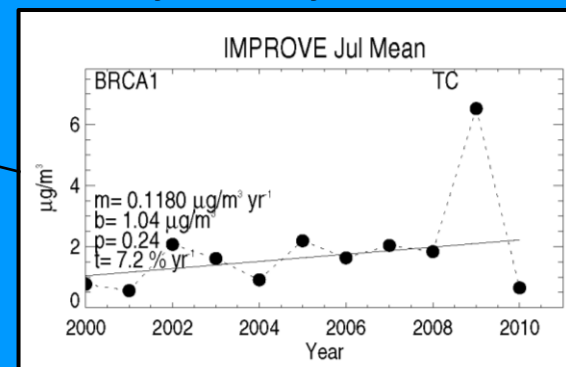
St. Marks, FL



Lostwood, ND

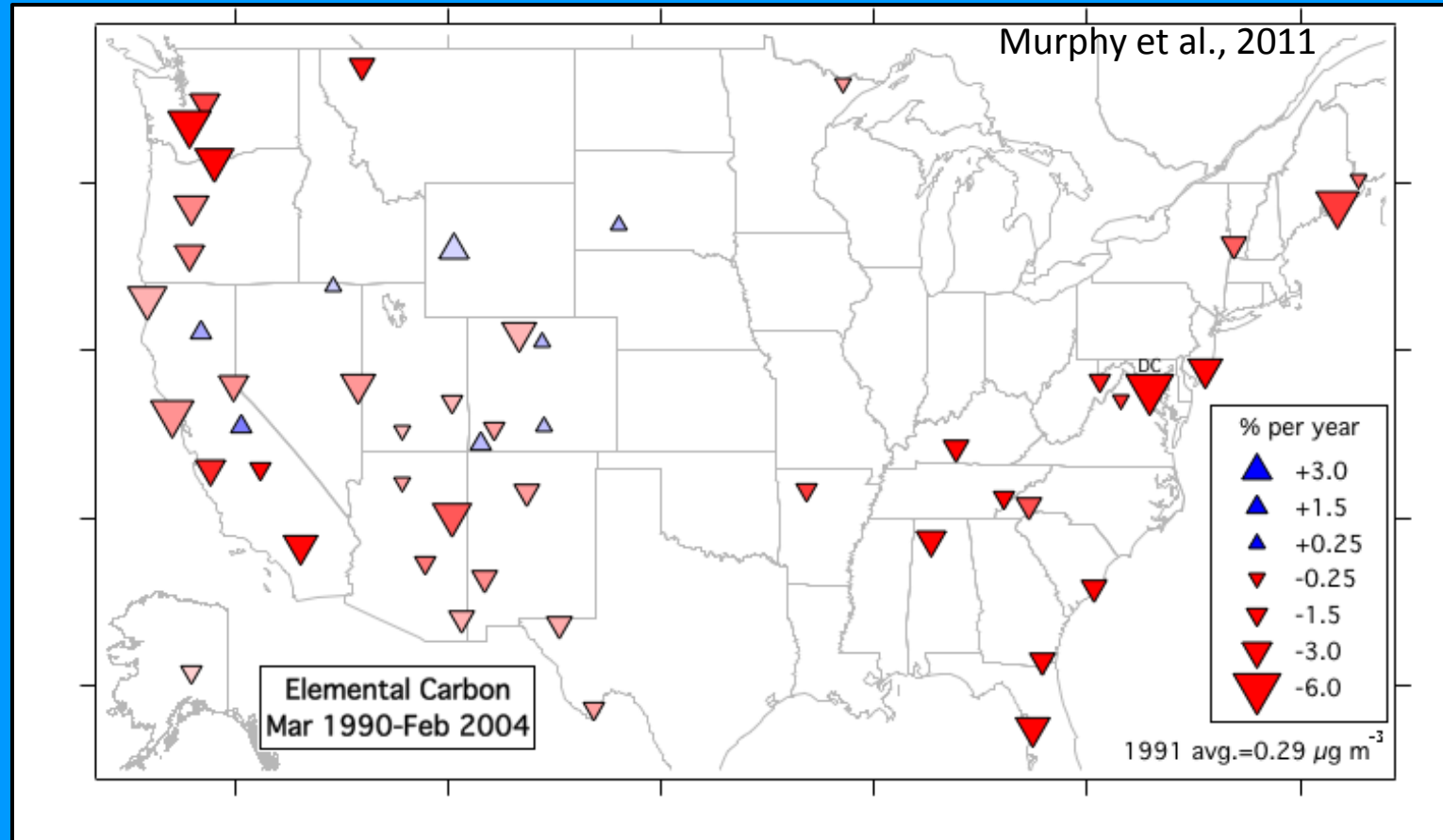


Bryce Canyon, UT



Widespread Decreases in Annual Elemental Carbon

EC trends from 1990 – 2004

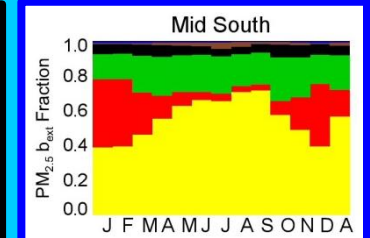
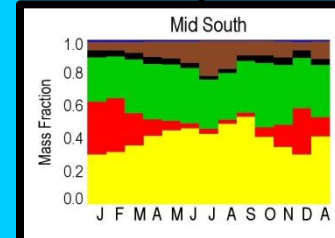
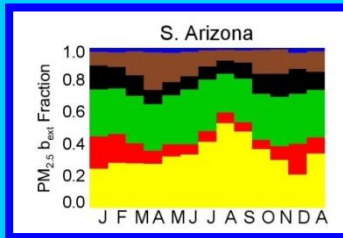
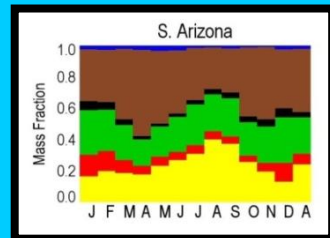
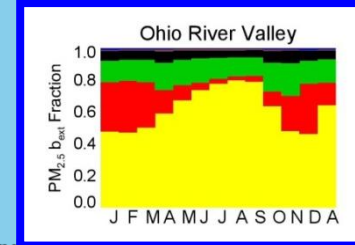
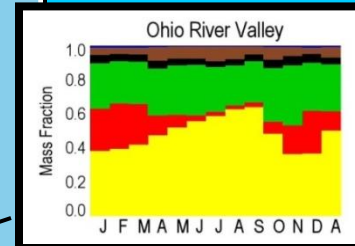
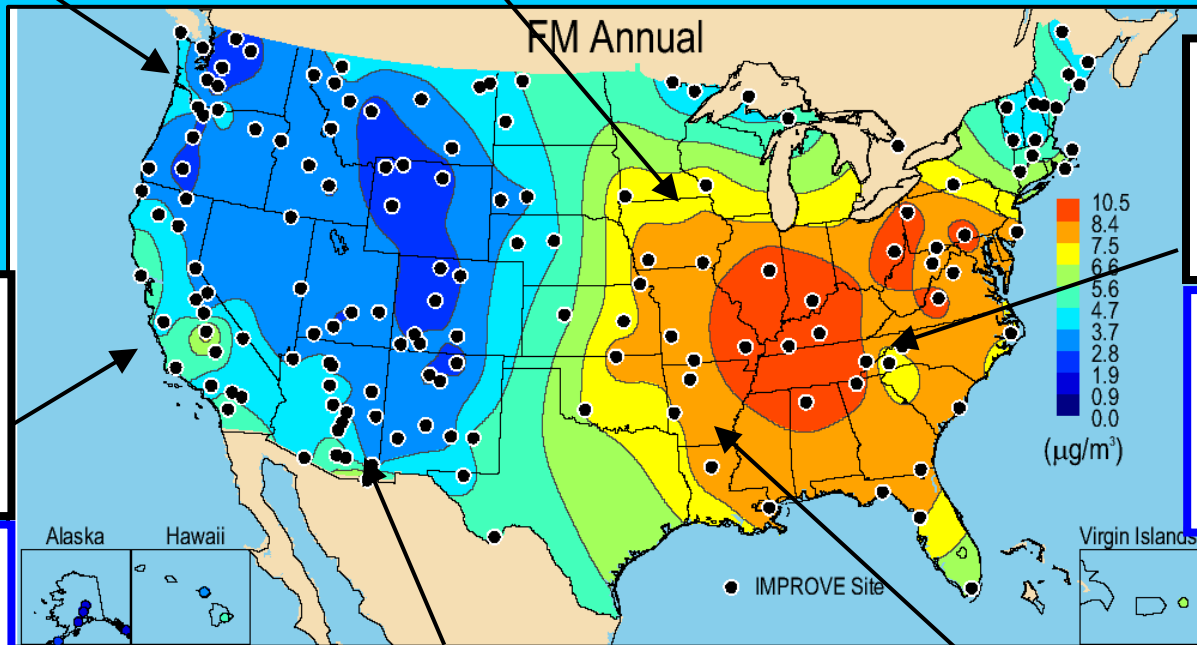
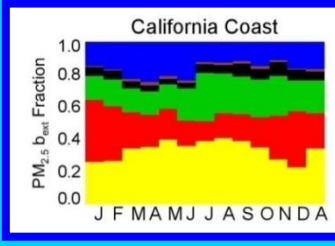
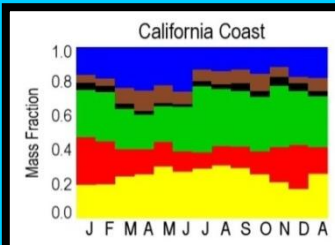
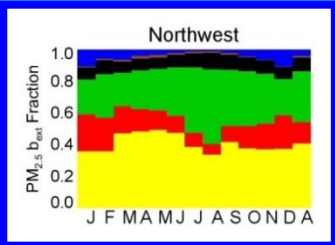
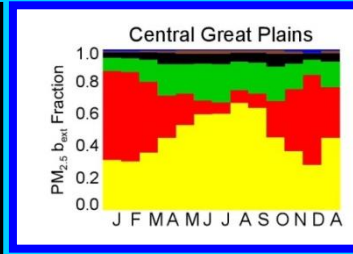
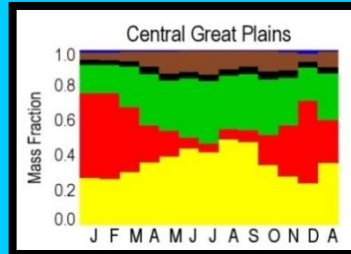
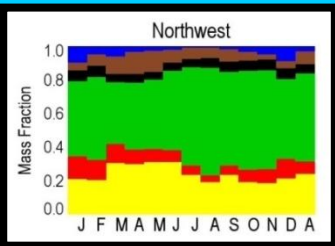


Symbol size: magnitude of trend

Color saturation: absolute amount of EC

2007–2010 Annual Mean PM_{2.5} Mass

Ammonium sulfate
 Ammonium nitrate
 Particulate organic matter
 Elemental carbon
 Soil
 Sea salt



Mass Fraction PM_{2.5} b_{ext} fraction

2005-2011 Trends in OMI tropospheric NO₂ observations

Russell et al., 2012, ACPD

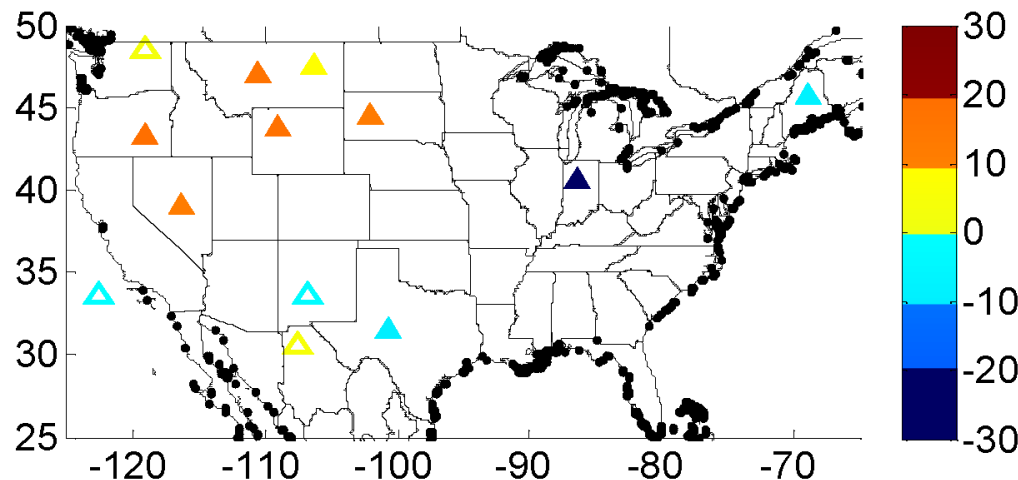
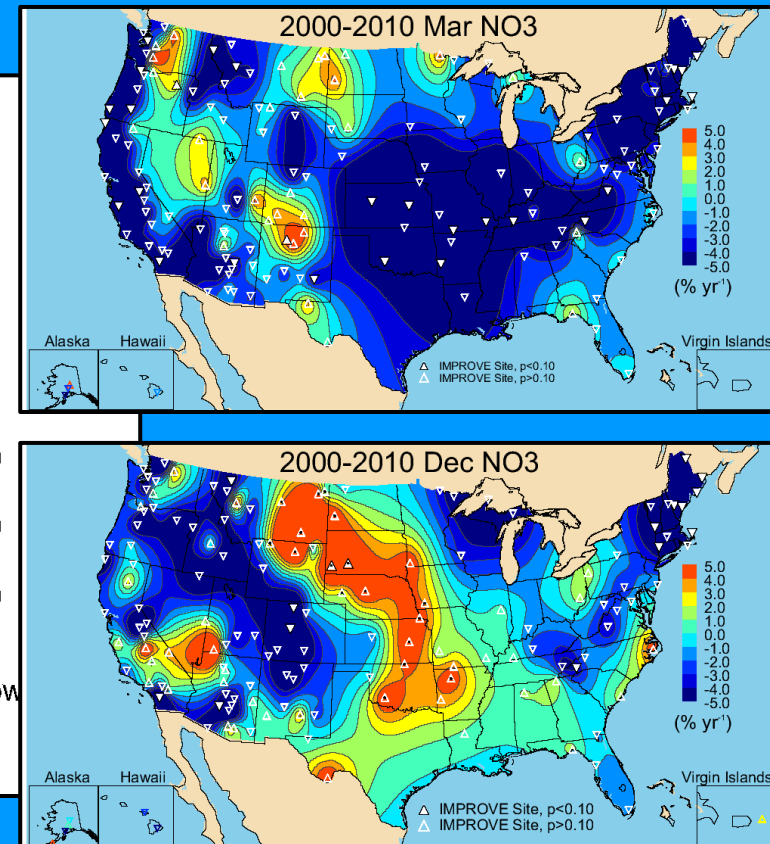
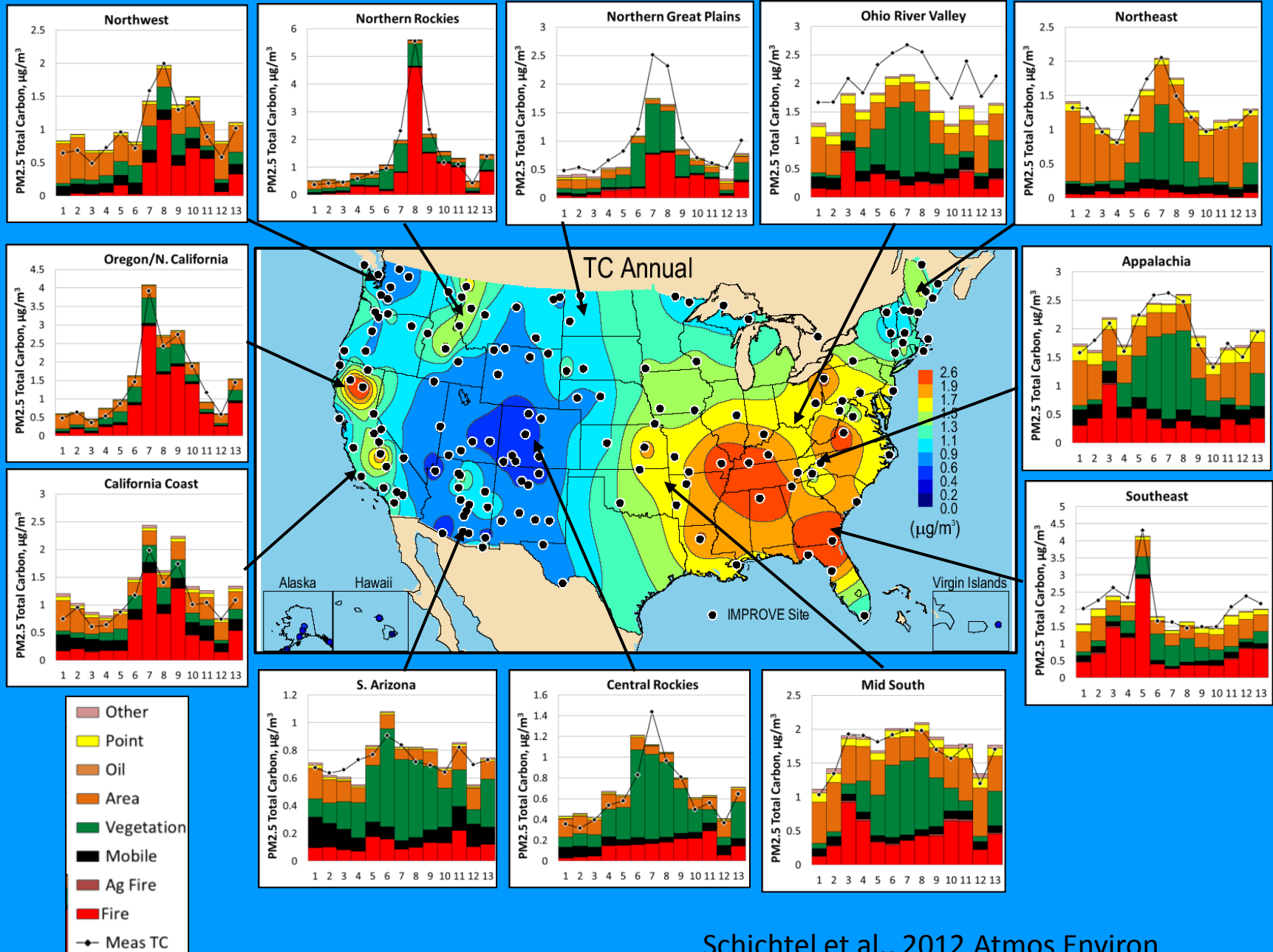


Fig. 7. Percent change in OMI-observed background NO₂ columns. Open triangles show the change is $< \pm 5\%$ while the solid triangles show where the change is $\geq \pm 5\%$.

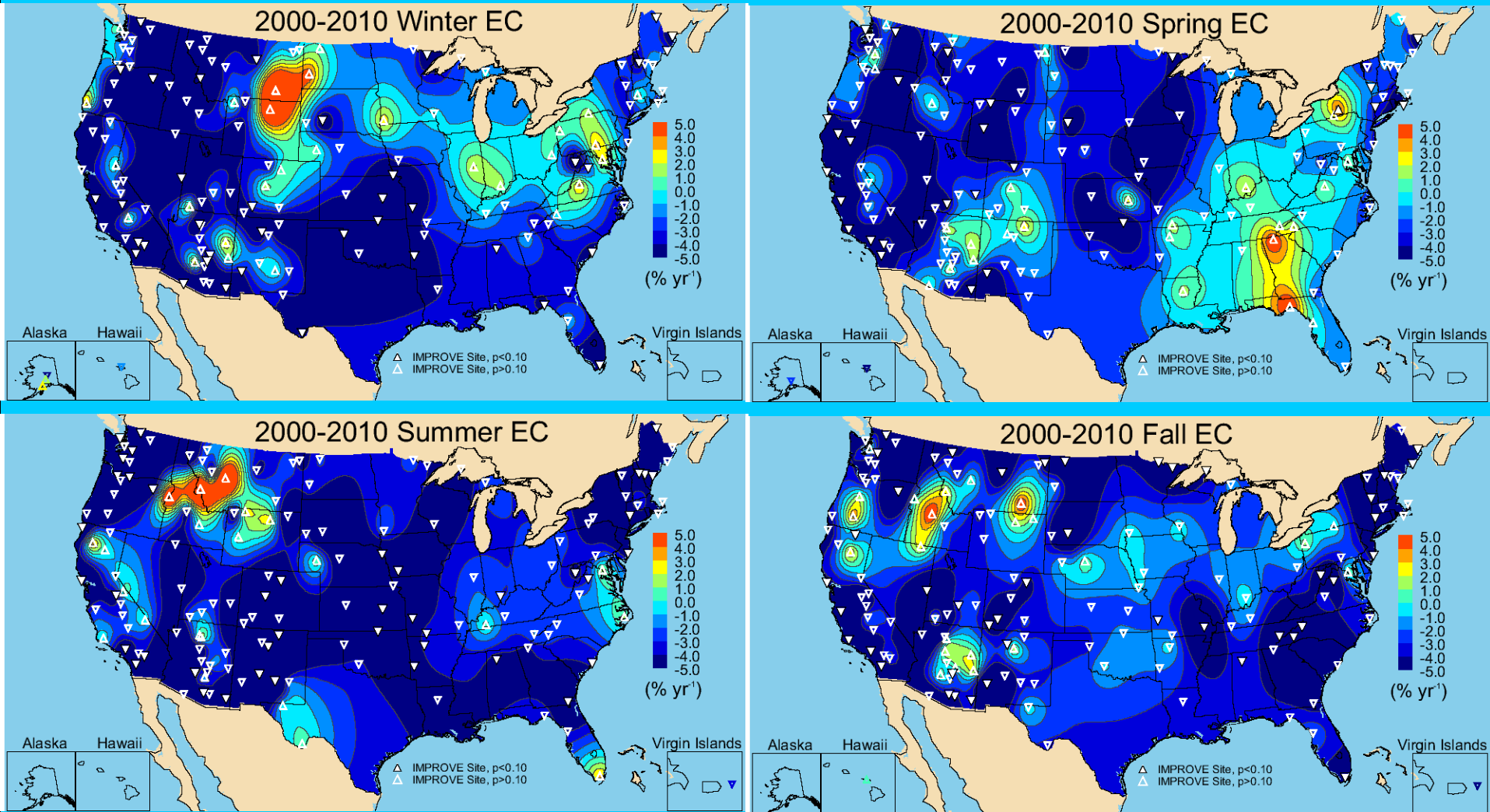


No consistent seasonal cycle was observed
Low concentrations leading to uncertainties?
Large decreases in anthropogenic emissions in urban regions

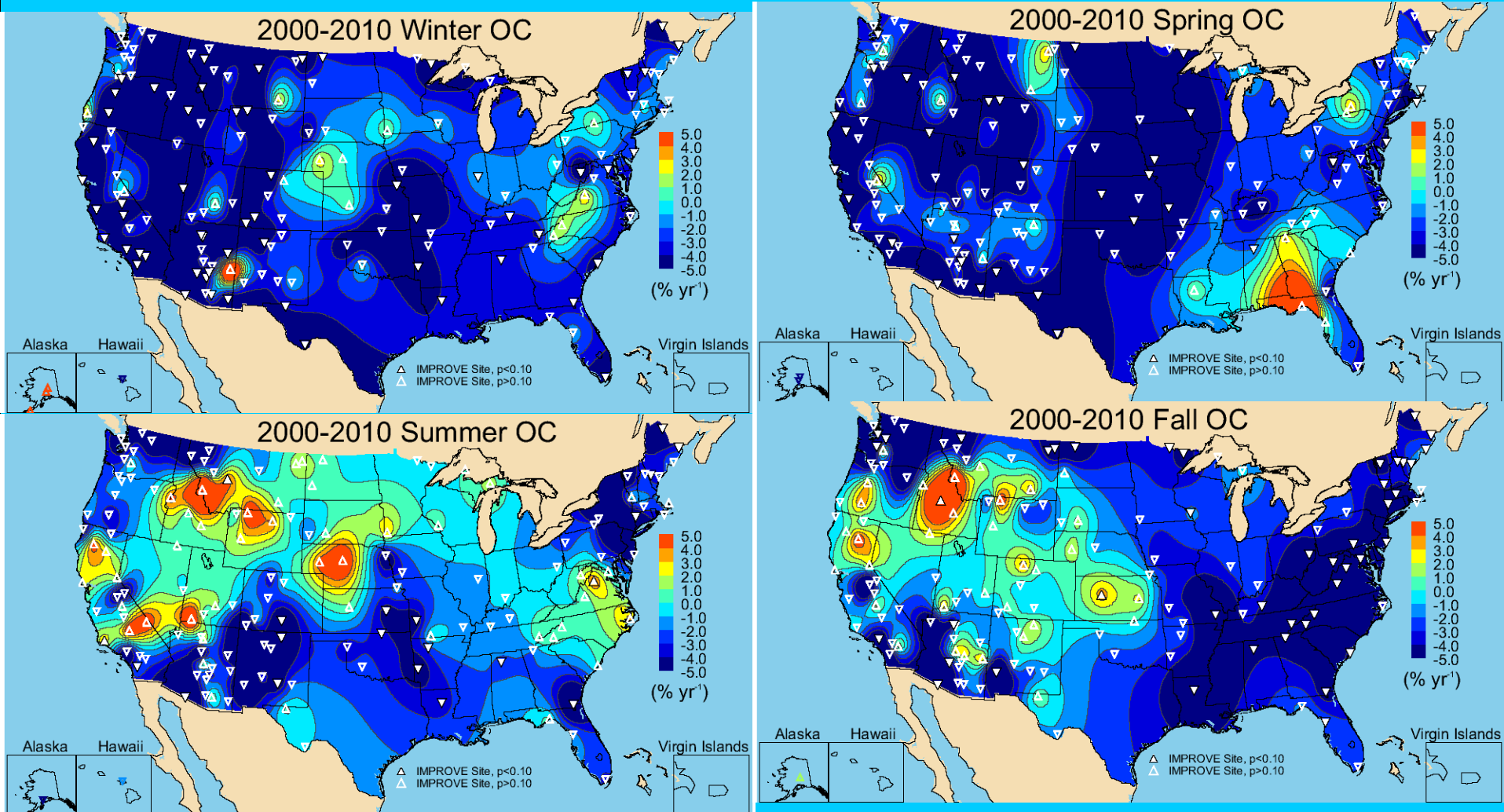
2006–2008 Carbon Source Apportionment



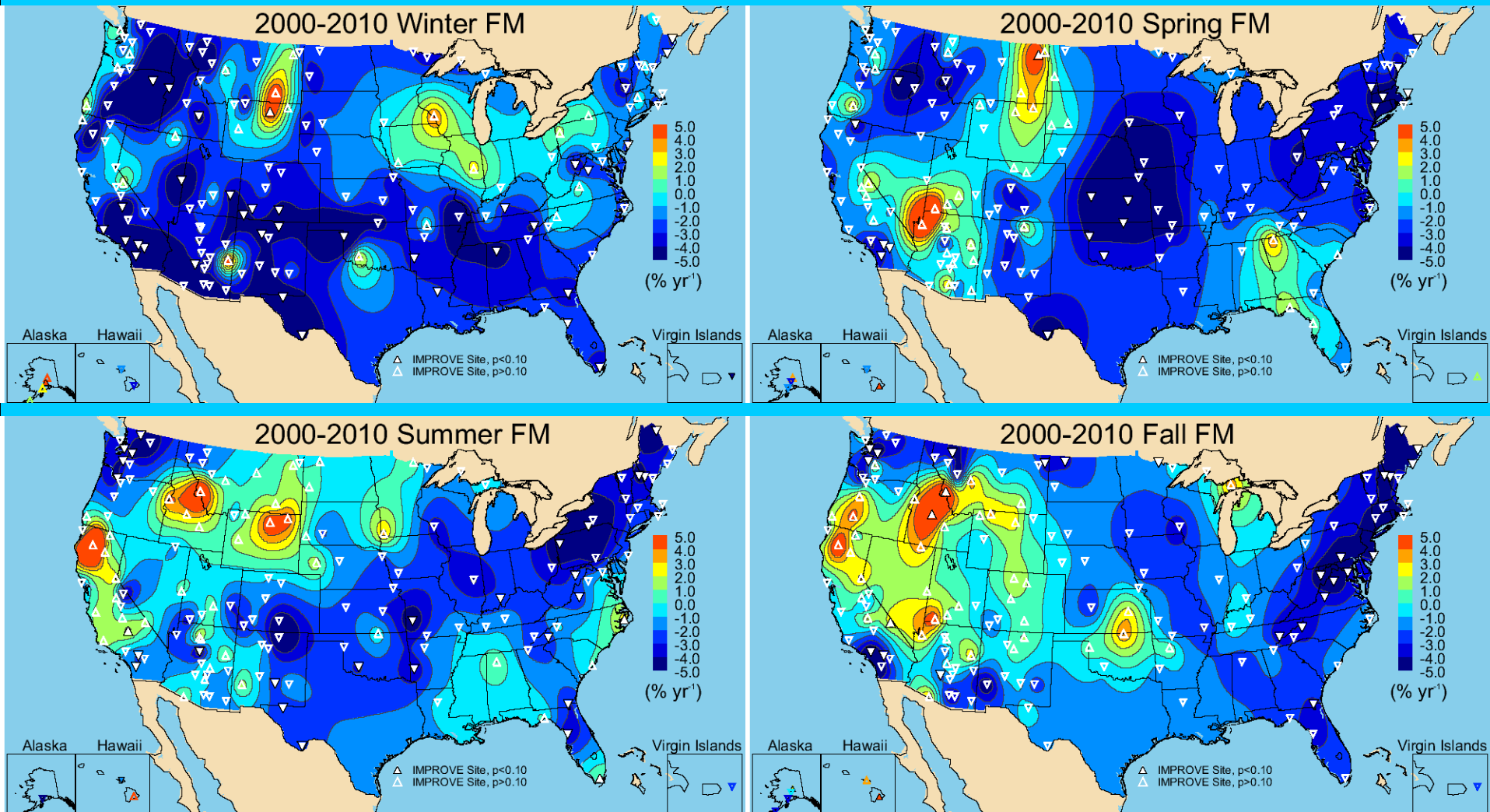
2000-2010 Trends in Seasonal Mean EC



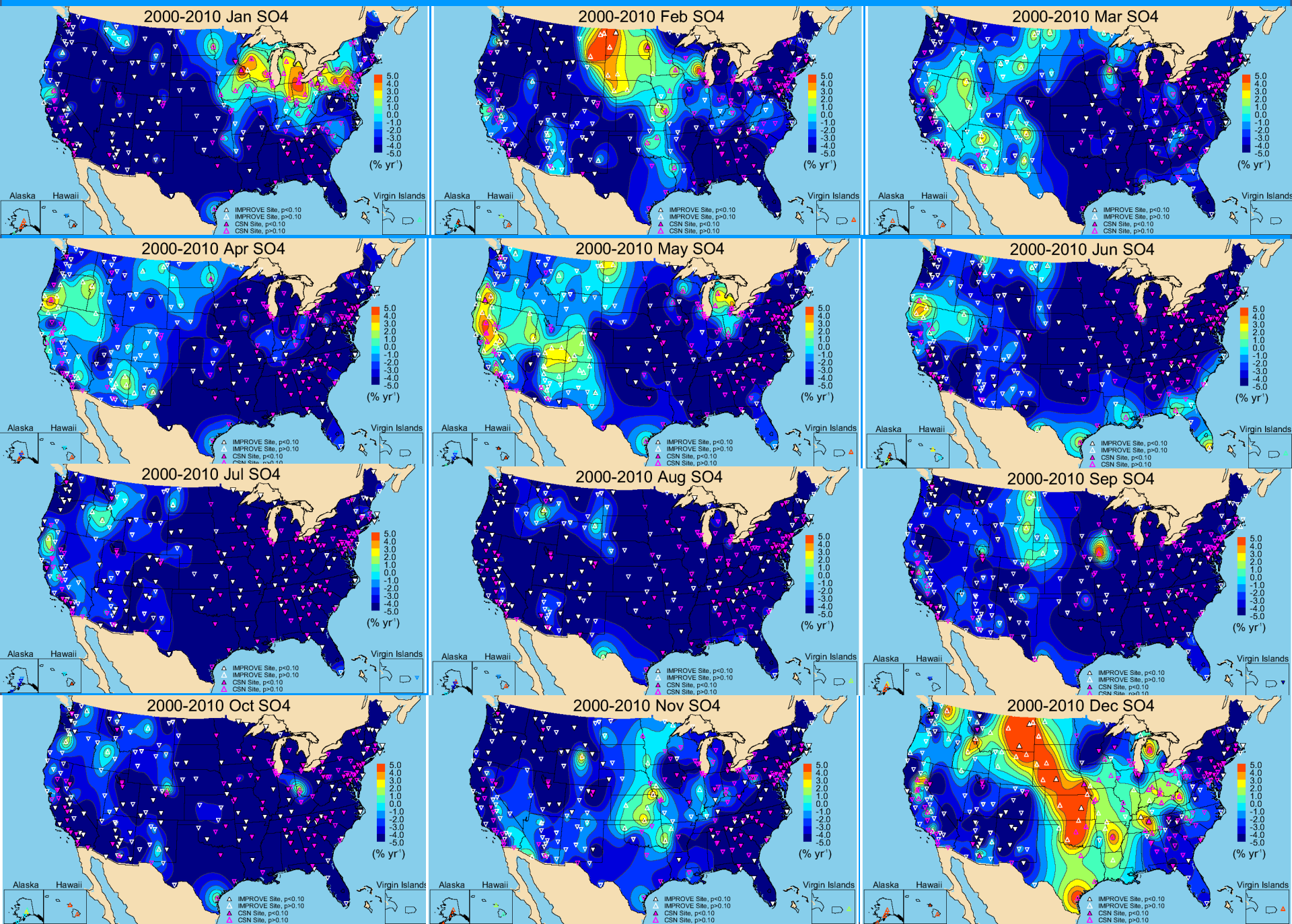
2000-2010 Trends in Seasonal Mean OC



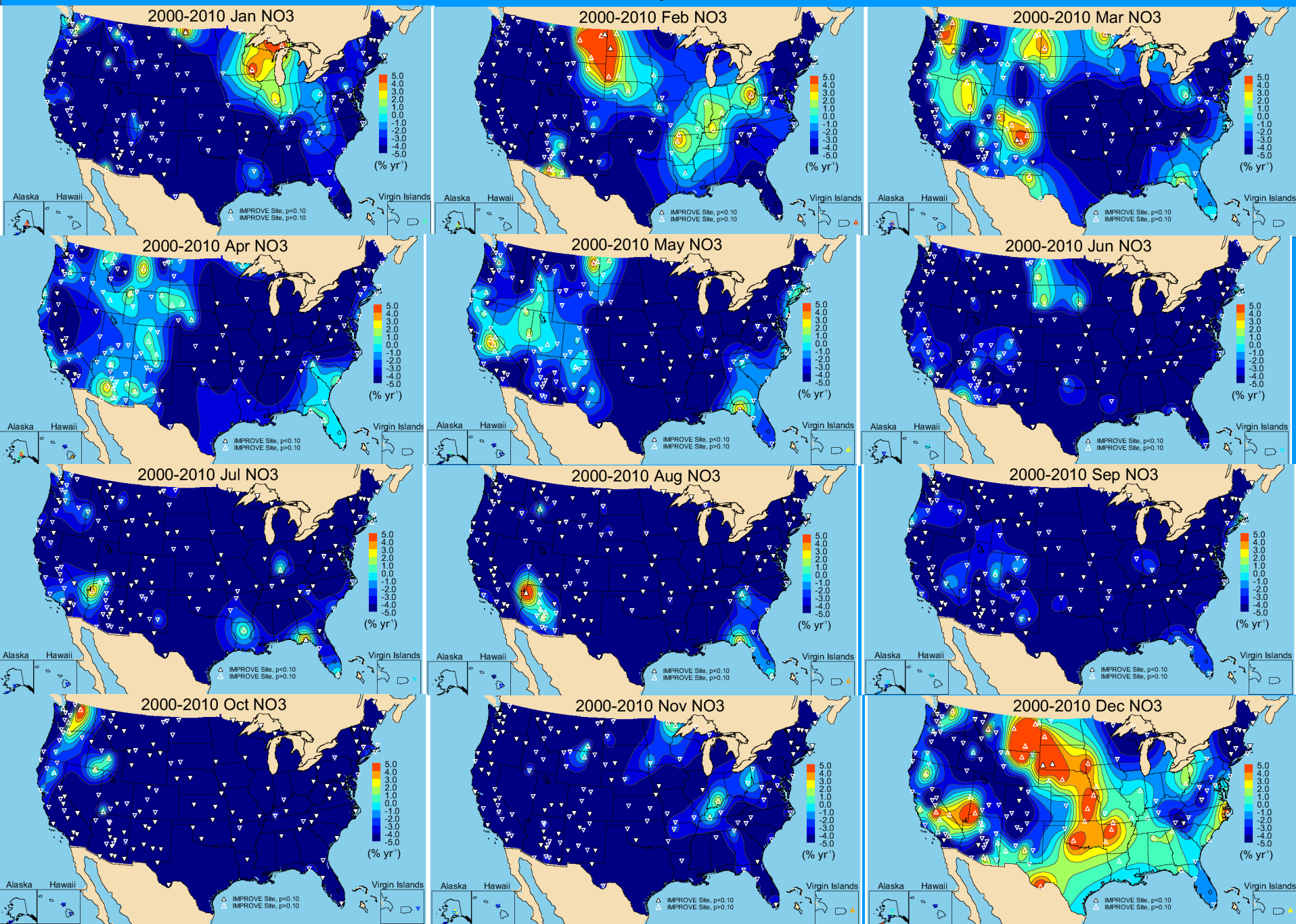
2000-2010 Trends in Seasonal Mean FM



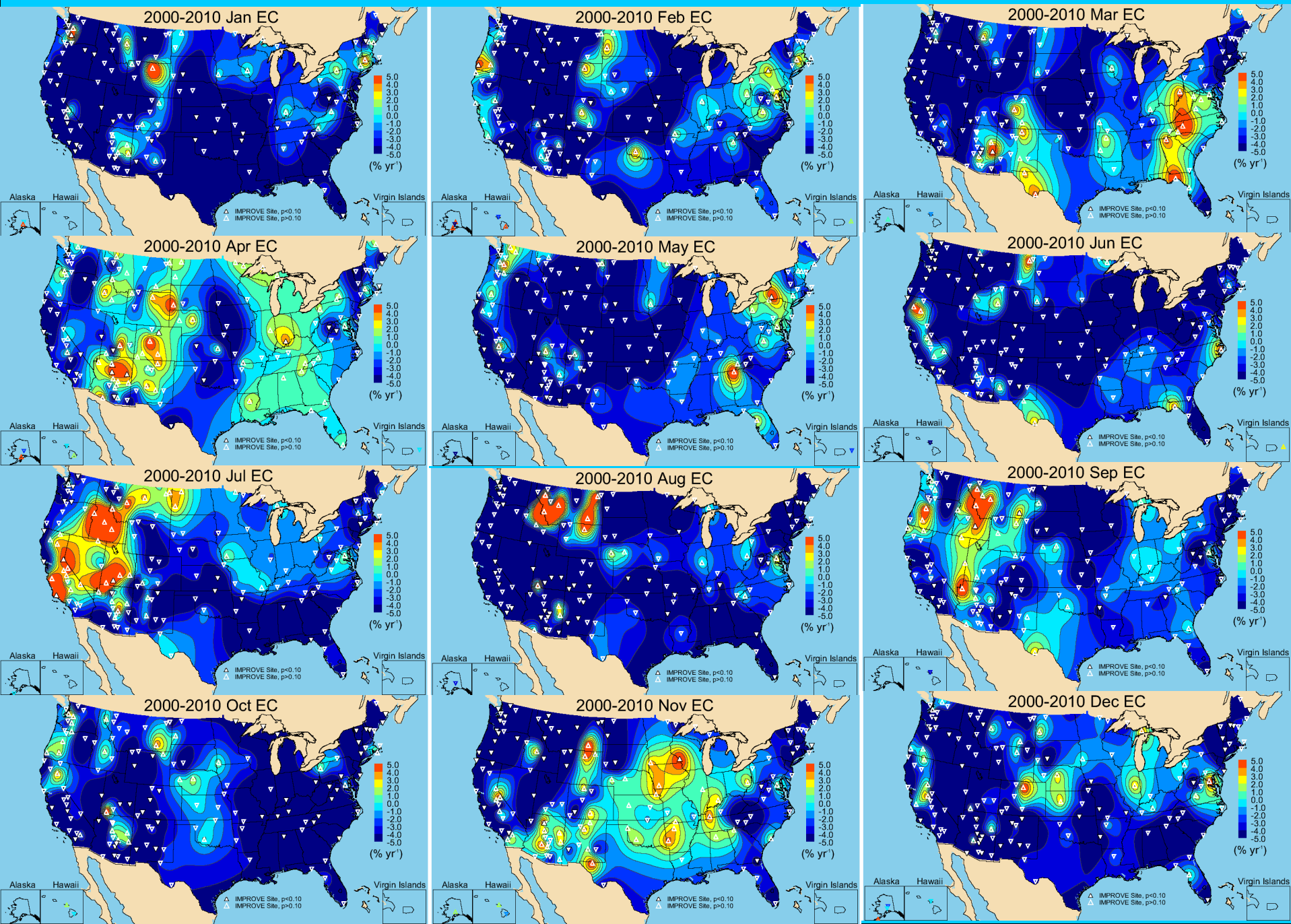
2000-2010 Monthly Mean Sulfate Trends



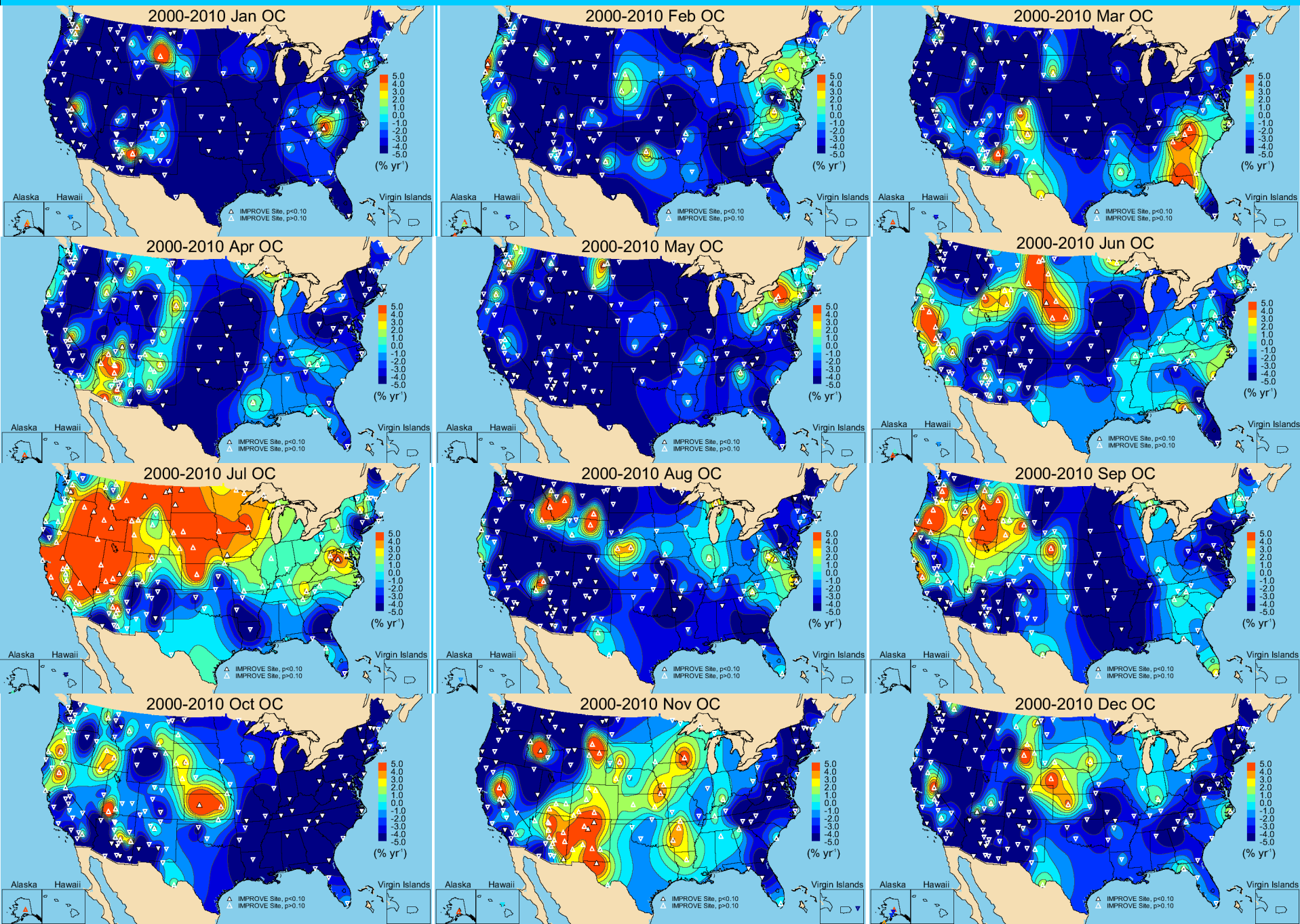
2000-2010 Monthly Mean Nitrate Trends



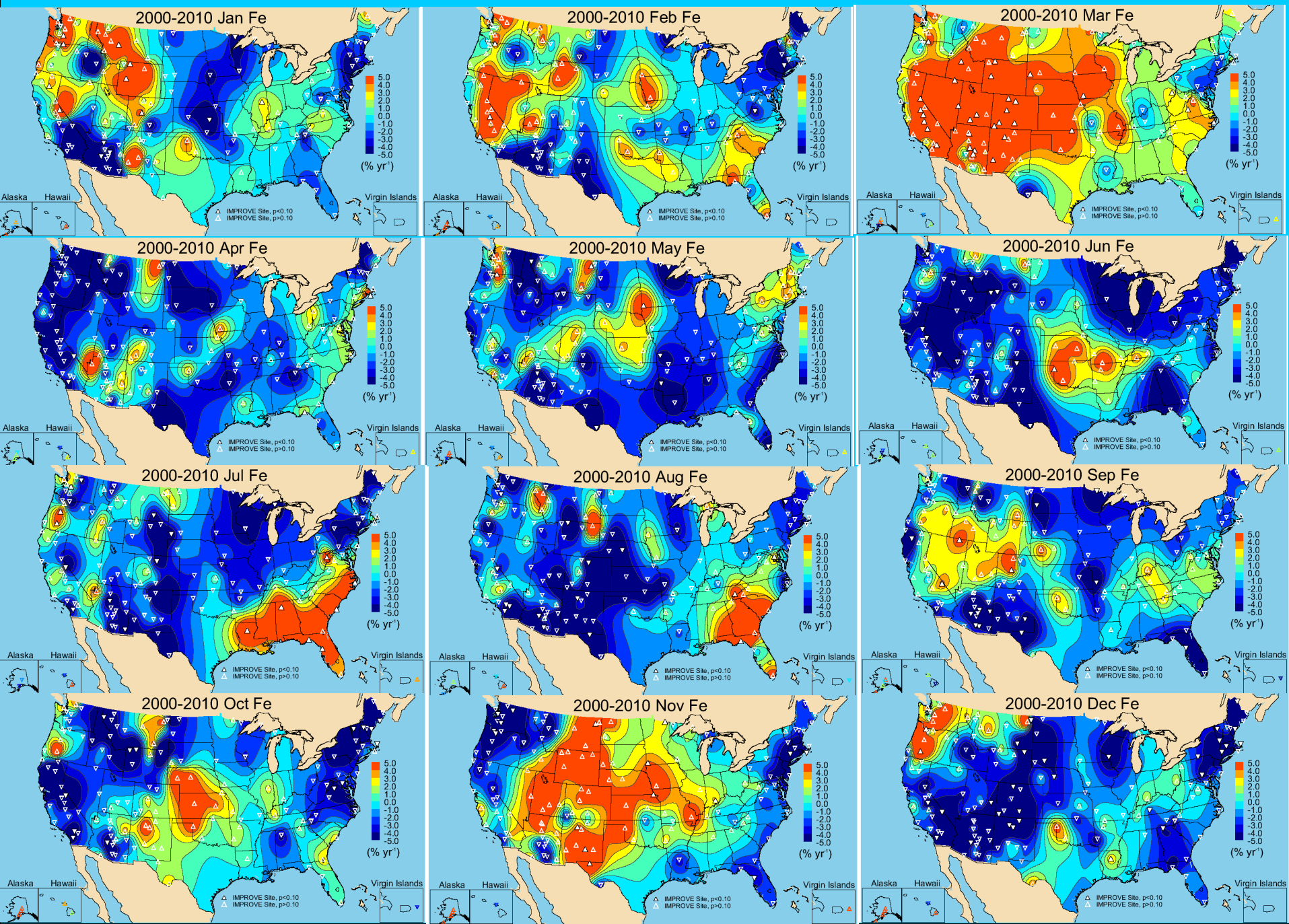
2000-2010 Monthly Mean EC Trends



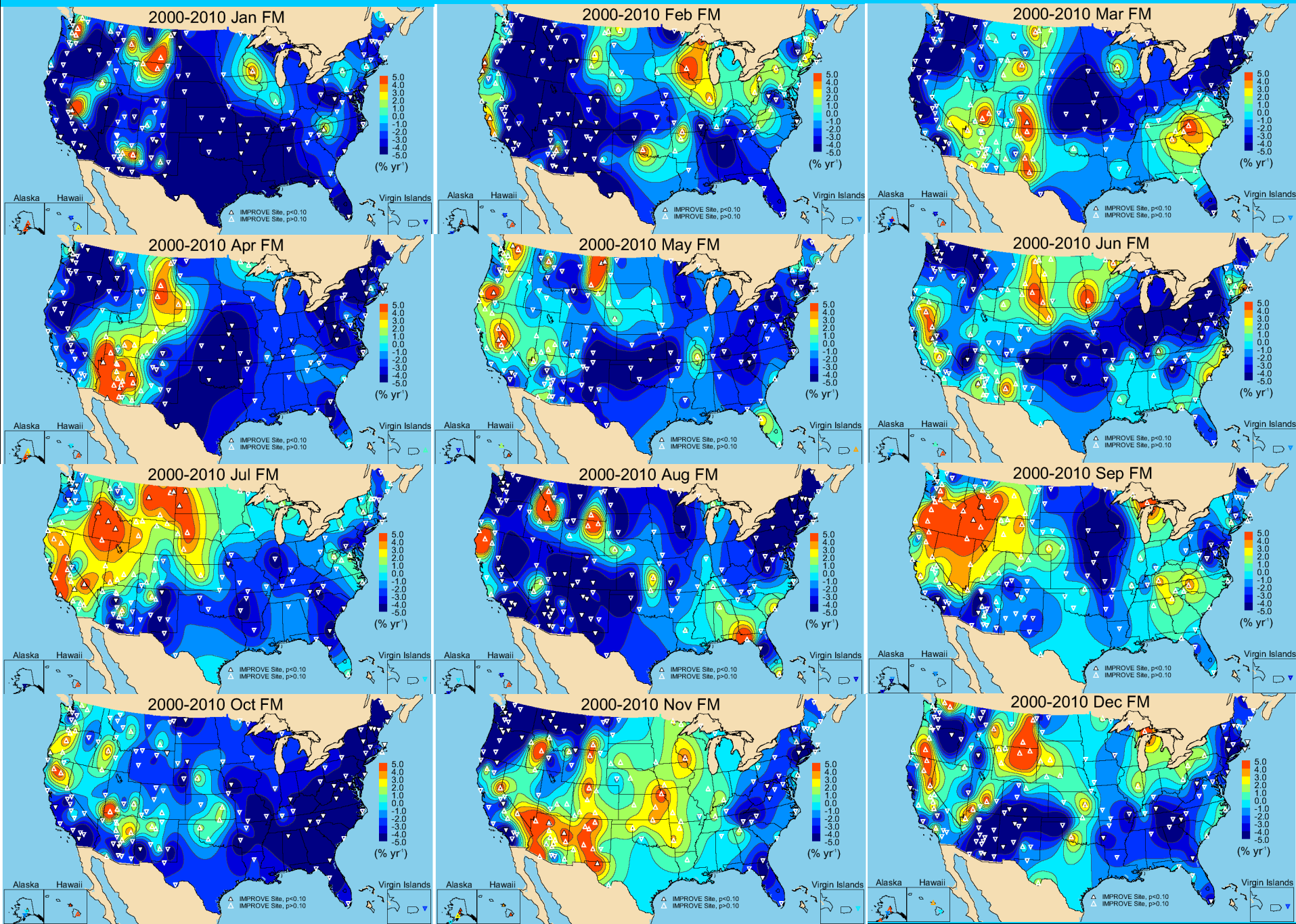
2000-2010 Monthly Mean OC Trends



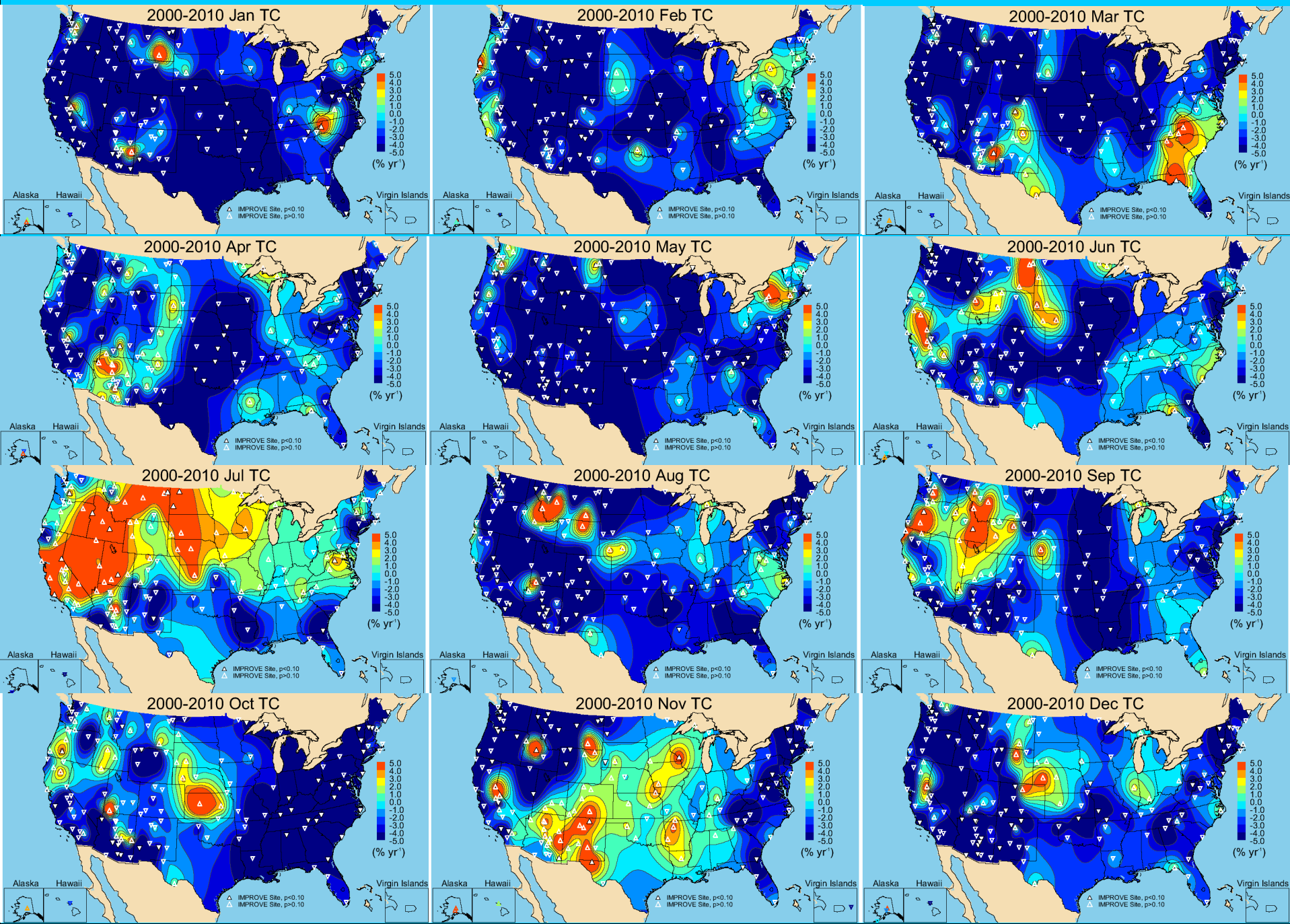
2000-2010 Monthly Mean Fe Trends



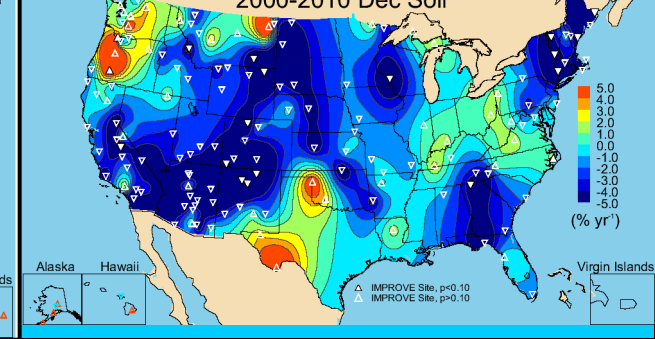
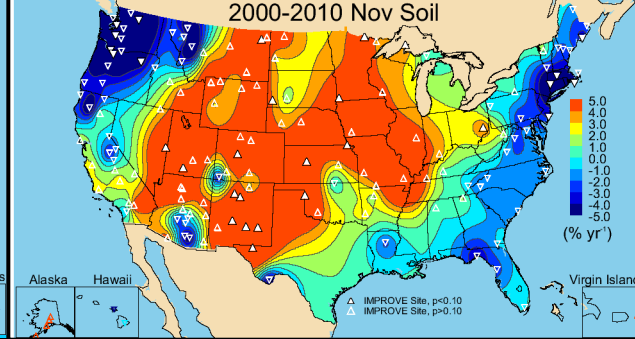
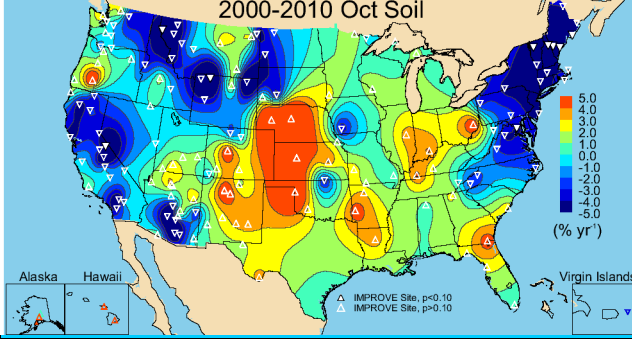
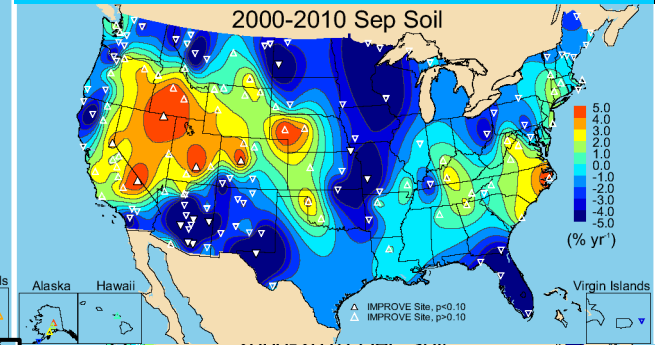
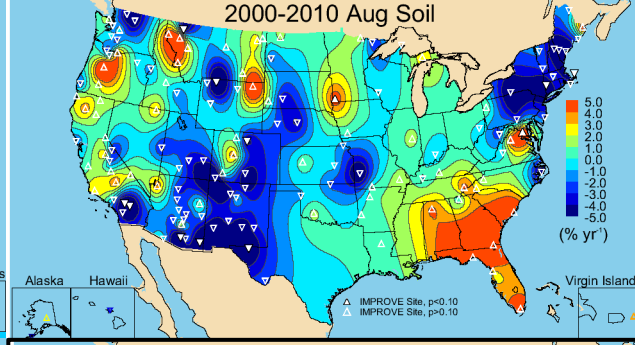
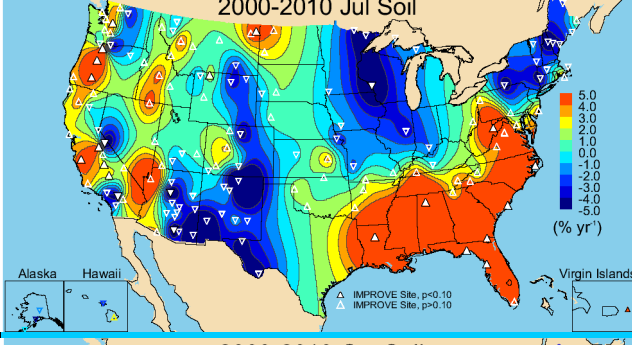
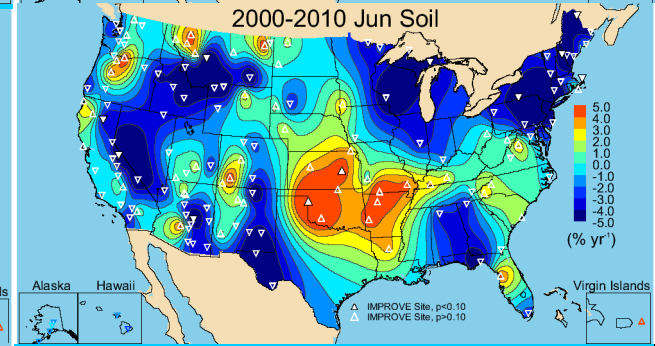
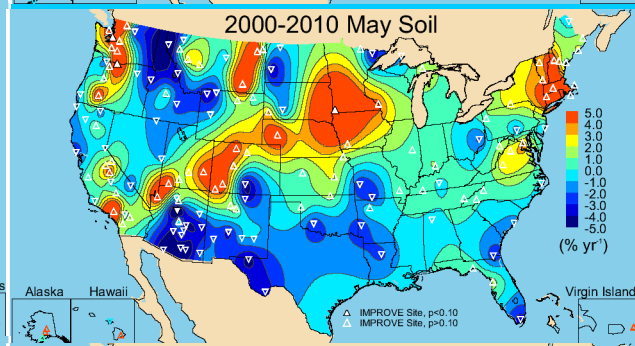
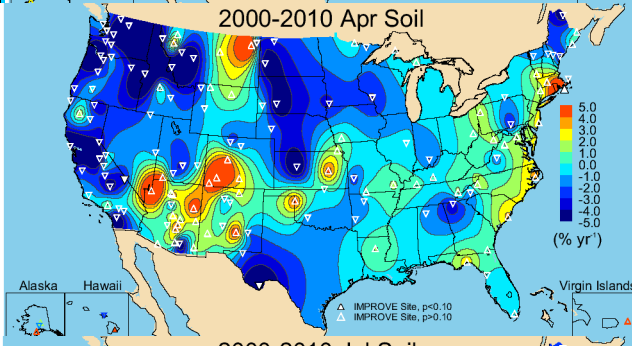
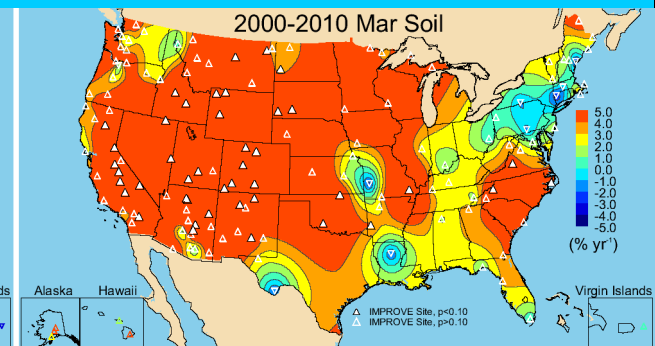
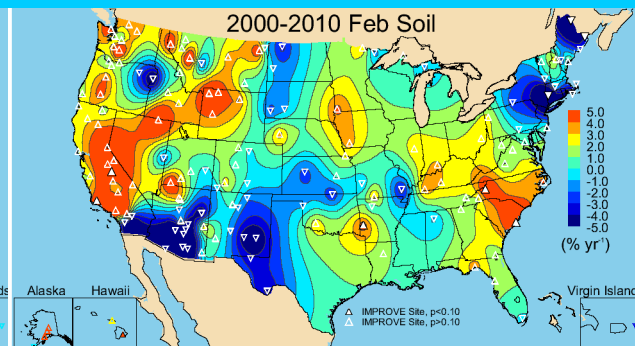
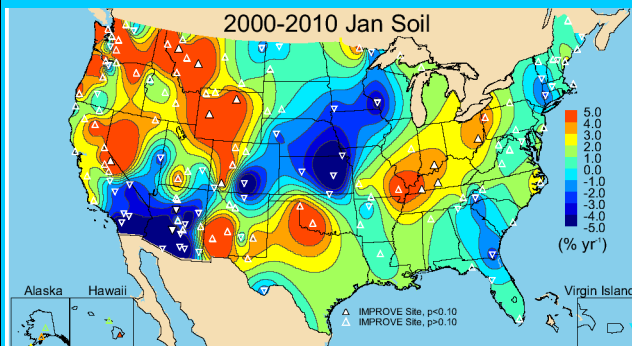
2000-2010 Monthly Mean FM Trends



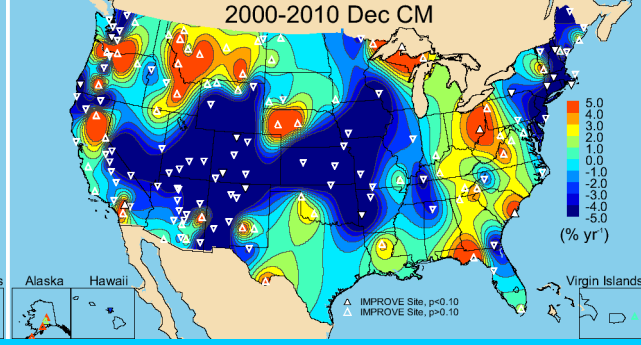
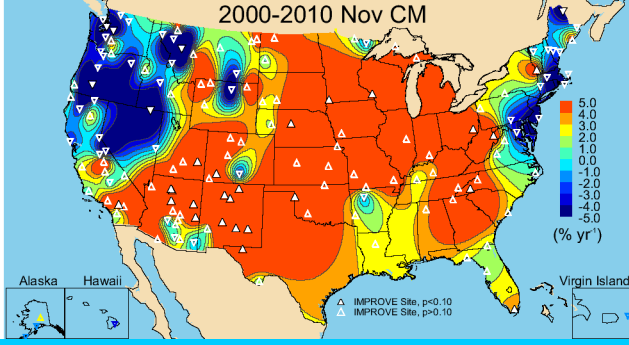
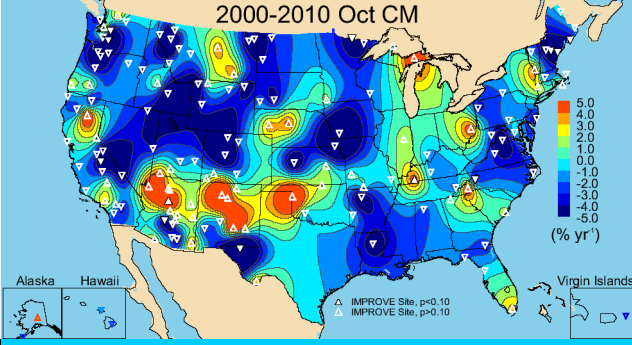
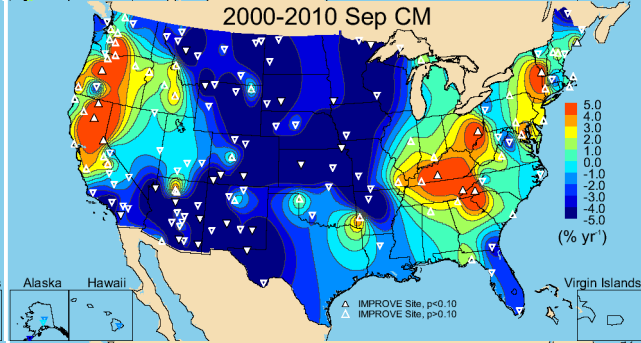
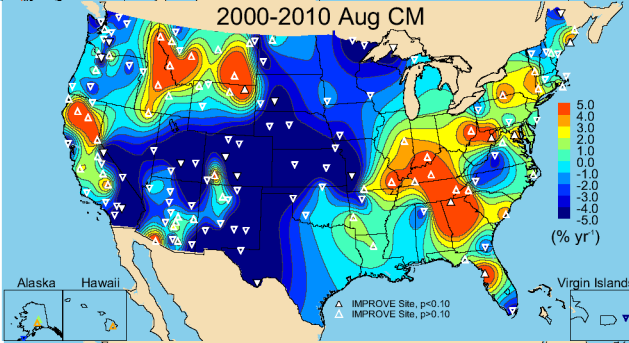
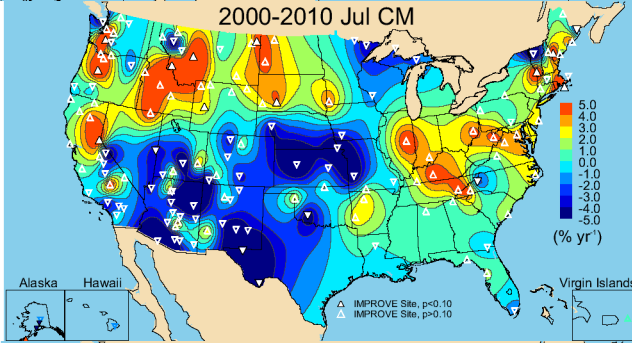
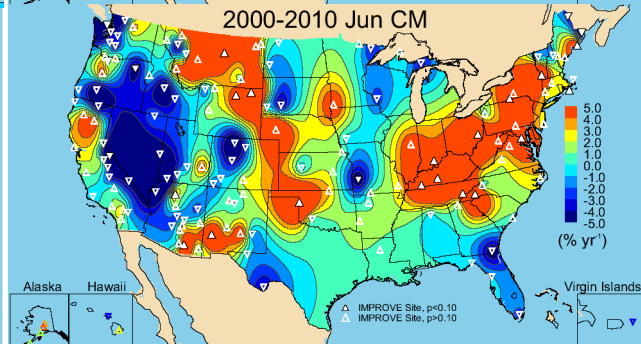
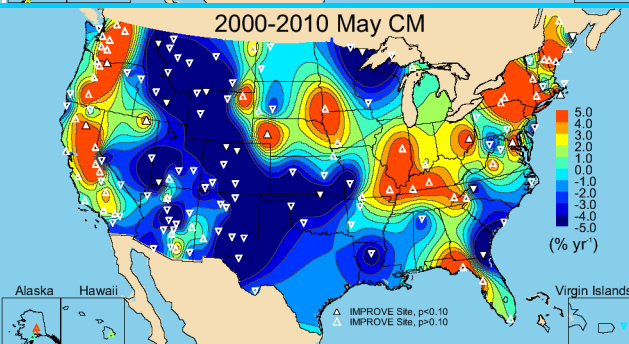
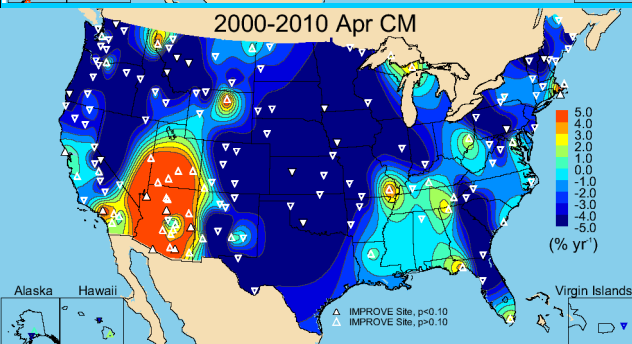
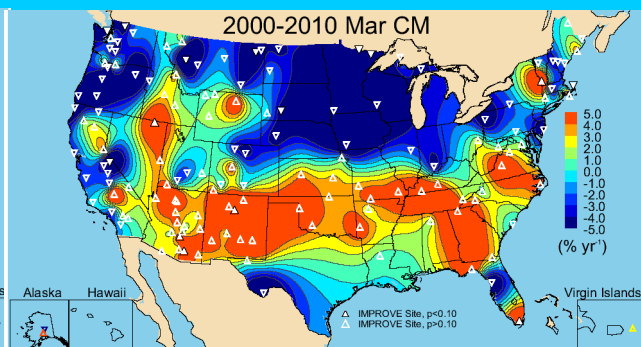
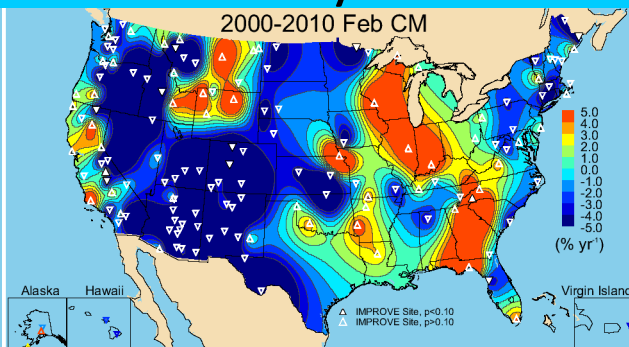
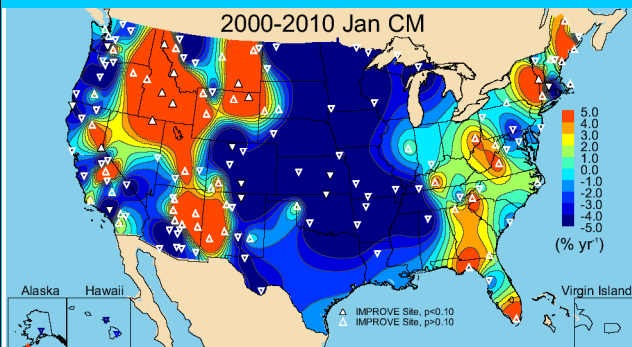
2000-2010 Monthly Mean Total Carbon Trends



2000-2010 Monthly Mean Soil Trends



2000-2010 Monthly Mean CM Trends



Standardized Anomaly

$$x' = \frac{x - \bar{x}}{s_x}$$

x' = standardized anomaly

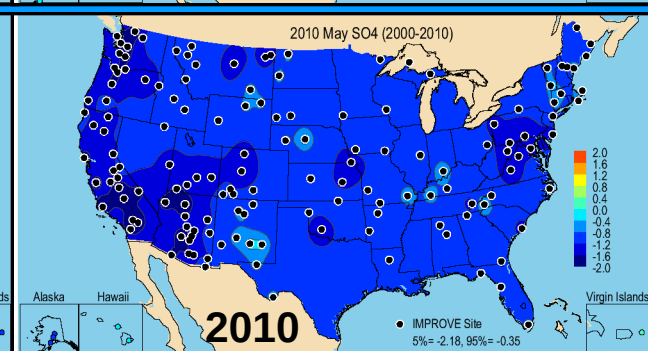
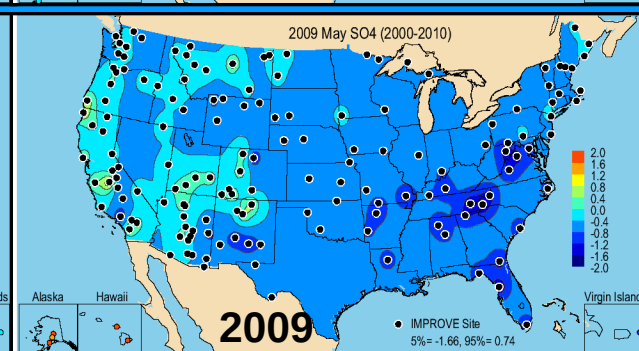
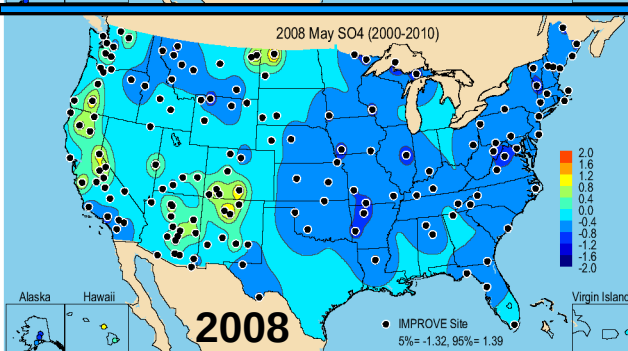
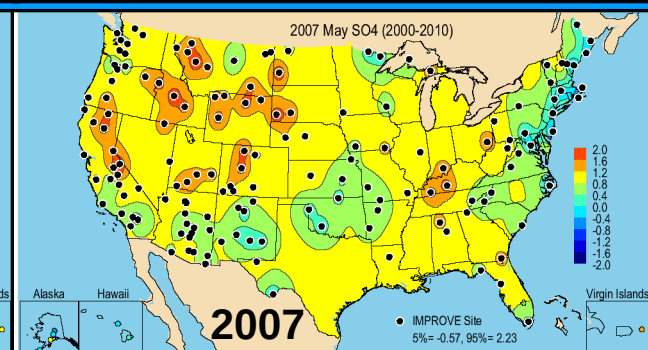
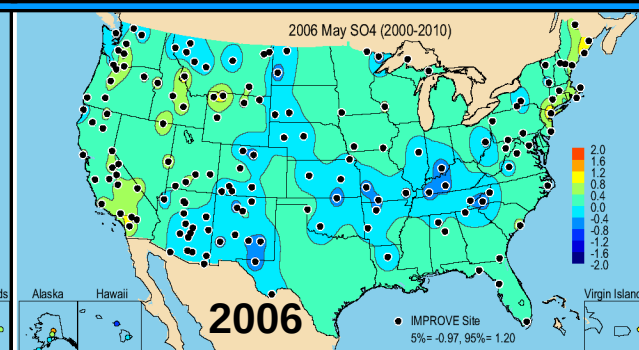
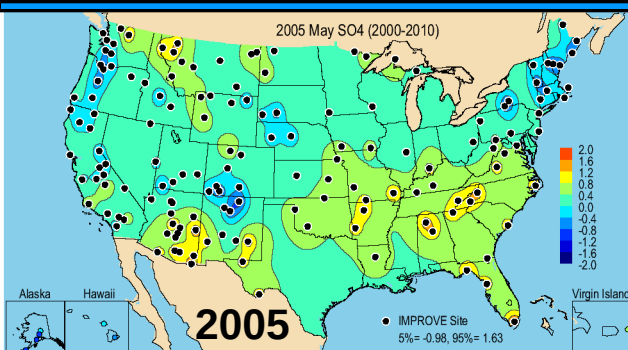
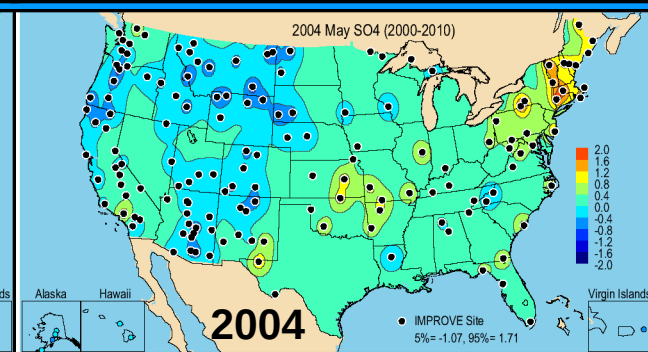
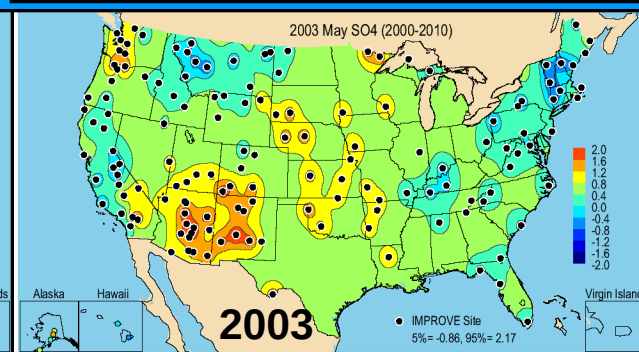
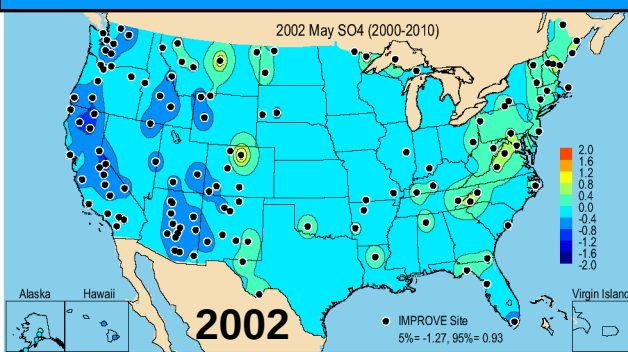
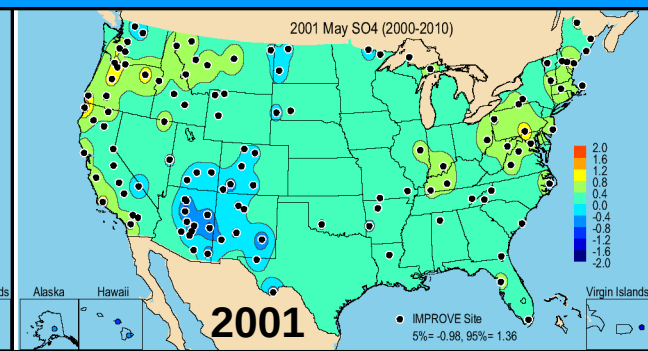
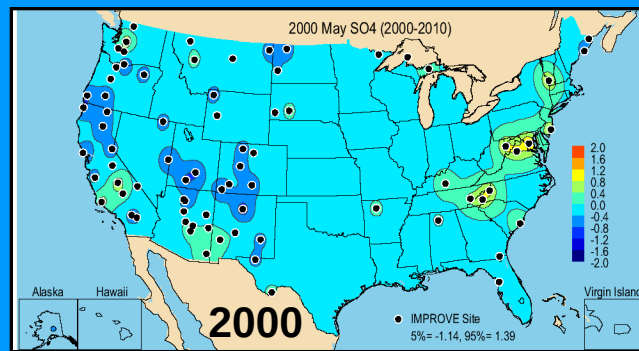
x = data (monthly mean)

$\bar{x}$ = climatological monthly mean

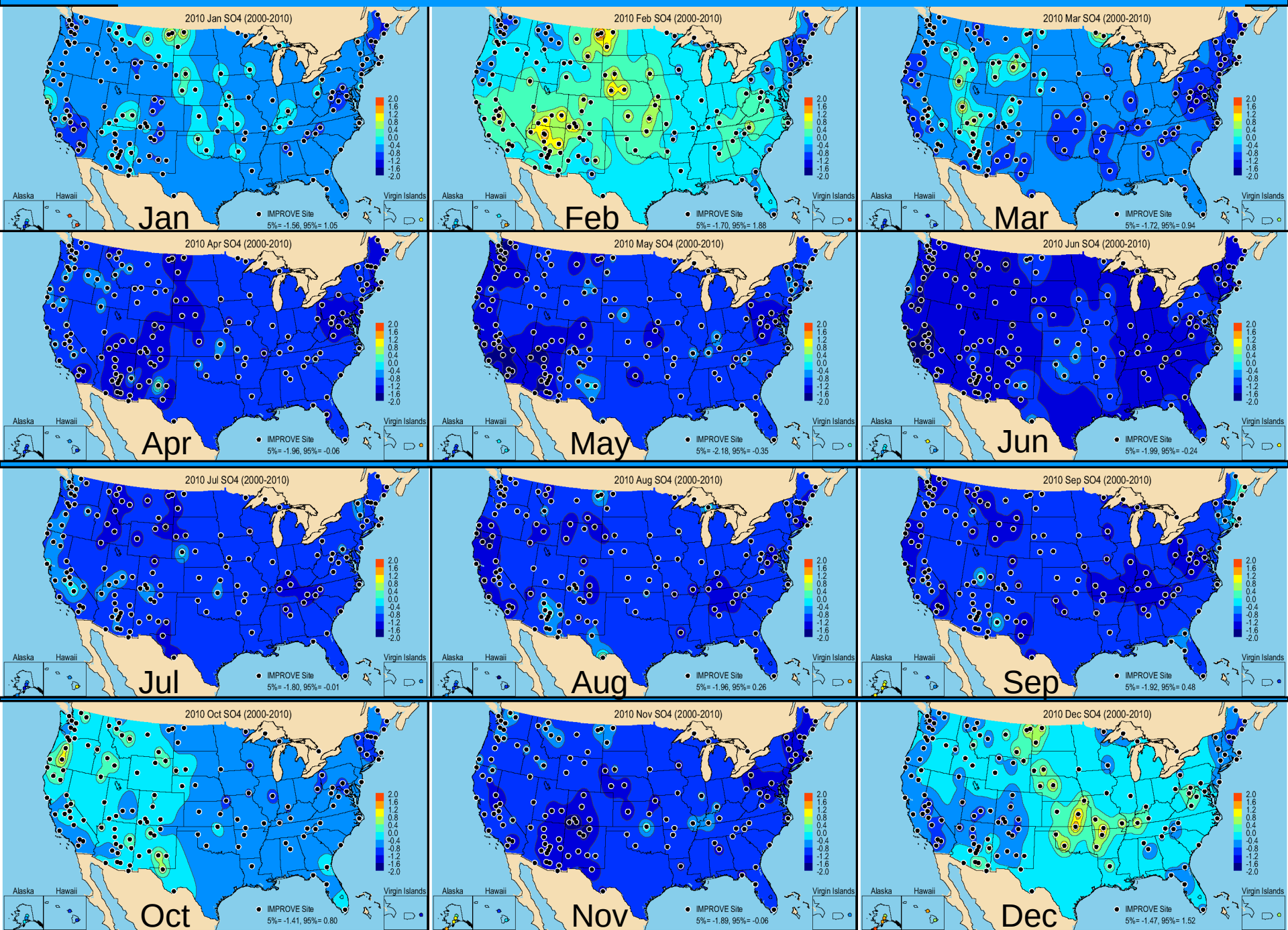
s_x = climatological standard deviation

- Removes influences of location and spread
- $x' = 0$ for mean, $x' = 1$ for 1 standard deviation

Sulfate 2000-2010 Standardized Anomaly for May

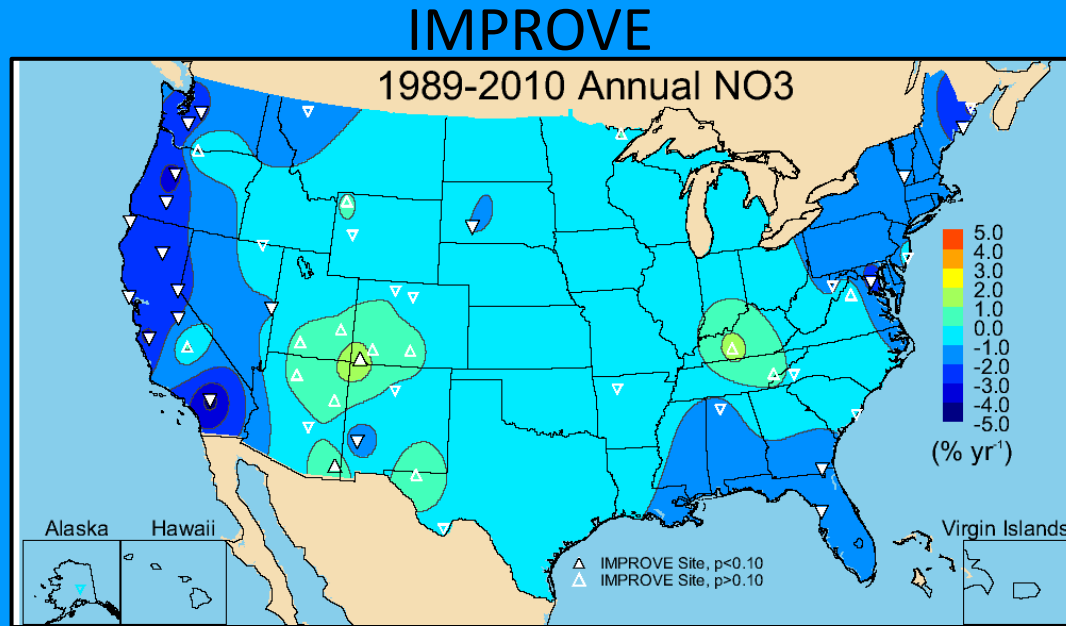


2010 Sulfate concentrations are significantly low



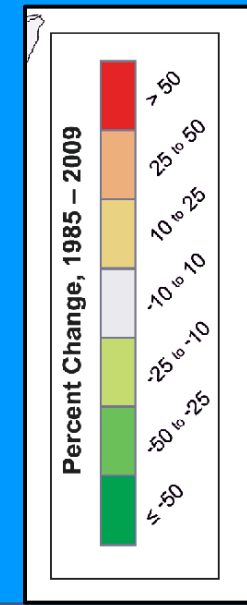
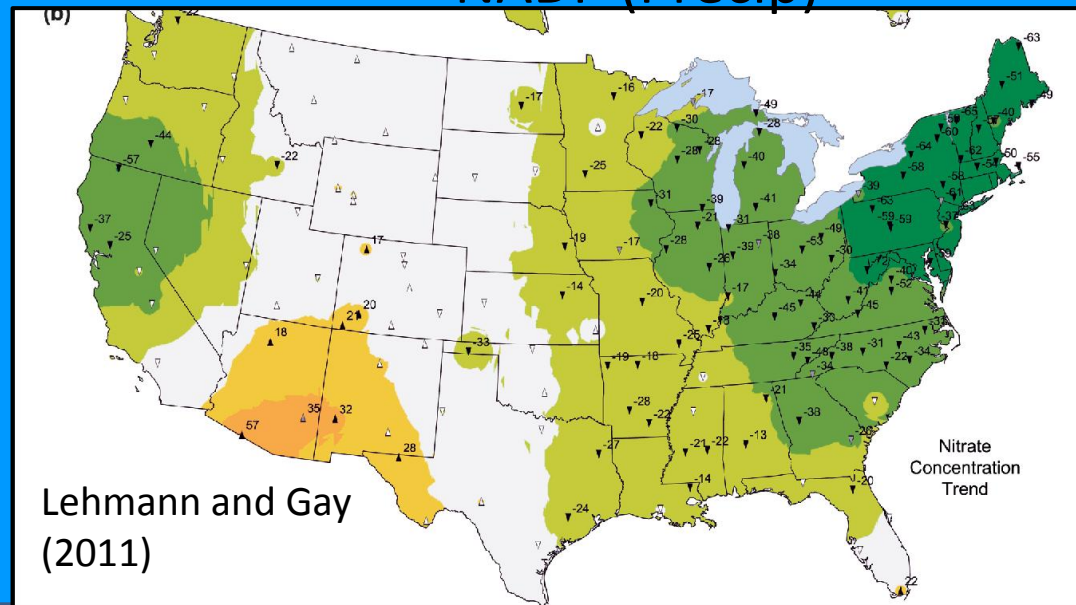
Trends in IMPROVE and NADP Nitrate

IMPROVE
1989-2010



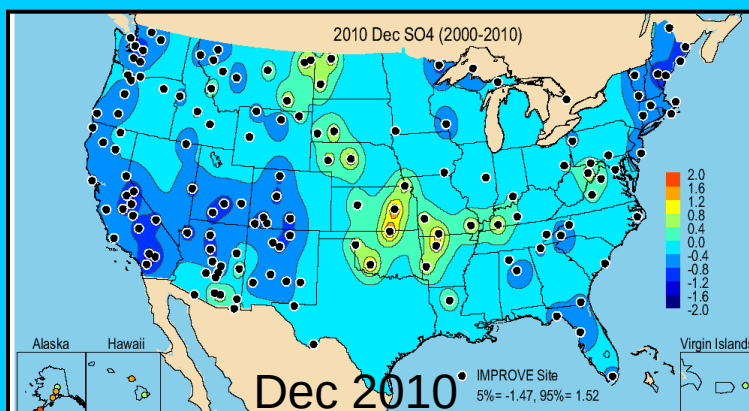
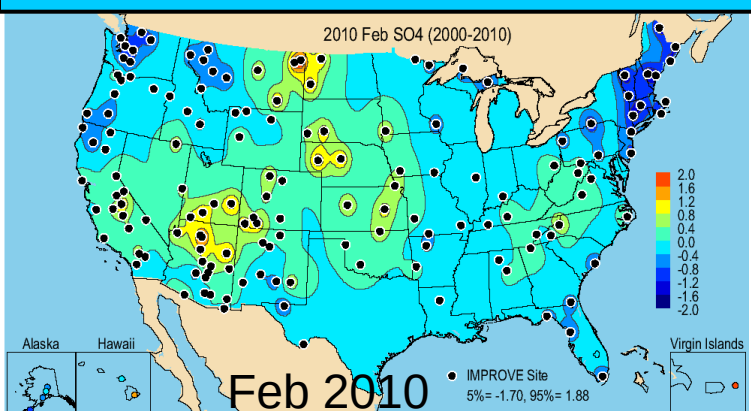
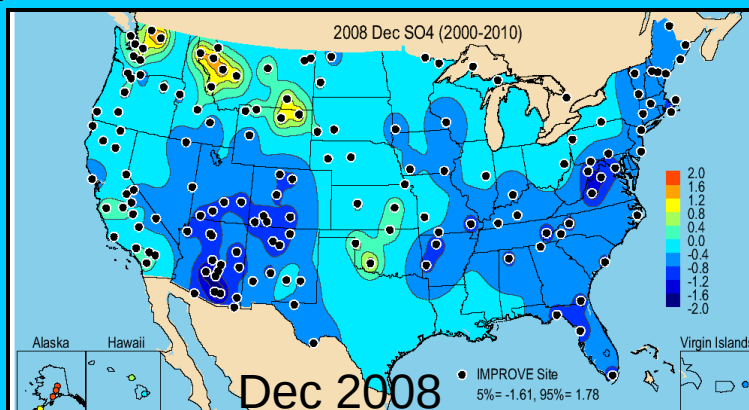
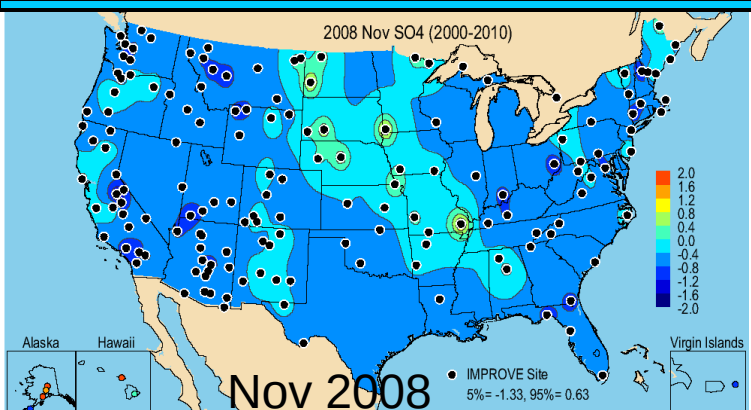
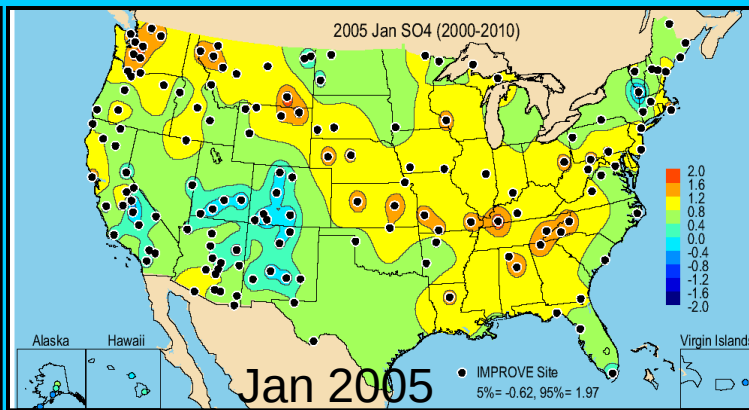
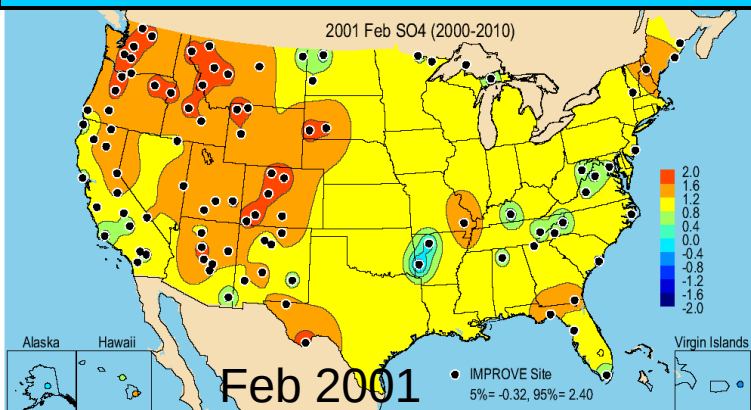
NADP (Precip)

NADP
1985-2009

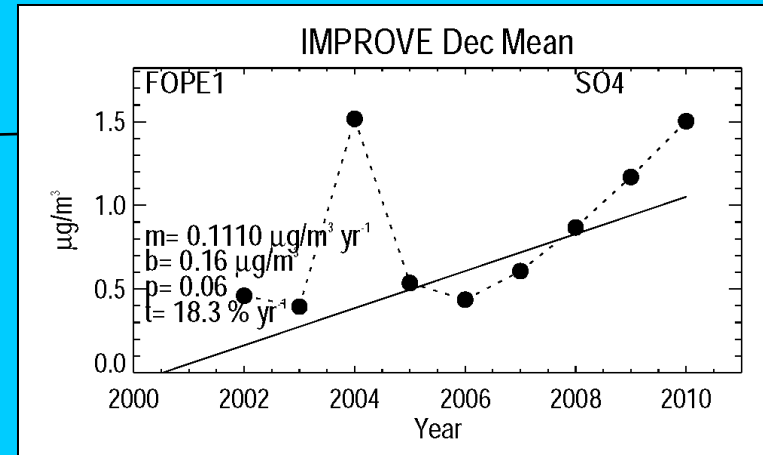
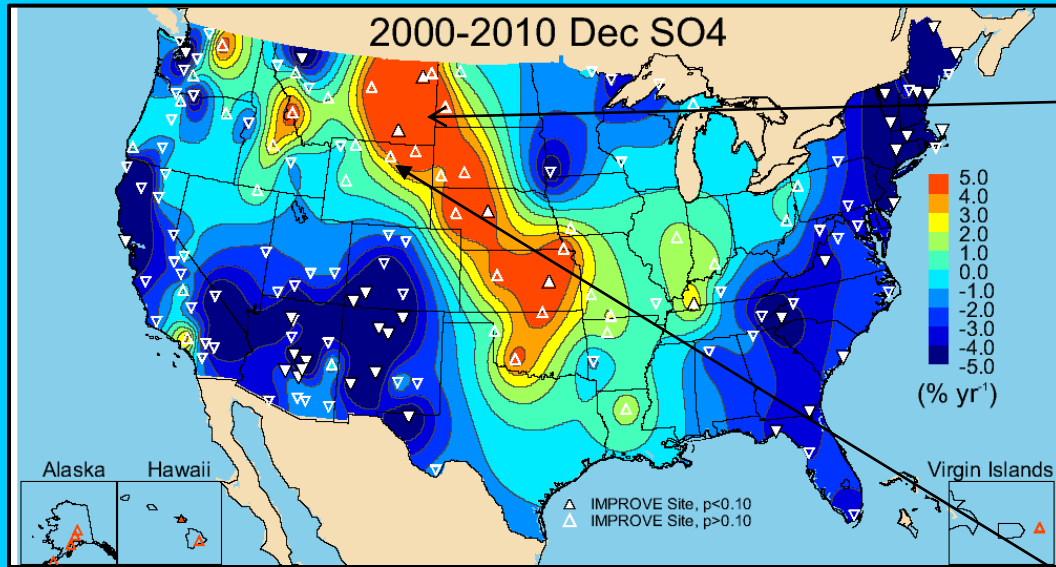


Winter Sulfate Standardized Anomalies

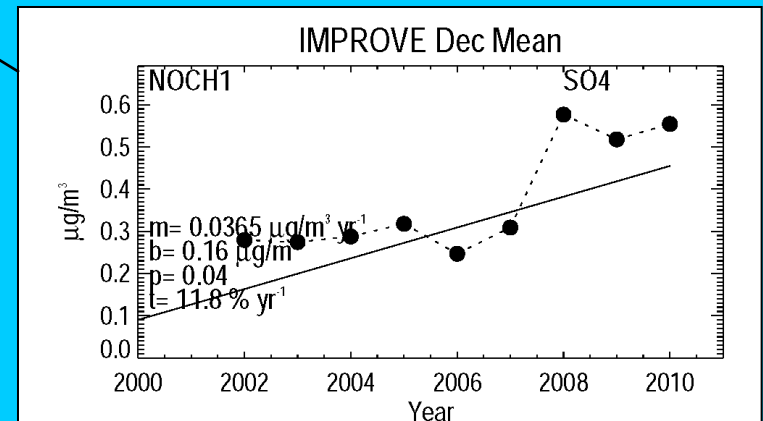
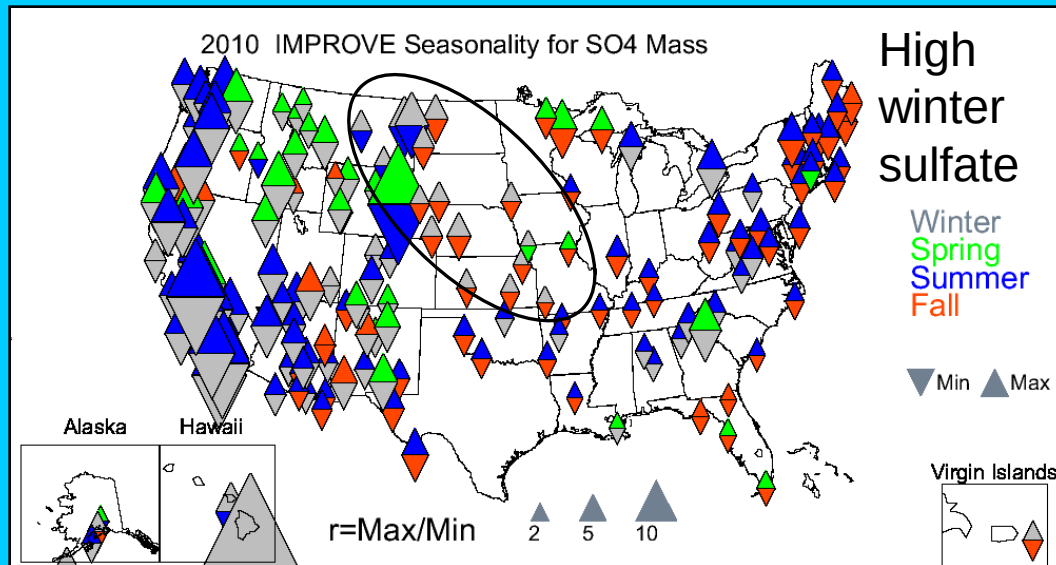
Examples of
other periods
with Midwestern
winter episodes



Winter Sulfate in the Midwest



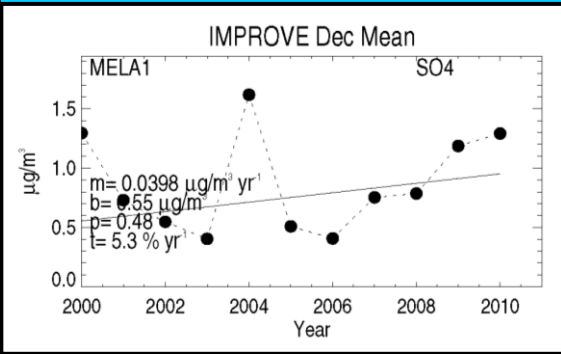
Fort Peck, MT



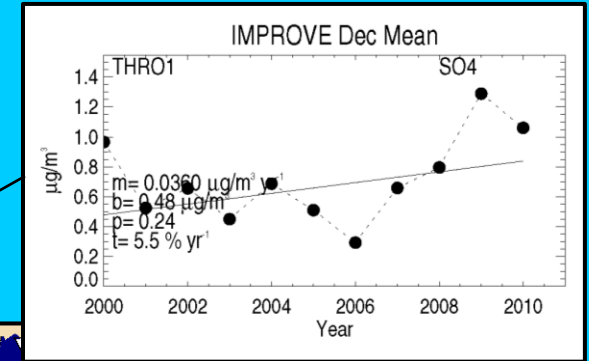
Northern Cheyenne, MT

December Mean Trends

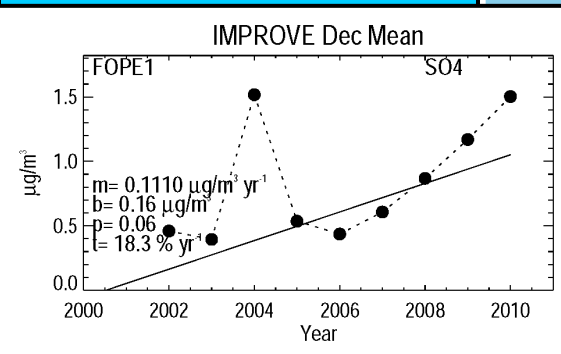
Medicine Lake, MT



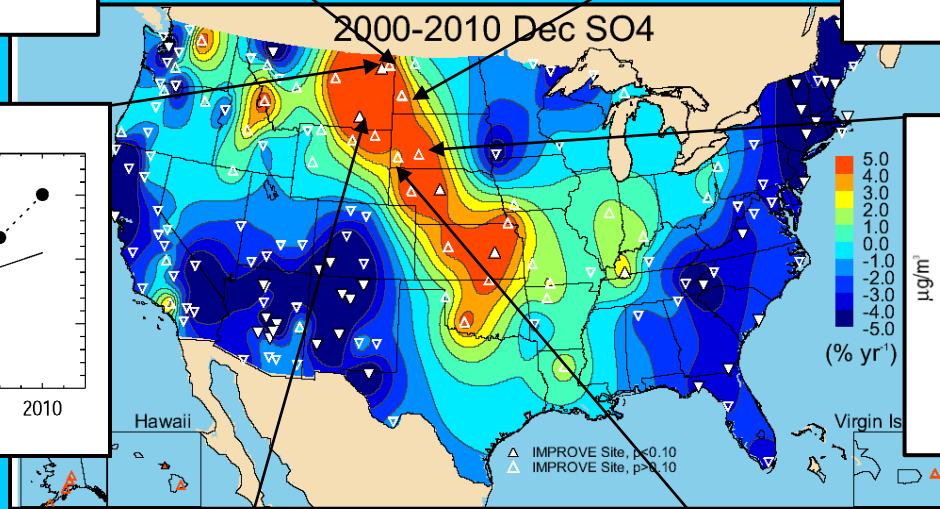
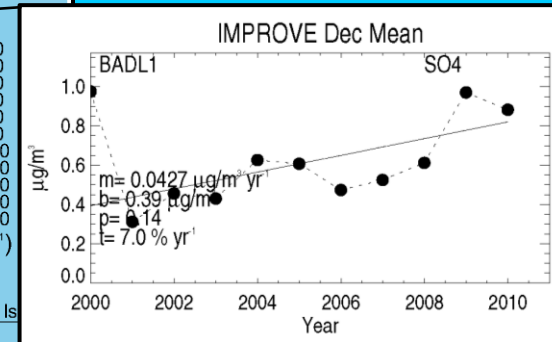
Theodore Roosevelt, ND



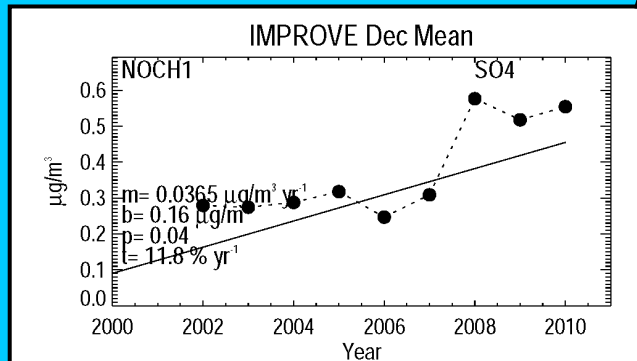
Fort Peck, MT



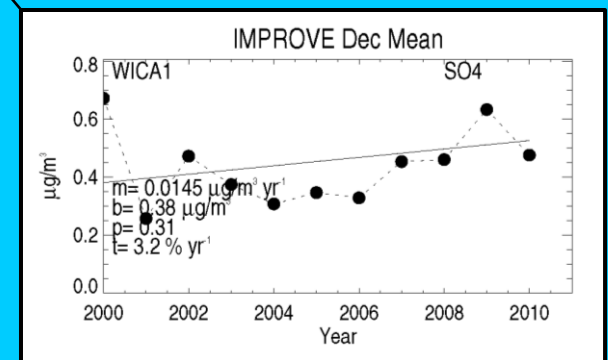
Badlands, SD



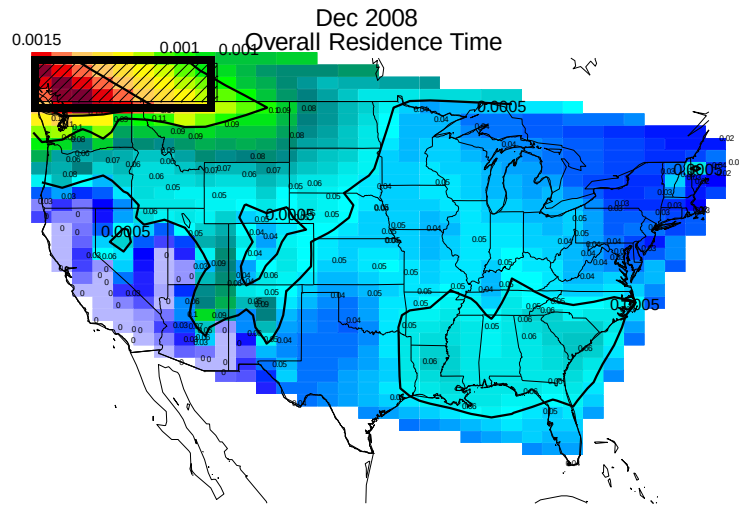
Northern Cheyenne, MT



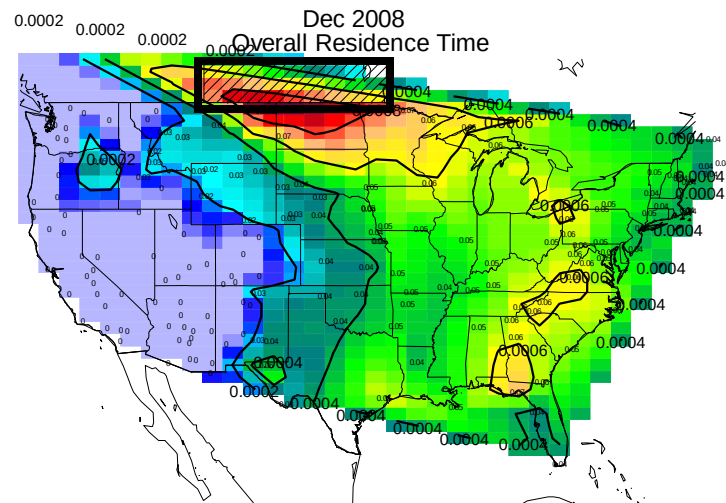
Wind Cave, SD



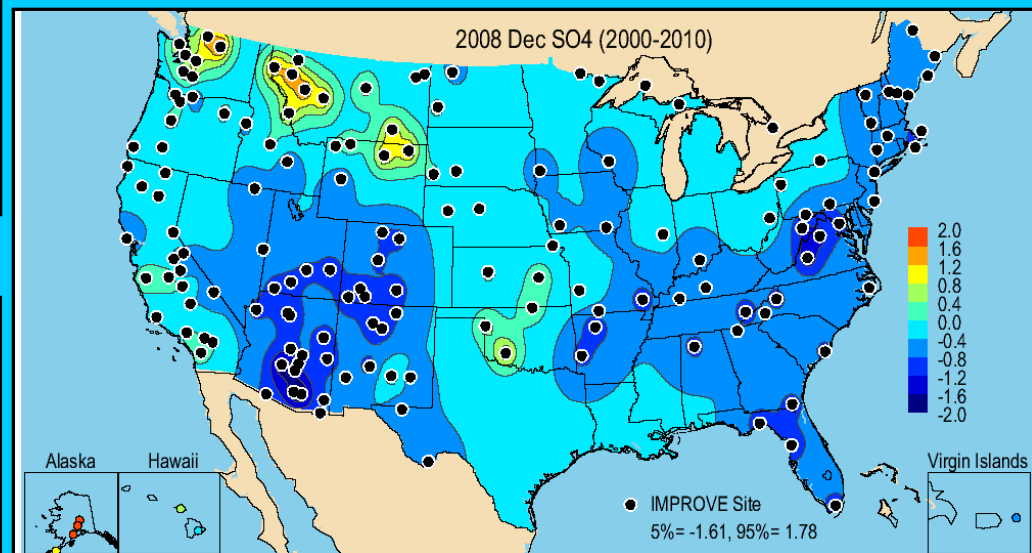
ATAD Back trajectories



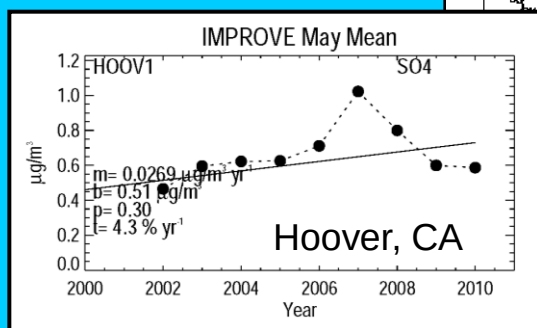
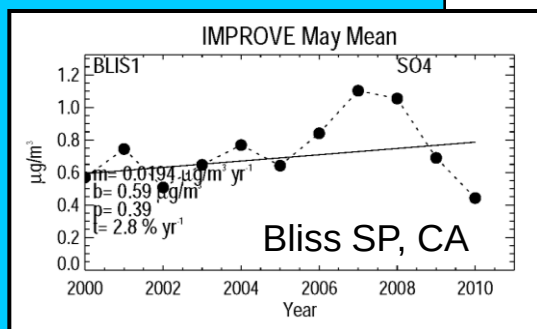
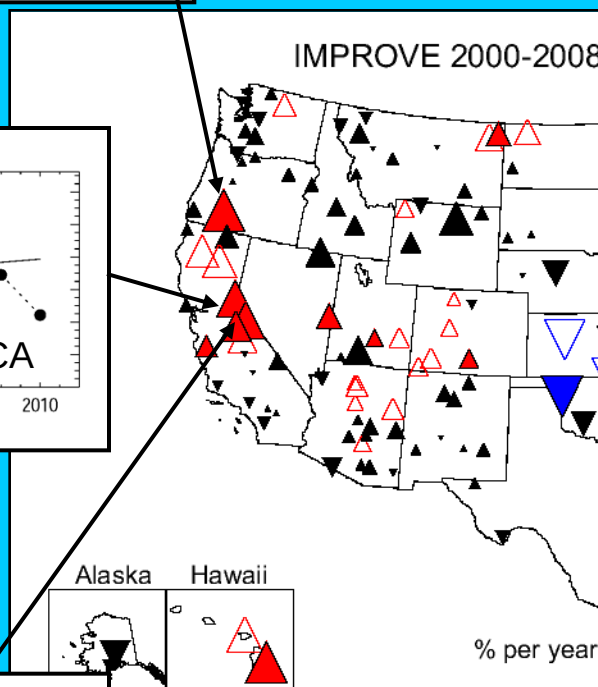
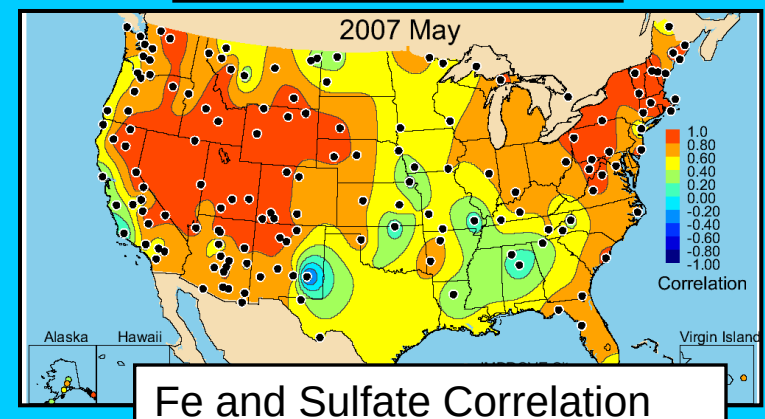
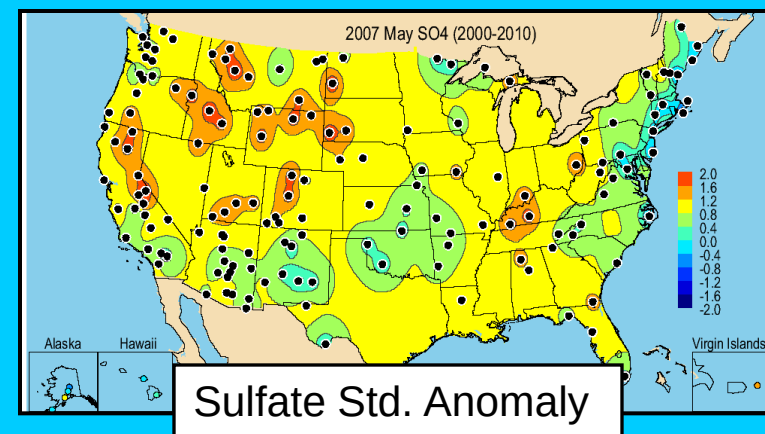
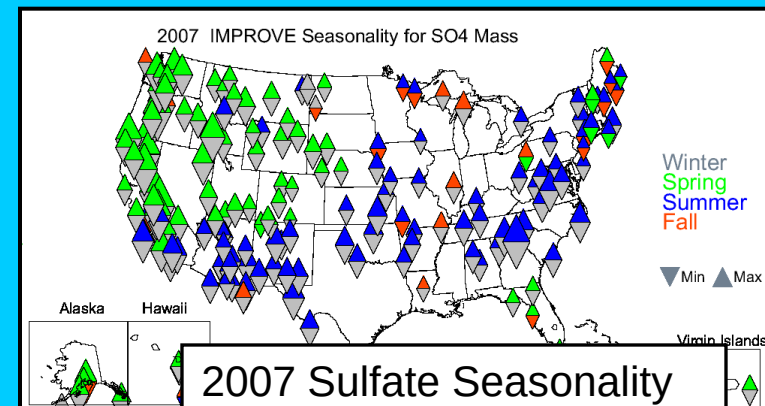
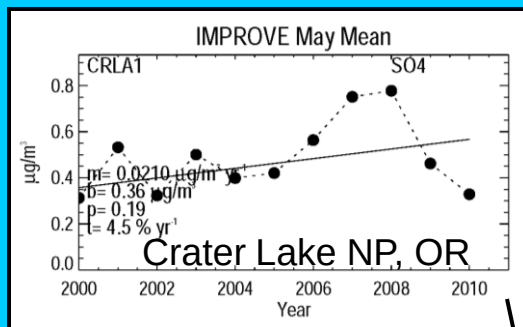
Source: CanadaABBC Lon -125 to -110 , Lat 49 to 51.5 , n.min= 1



Source: CanadaSKMN Lon -110 to -95 , Lat 49 to 51.5 , n.min= 1

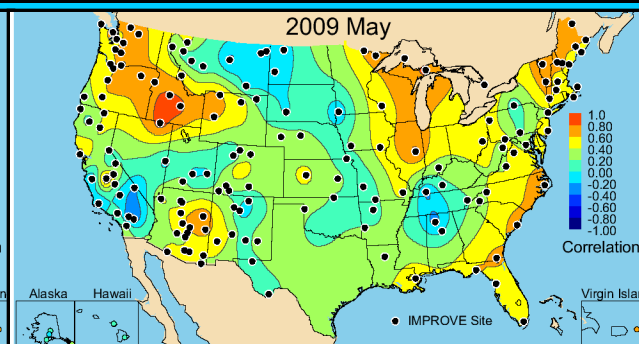
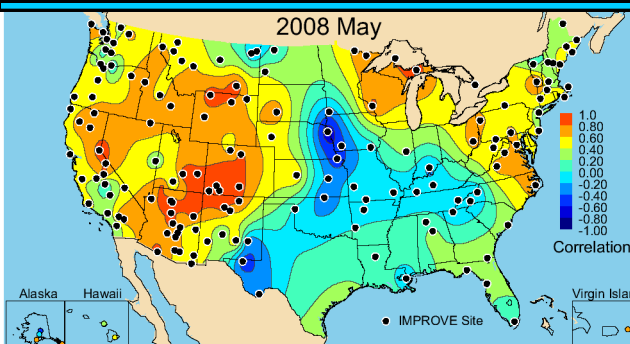
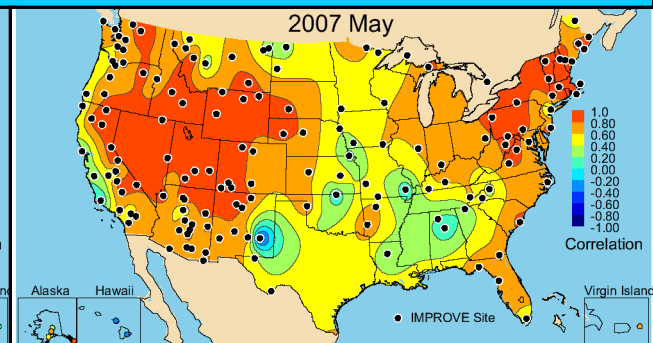
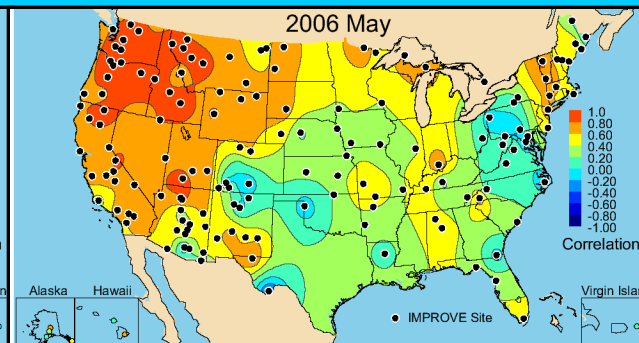
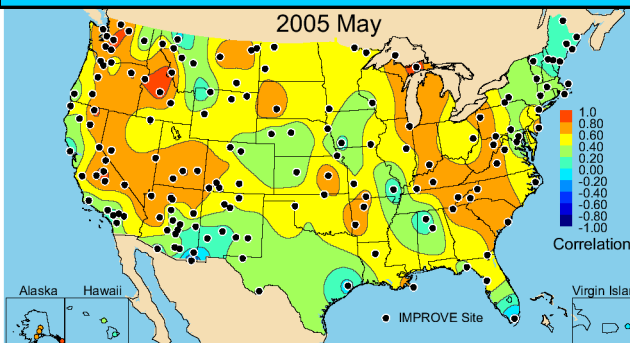
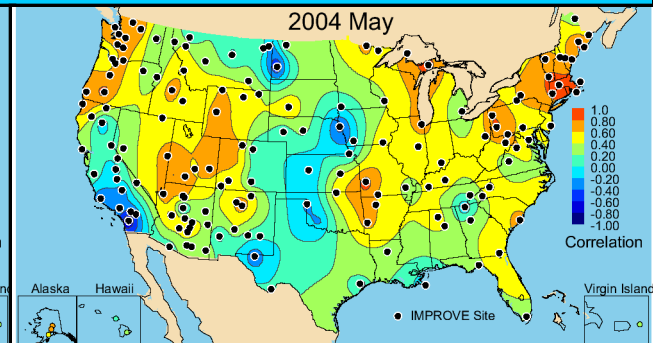
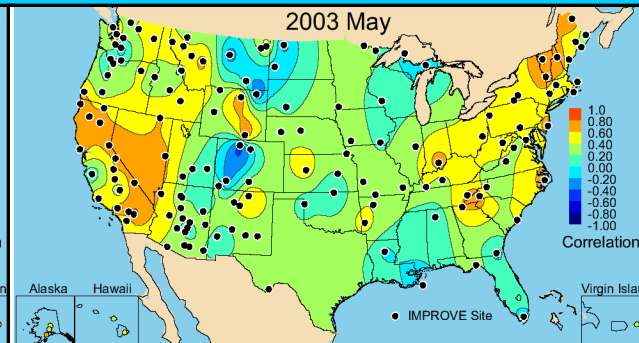
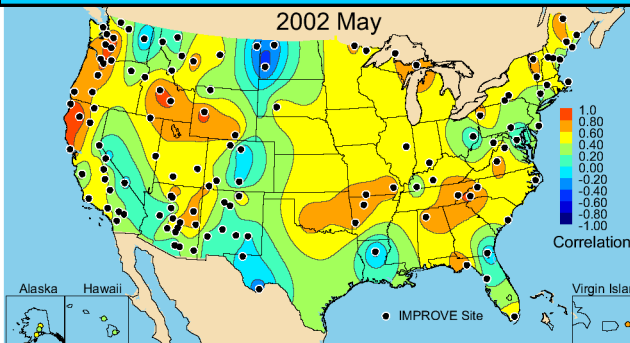
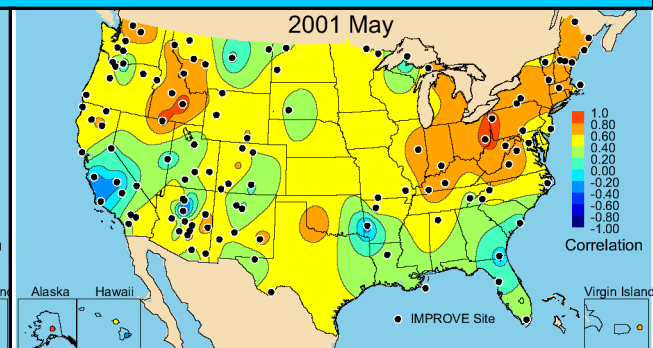
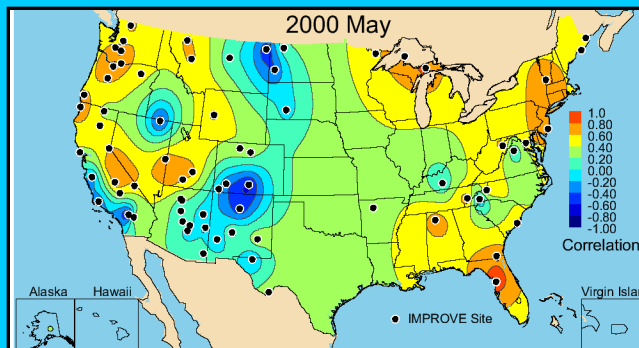


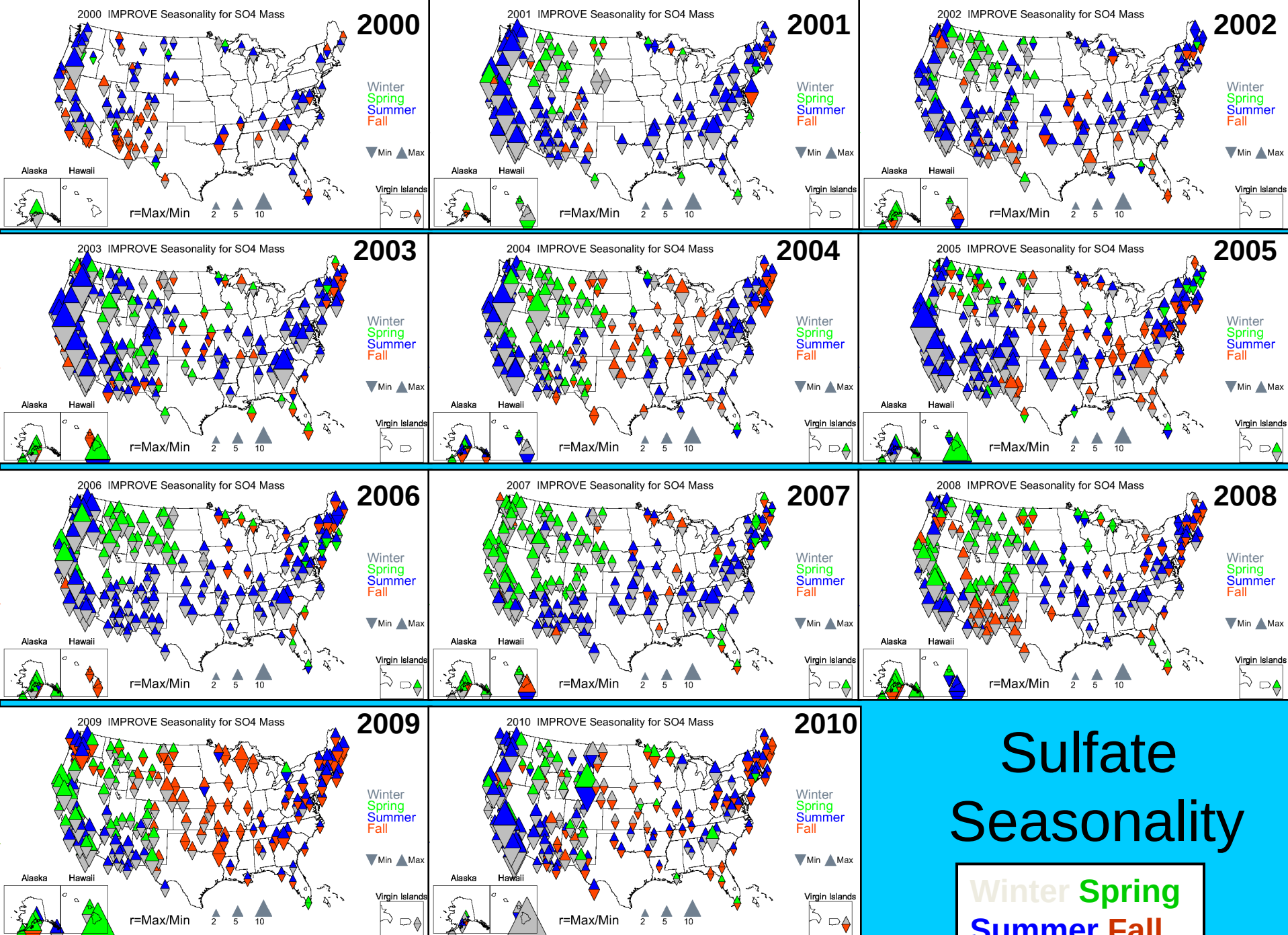
May Sulfate Patterns

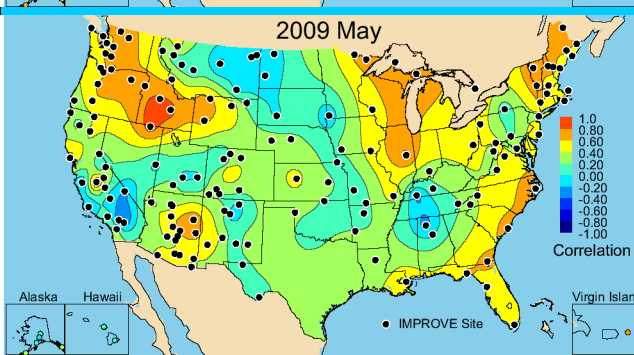
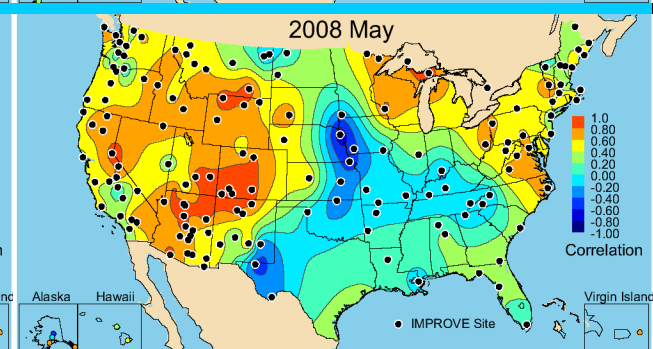
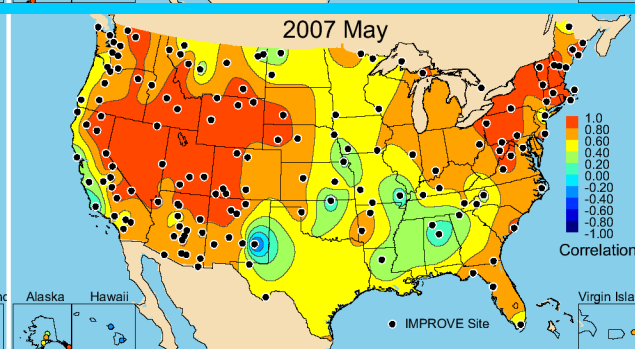
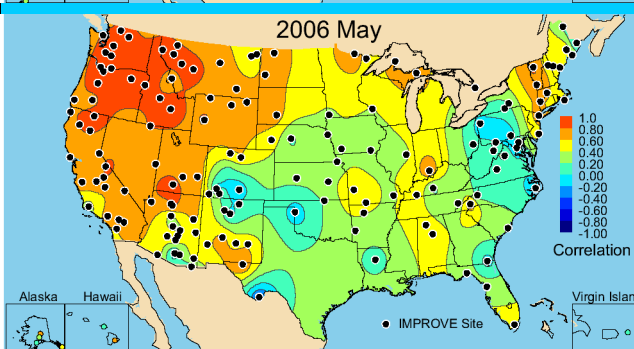
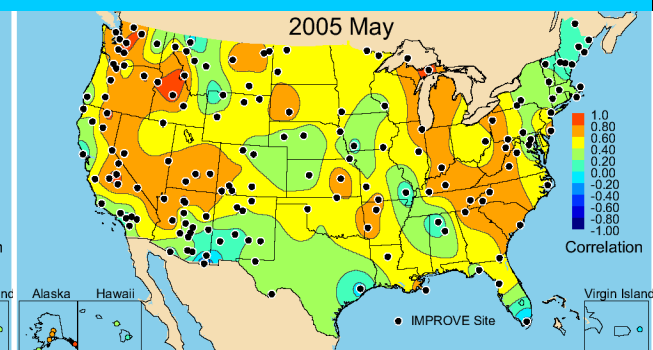
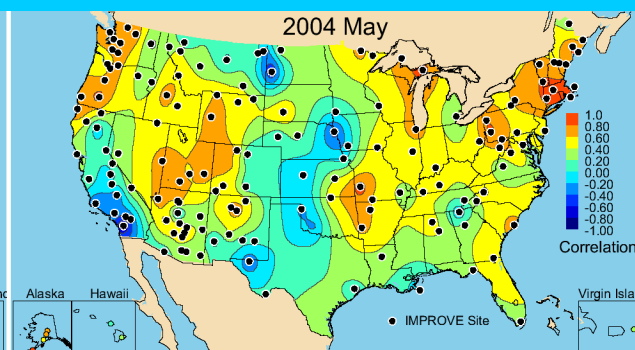
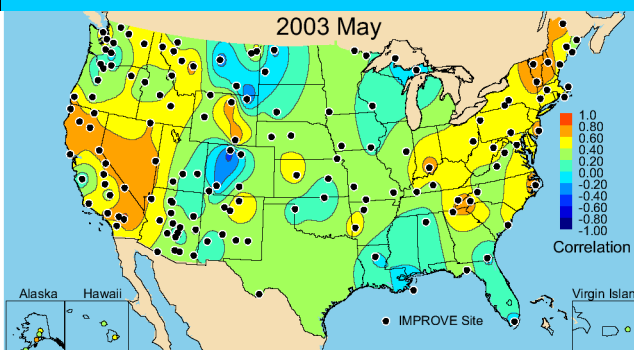
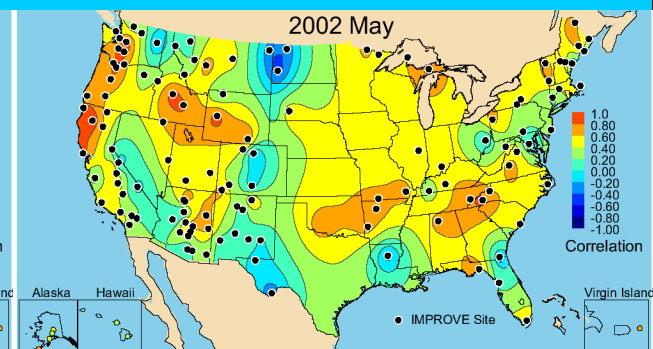
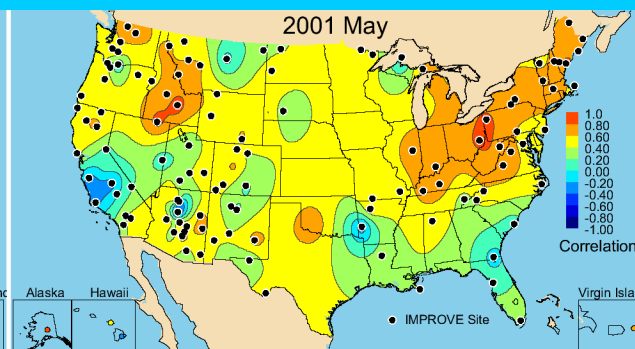
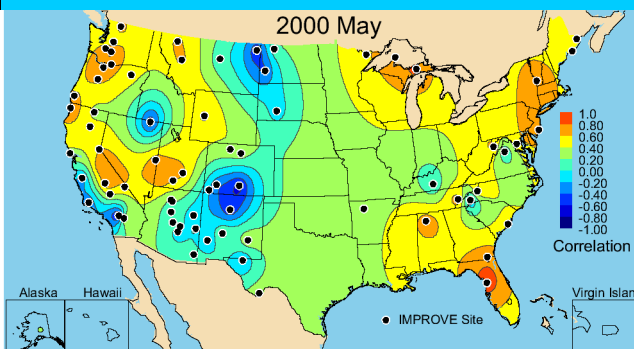


Sulfate and Iron Correlation

May 2000-2009

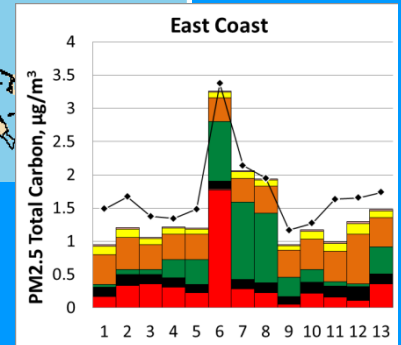
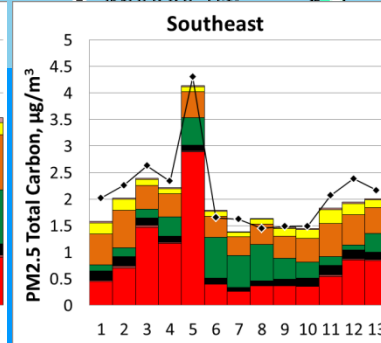
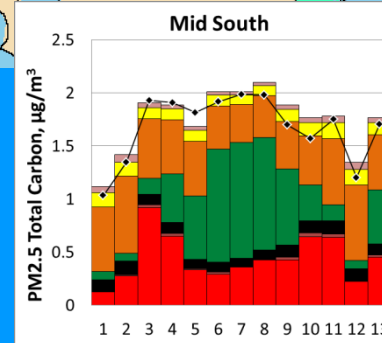
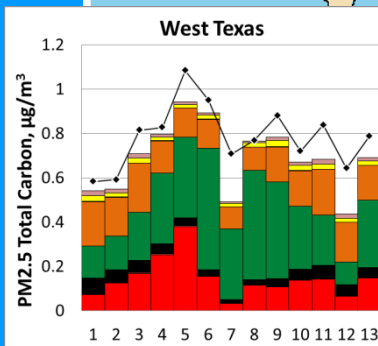
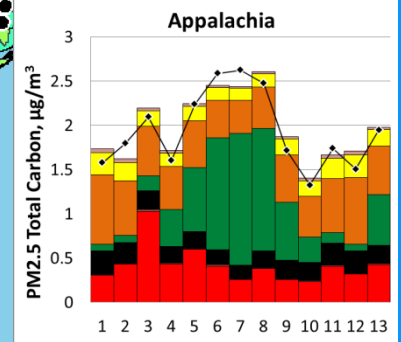
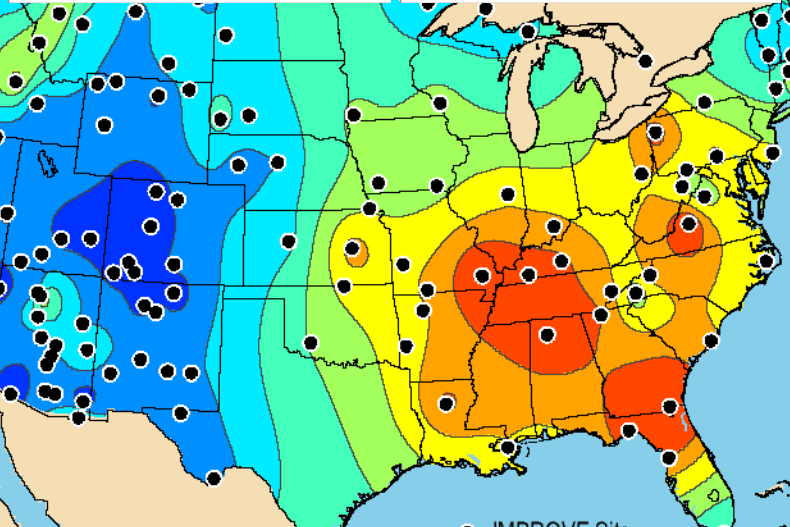
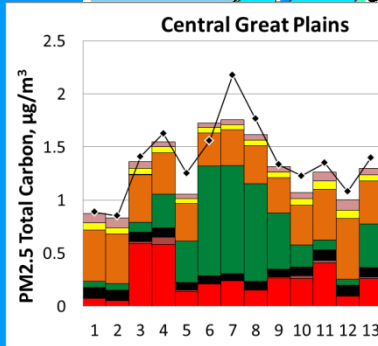
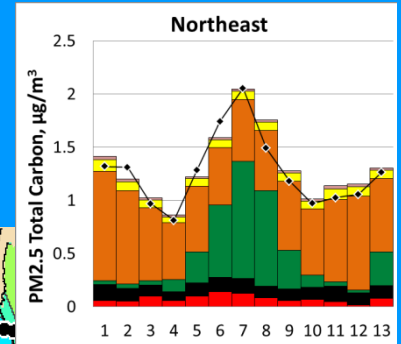
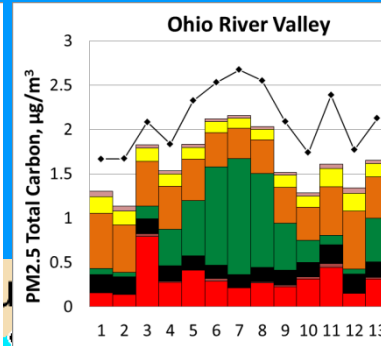
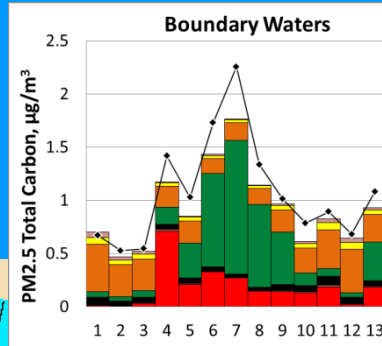
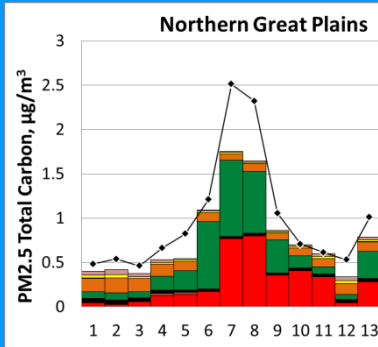




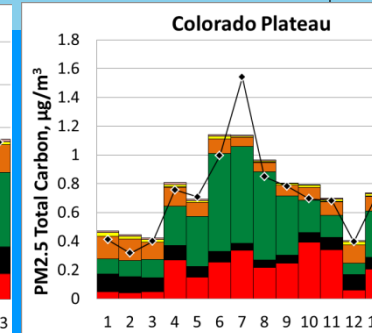
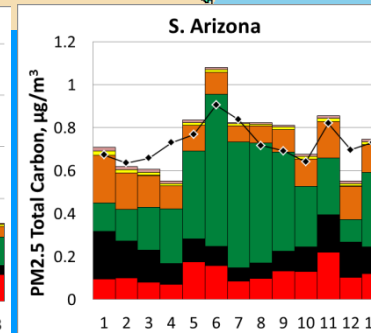
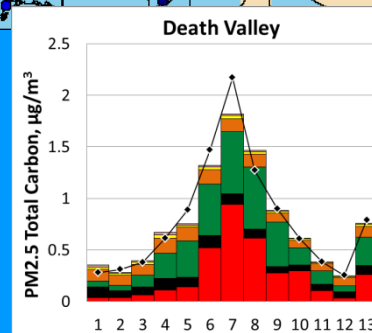
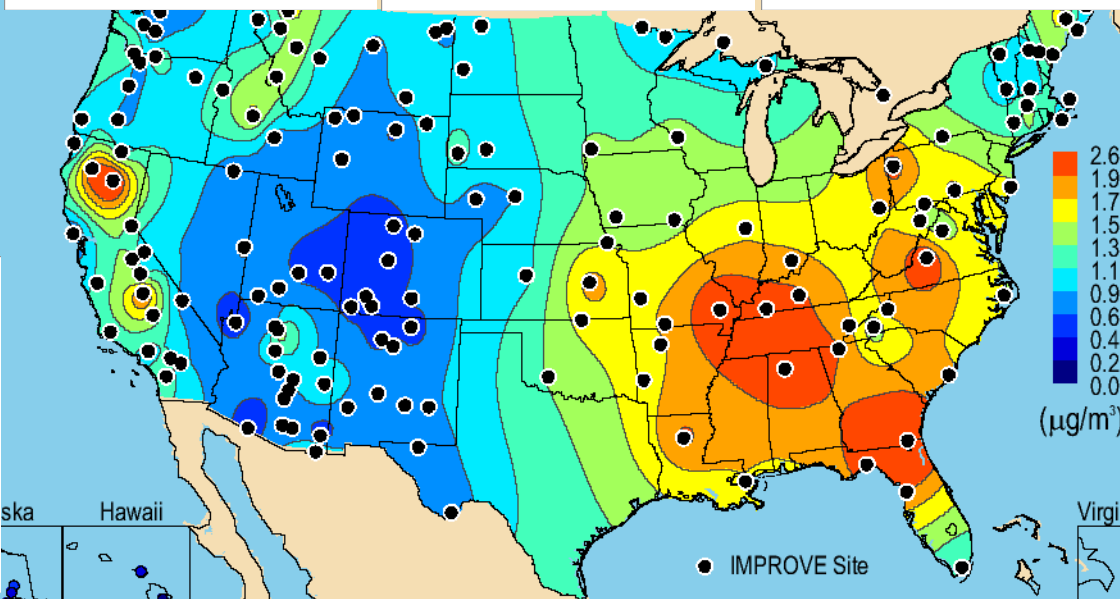
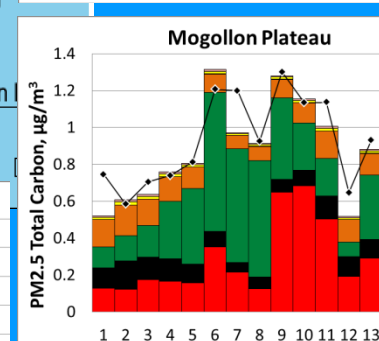
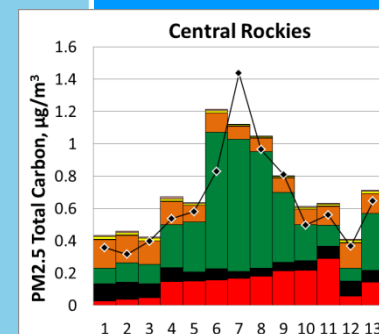
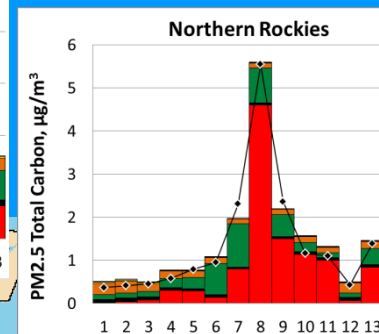
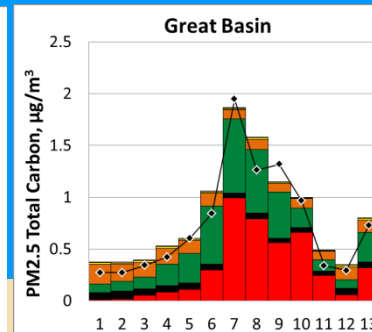
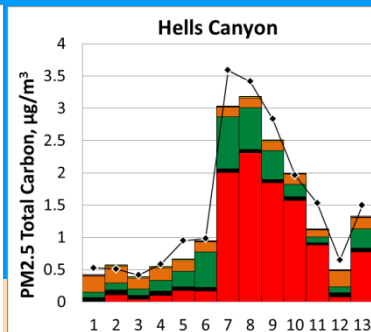
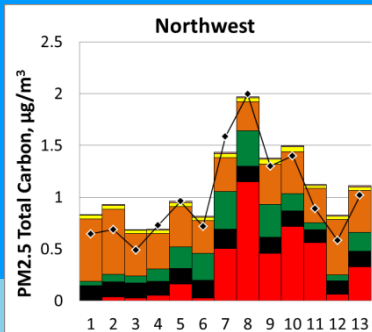
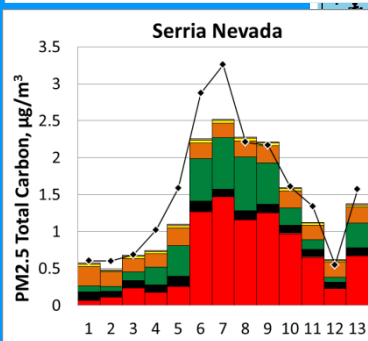
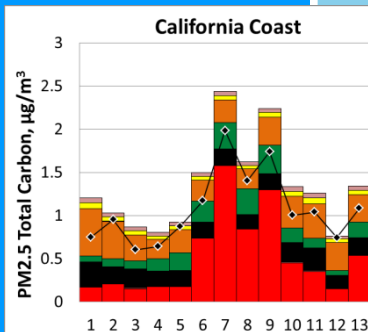
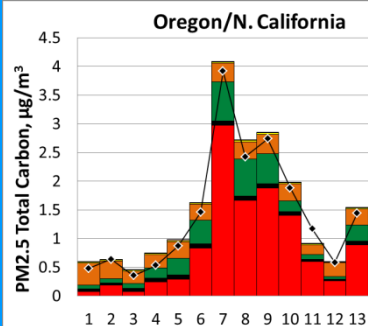
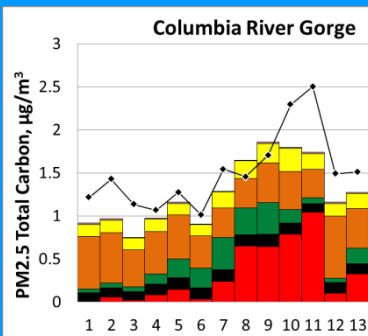


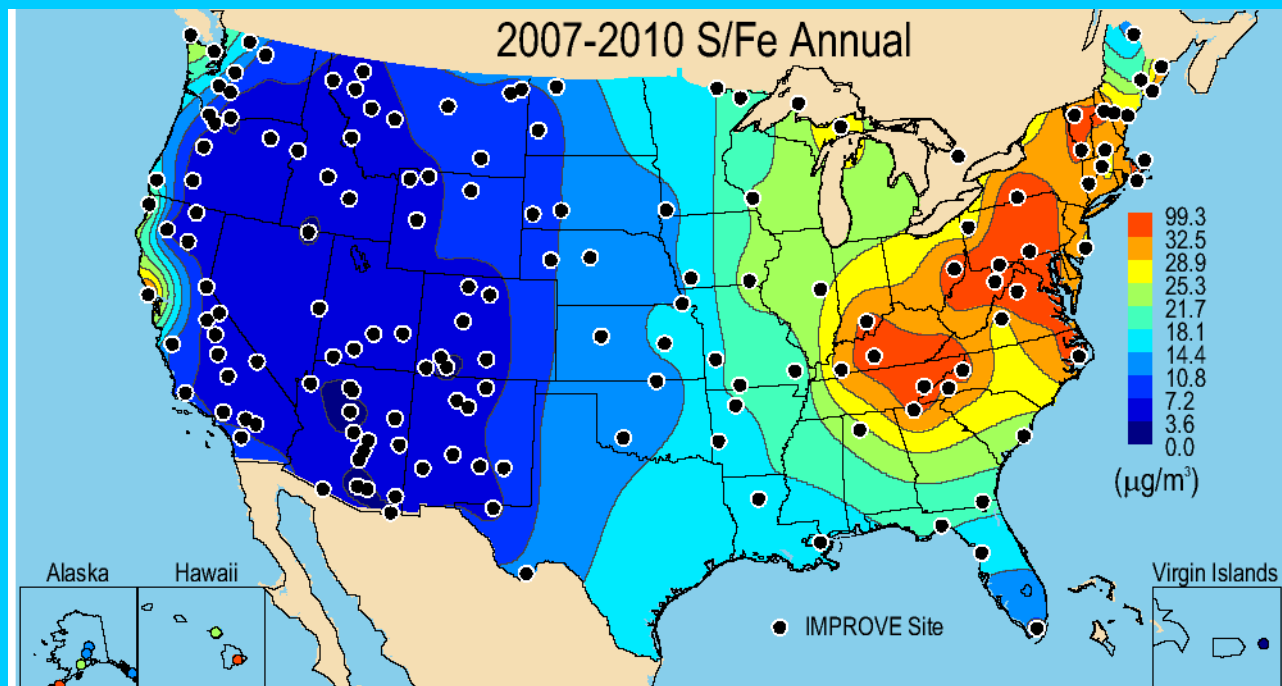
May 2000-2009

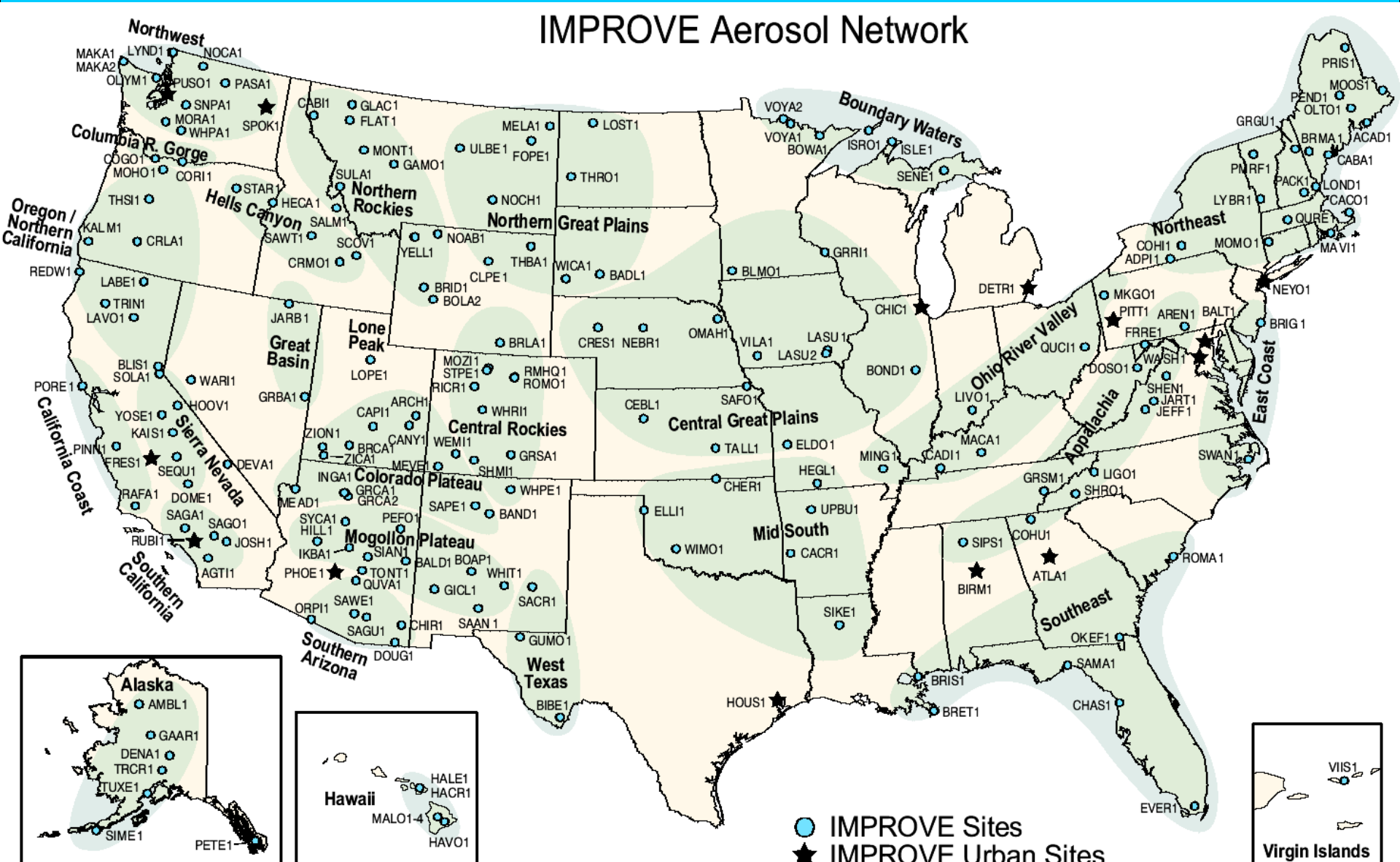
Iron and sulfate correlation



Refined 2006–08 Source App



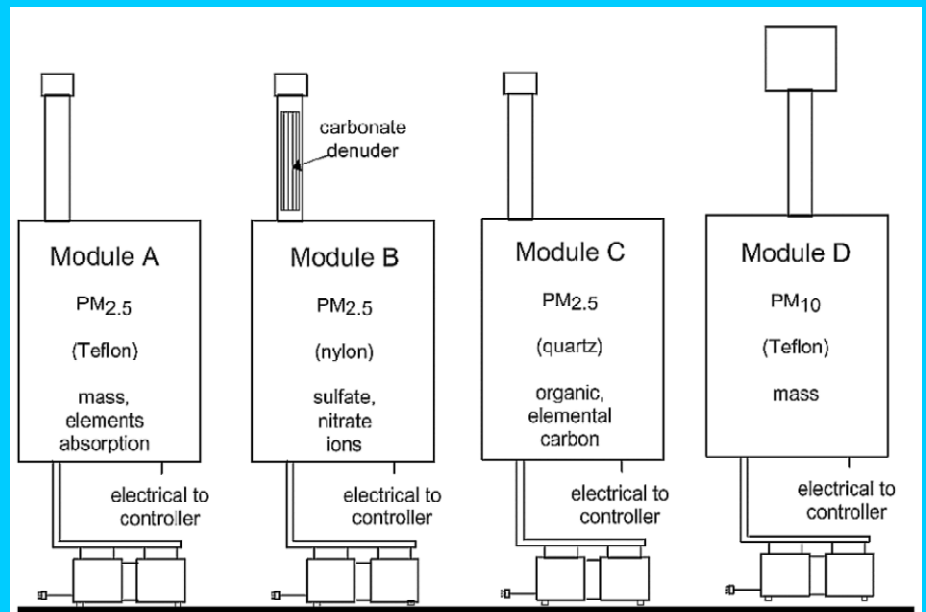




IMPROVE Monitoring Site



Big Bend National Park





JARI



SIPS



19?

Cohutta, GA

2010



DOME

- The IMPROVE program is a cooperative measurement effort designed to
 - establish current visibility and aerosol conditions in mandatory Class I areas;
 - identify chemical species and emission sources responsible for existing anthropogenic and natural visibility impairment;
 - document long-term trends for assessing progress towards the national visibility goal;
 - and, with the enactment of the Regional Haze Rule, provide regional haze monitoring representing all Class I areas where practical.

IMPROVE

- 1987: Monitoring began at ~30 sites
- 2000: Expansion of network to 159 sites in support of the Regional Haze Rule
- 2011: 170 operating and 42 discontinued sites