The Effects of SOx and NOx Emission Reductions on Sulfate and Nitrate Particulate Concentrations

by

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Introduction

The Regional Acid Deposition Model / Regional Particulate Model (RADM / RPM) was used to model the effects of large reductions in sulfur oxide (SOx) and nitrogen oxide (NOx) emissions. RADM/RPM was used to simulate atmospheric sulfate and nitrate concentrations. The performance of RPM in replicating observed concentrations has not been evaluated extensively. Some preliminary evaluations indicate that RPM performs well in reproducing observed sulfate particulate concentrations, but RPM appears to underpredict nitrate particulate concentrations by approximately a factor of 2. The base emissions inventory for these reductions was the National Particulate Inventory (NPI) Version 2. Adjustments were made to the ammonia inventory to correct for underestimation. Primary direct PM-10 and PM-2.5 emissions are not included in the RADM/RPM modeling. The RADM / RPM was used to simulate atmospheric concentrations for the base 1990 emissions inventory, a scenario with a 75% across-the-board reduction in SOx emissions, and a scenario with a 75% across-the-board reduction in SOx and NOx emissions. The modeling was performed for the eastern half of the United States with 80-kilometer on a side grid squares. The 1330 grid square modeling domain is shown in Figure 1. Thirty 3-day meteorological periods spanning the four seasons of the year were modeled with RADM / RPM. The model results for the 90 days were then averaged to approximate annual averages. Sulfate and nitrate particulate concentrations were compared for the base case and the 75% reduction scenarios.

Results

The 1990 base case sulfate particulate concentrations are shown in Figure 2. Note that the results in this figure and all other color plots represent annual averages, so disregard the January 0, 1991 date and hour that appears on these plots. The maximum sulfate particulate concentrations of 8 to 12 micrograms per cubic meter (ug/m3) occur near the center of the modeling domain. Sulfate particulate concentrations are smallest, less than 2 ug/m3, in the northwestern portion of the modeling domain. The 1990 base case nitrate particulate concentrations are shown in Figure 3. The maximum nitrate particulate concentrations of 1 to 1.5 micrometers per cubic meter occur in the western half of the modeling domain. Nitrate particulate concentrations in the extreme eastern portion of the modeling domain were generally less than 0.25 ug/m3. Comparing figures 2 and 3 show that base case 1990 sulfate particulate concentrations are much higher than nitrate particulate concentrations.

The percent reduction in the RADM/RPM annual sulfate plus nitrate particulate concentrations when SOx emissions are reduced 75% across the board are shown in Figure 4. Reducing SOx emissions by 75% reduces sulfate plus nitrate particulate concentrations by greater than 55% in the majority of the modeled area. The percent reduction in sulfate plus nitrate concentrations when SOx and NOx emissions are reduced 75% across the board are shown in Figure 5. Comparing Figures 4 and 5 shows that in all areas of the country a greater percent reduction in sulfate plus nitrate particulate concentrations was achieved when NOx was reduced along with SOx by 75%. The further percent reduction in sulfate plus nitrate concentrations over the base case concentrations when NOx is reduced in addition to SOx by 75% percent are shown in Figure 6. In much of the eastern United States the added benefit of the NOx reduction is fairly small, less than 4 percent. In the western portion of the modeling domain, where base case nitrate concentrations are highest, the added benefit of the NOx reduction on sulfate plus nitrate concentrations ranges from 12 to 24 percent over a large area. In Figure 7 each data point represents the concentration in ug/m3 for each of the 1330 grid squares in the modeling domain. The vertical axis corresponds to the sulfate plus nitrate particulate concentrations for the 75% reduction in SOx emissions scenario and the horizontal axis corresponds to the sulfate plus nitrate particulate concentrations for the 75% reduction in SOx and NOx scenario. The left line is a regression line through the center of the 1330 points. The line further to the right is a one-to-one correspondence line. Examining the regression line in Figure 7 shows that reducing NOx emissions in addition to SOx emissions by 75 percent leads to on average about a 12 percent reduction in sulfate plus nitrate particulate concentrations when considering all grid squares in the modeling domain. Figure 8 shows the added percent reduction in sulfate plus nitrate concentrations of a 75% reduction in NOx over the 75% reduction in SOx. Although as supported by Figure 7 the average added percent reduction in sulfate plus nitrate by reducing NOx by 75% is approximately 12 percent, the added benefit is generally 5 to 10 percent in a large area of the eastern portion of the modeling domain and 20 to 30 percent in a large area of the western portion of the modeling domain where nitrate concentrations are highest.

The percent reduction in the RADM/ RPM annual sulfate particulate concentrations when SOx emissions are reduced 75% across-the-board are shown in Figure 9. The percent reduction in sulfate particulate concentrations when SOx and NOx emissions are reduced 75% across-the-board are shown in Figure 10. The percent reductions in Figures 9 and 10 are nearly identical with a 65% or greater reduction across much of the eastern United States. It had been postulated that a decrease in NOx emissions could theoretically lead to a significant increase in sulfate particulate concentrations. Since Figure 10 is almost identical to Figure 9, that effect was not shown in these model results. This is further supported in Figure 11 where each data point represents the concentration in ug/m3 for each of the 1330 grid squares in the modeling domain. The vertical axis corresponds to the sulfate particulate concentrations for the 75% reduction in SOx emissions scenario and the horizontal axis corresponds to the sulfate particulate concentrations for the 75% reduction in SOx and NOx scenario. The regression line through the center of the 1330 points is on top of the one-to-one correspondence line. Thus, the results in Figure 11 show a strong one-to-one relationship in sulfate particulate concentrations for the two scenarios. This again indicates that NOx emission reductions do not lead to an increase in sulfate particulate concentrations.

In the presence of both sulfate and nitric acid, ammonia prefers to react with sulfate to form ammonium sulfate particles than to react with nitric acid to form ammonium nitrate particles. When SOx emissions are reduced by 75%, ammonia is available to react to form ammonium nitrate particles. The increase in the RADM/RPM annual nitrate particulate concentrations when SOx emissions are reduced 75% across the board are shown in Figure 12. Note that since Figure 12 shows the difference in base case minus the 75% reduction in SOx concentrations, the negative numbers indicate an increase in nitrate concentrations. The northeast quadrant of the modeling domain shows an on average increase of 0.2 to 0.4 ug/m3 increase in nitrate particulate concentrations. The increase over the western and southeastern portion of the modeling domain is generally less than 0.2 ug/m3. Nitrate particulate concentrations for the base case minus the 75% reduction in SOx and NOx case are shown in Figure 13. Positive numbers reflect a decrease in nitrate particulate concentrations and negative numbers reflect an increase in nitrate particulate concentrations. Figure 13 shows that the addition of a 75% reduction in NOx emissions results in a decrease in nitrate particulate concentrations from the base case for the majority of the modeling domain. Significant reductions in nitrate particulate concentrations, 0.4 to 0.8 ug/m3, were obtained in portions of the western modeling domain where base case nitrate concentrations were highest. The extreme northeast still shows a small, less than 0.2 ug/m3, increase in nitrate particulate concentrations due to the

75% reduction in SOx. In Figure 14, each data point represents the concentration in ug/m3 for each of the 1330 grid squares in the modeling domain. The vertical axis corresponds to the nitrate particulate concentrations for the 75% reduction in SOx emissions scenario and the horizontal axis corresponds to the nitrate particulate concentrations for the 75% reduction in NOx and SOx scenario. The left line is a regression line through the center of all 1330 grid points. The line further to the right is a one-to-one correspondence line. Examining the regression line in Figure 14 shows that reducing NOx emissions in addition to SOx emissions by 75 percent leads to on average a large, 50 percent, reduction in nitrate particulate concentrations. Figure 15 shows spatially the added percent reduction in nitrate concentrations of a 75% reduction in NOx over a 75% reduction in SOx. Although as supported by Figure 14 the average added percent reduction by reducing NOx by 75% is approximately 50%, there is some spatial variation in that percent reduction. In general, higher percent reductions are shown in the southern portion of the modeling domain.

Conclusions

- Base case 1990 sulfate particulate concentrations are much higher than nitrate particulate concentrations in the eastern United States.

- Reducing SOx emissions by 75 % in the eastern United States reduced sulfate plus nitrate particulate concentrations by greater than 55 percent in the majority of the modeled area.

- Reducing NOx in addition to SOx emissions by 75 % in the eastern United States further reduced sulfate plus nitrate particulate concentrations by an average of 12 percent. The reduction is 5 to 10 percent in large areas of the eastern portion of the modeled domain and 20 to 30 percent in a large area of the western portion of the modeling domain.

■ Reducing SOx emissions by 75 % in the eastern United States reduces sulfate particulate concentrations by greater than 65 % over the majority of the modeled area. This 75% reduction in SOx emissions does lead to a small, generally less than 0.4 ug/m3, increase in nitrate particulate concentrations.

■ Reducing NOx in addition to SOx emissions by 75% in the eastern United States reduces nitrate particulate concentrations by an average of 50 percent.

■ Decreasing NOx emissions in the eastern United States does not lead to an increase in annual sulfate particulate concentrations.

Figure 1
RADM / RPM Modeling Domain

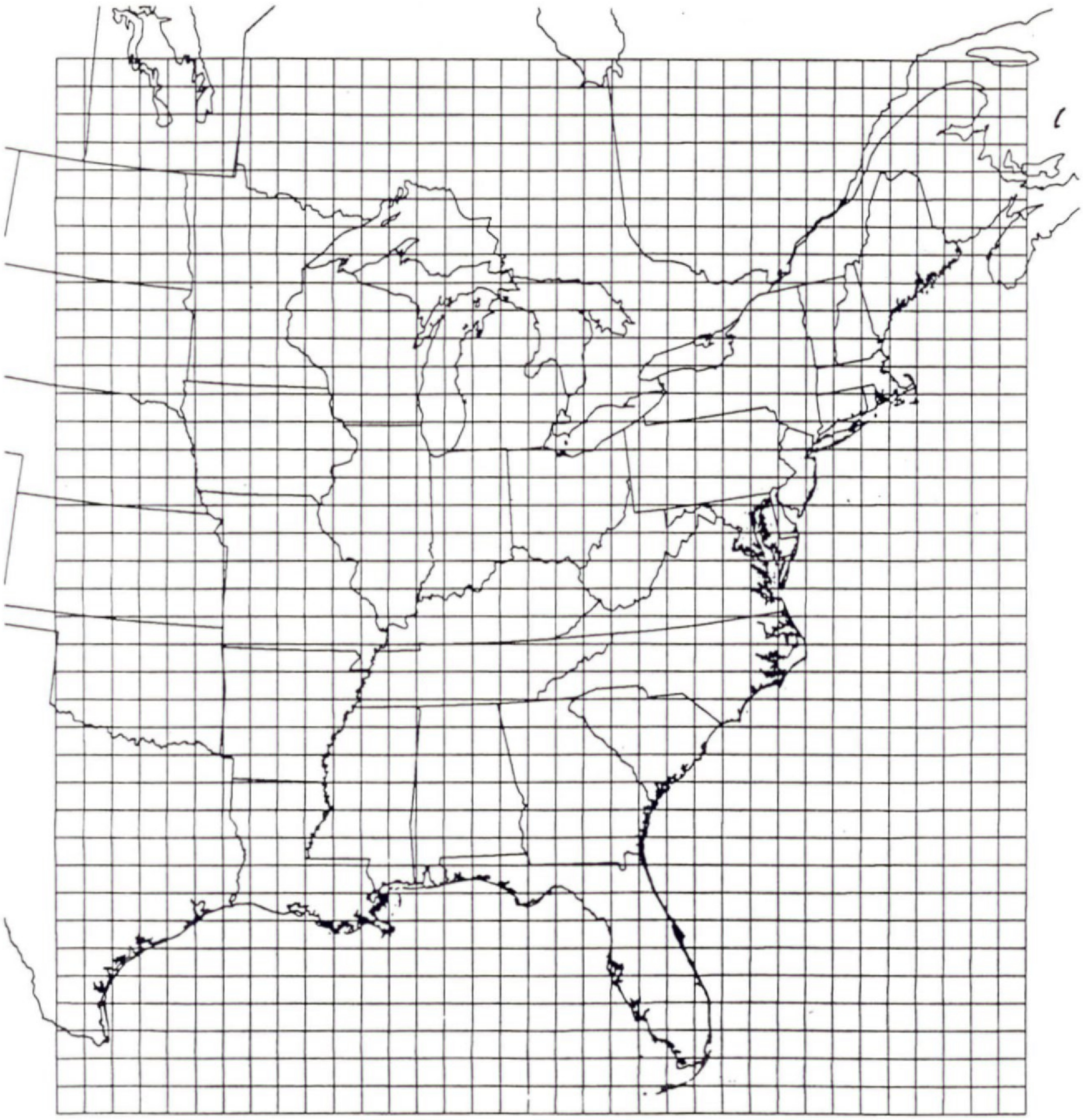


Figure 2

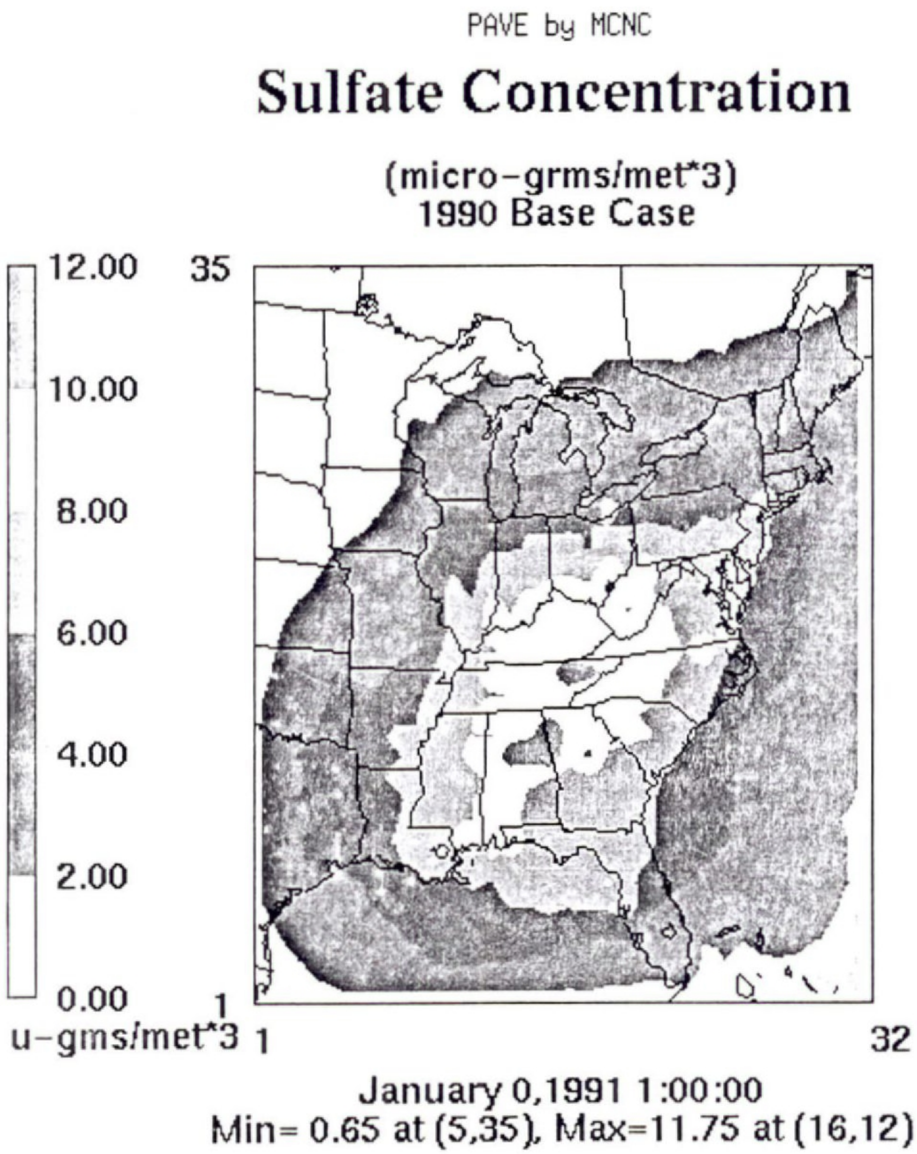
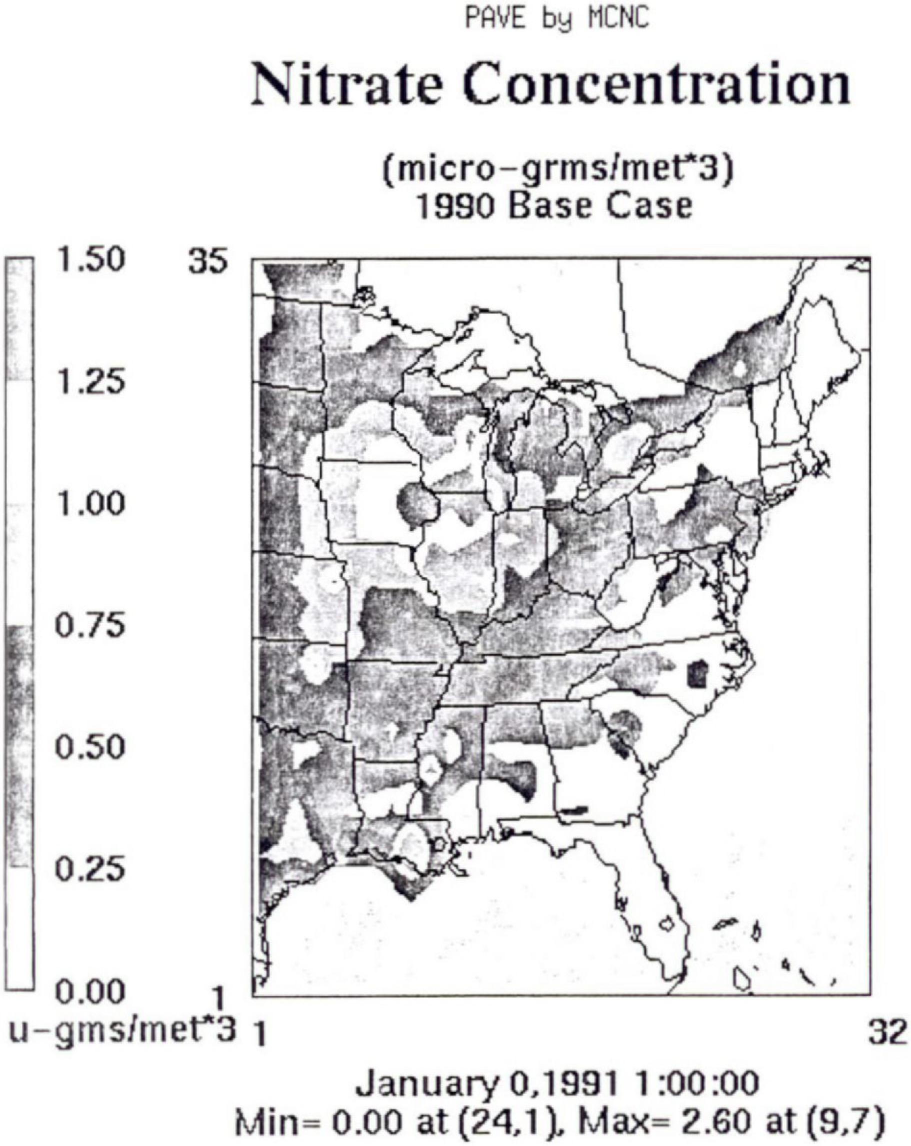

PAVE by MCNC
Sulfate Concentration
(micro-grms/met*3)
1990 Base Case
12.00
10.00
8.00
6.00
4.00
2.00
0.00
u-gms/met*3
35
1
1
32
January 0,1991 1:00:00
Min= 0.65 at (5,35), Max=11.75 at (16,12)

Figure 3



PAVE by MCNC
Nitrate Concentration
(micro-grms/met*3)
1990 Base Case
1.50
1.25
1.00
0.75
0.50
0.25
0.00
u-gms/met*3
35
1
1
32
January 0,1991 1:00:00
Min= 0.00 at (24,1), Max= 2.60 at (9,7)

Figure 4

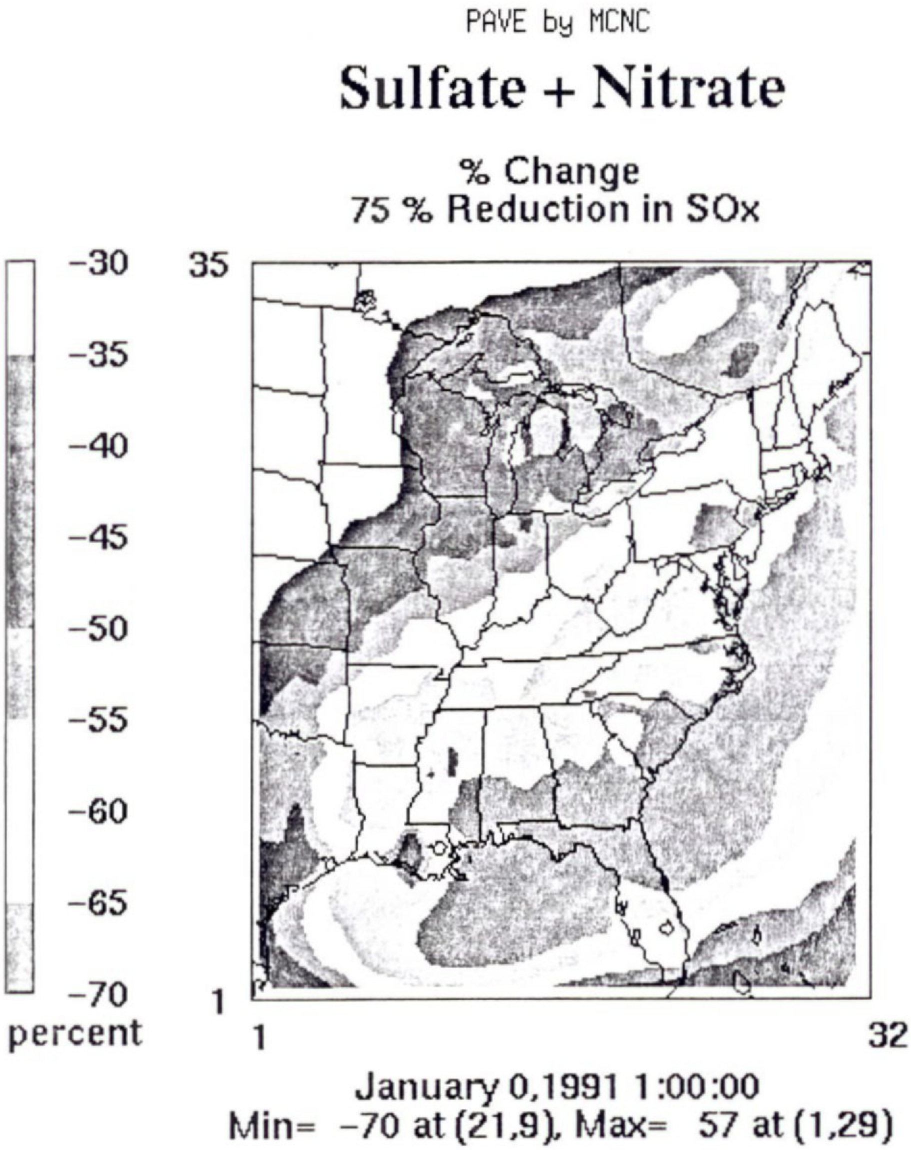
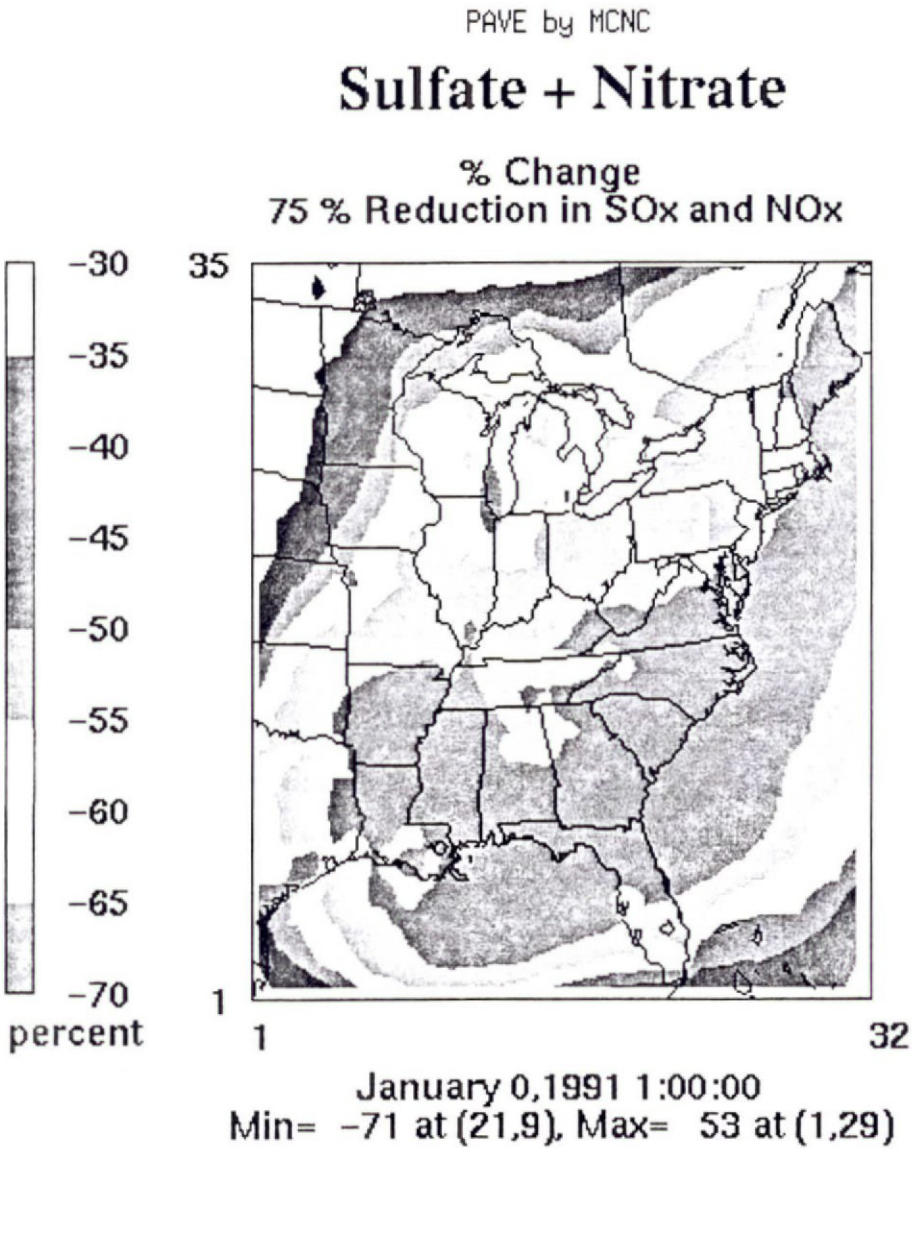
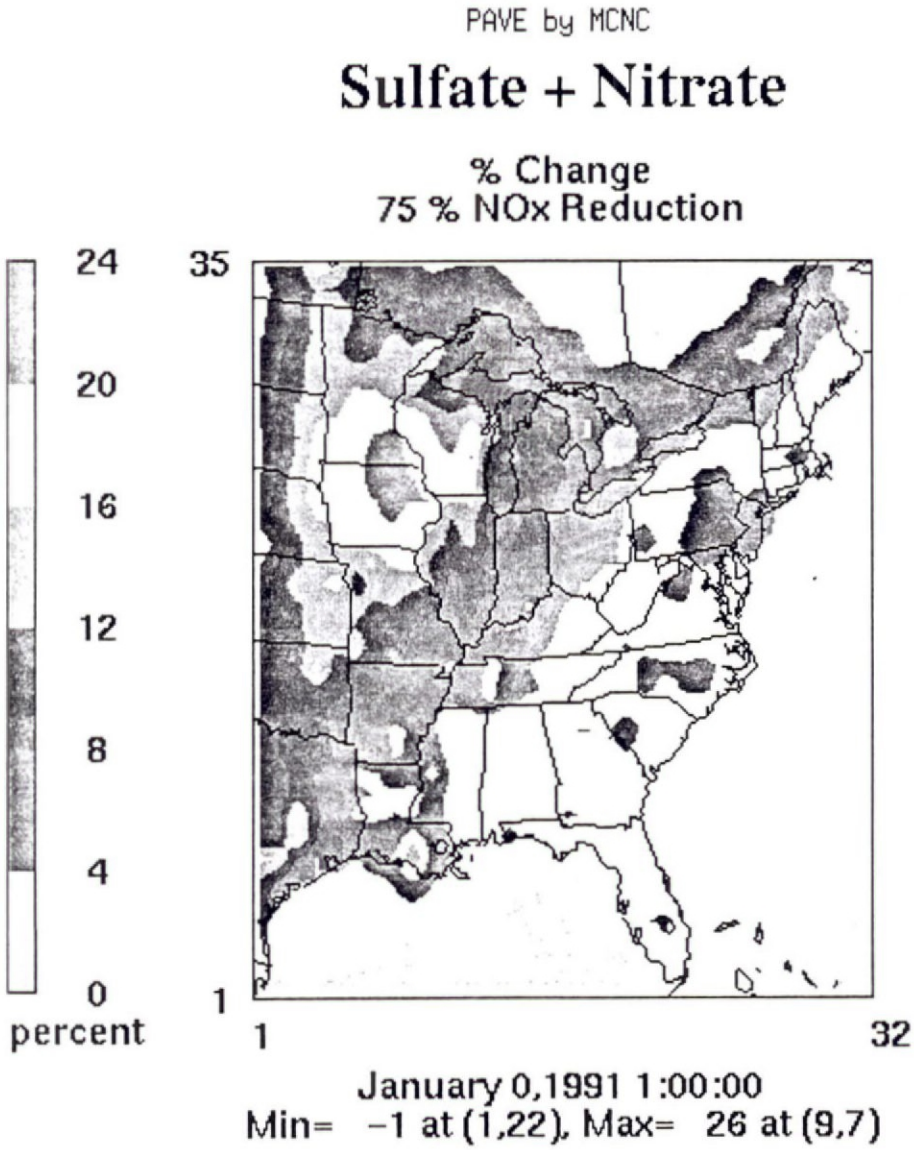

PAVE by MCNC
Sulfate + Nitrate
% Change
75 % Reduction in SOx
-30
-35
-40
-45
-50
-55
-60
-65
-70
percent
35
1
1
32
January 0,1991 1:00:00
Min= -70 at (21,9), Max= 57 at (1,29)

Figure 5



PAVE by MCNC
Sulfate + Nitrate
% Change
75 % Reduction in SOx and NOx
-30
-35
-40
-45
-50
-55
-60
-65
-70
percent
35
1
1
32
January 0,1991 1:00:00
Min= -71 at (21,9), Max= 53 at (1,29)

Figure 6



PAVE by MCNC
Sulfate + Nitrate
% Change
75 % NOx Reduction
24
20
16
12
8
4
0
percent
35
1
1
32
January 0,1991 1:00:00
Min= -1 at (1,22), Max= 26 at (9,7)

Figure 7
Sulfate + Nitrate Concentrations (ug/m3)

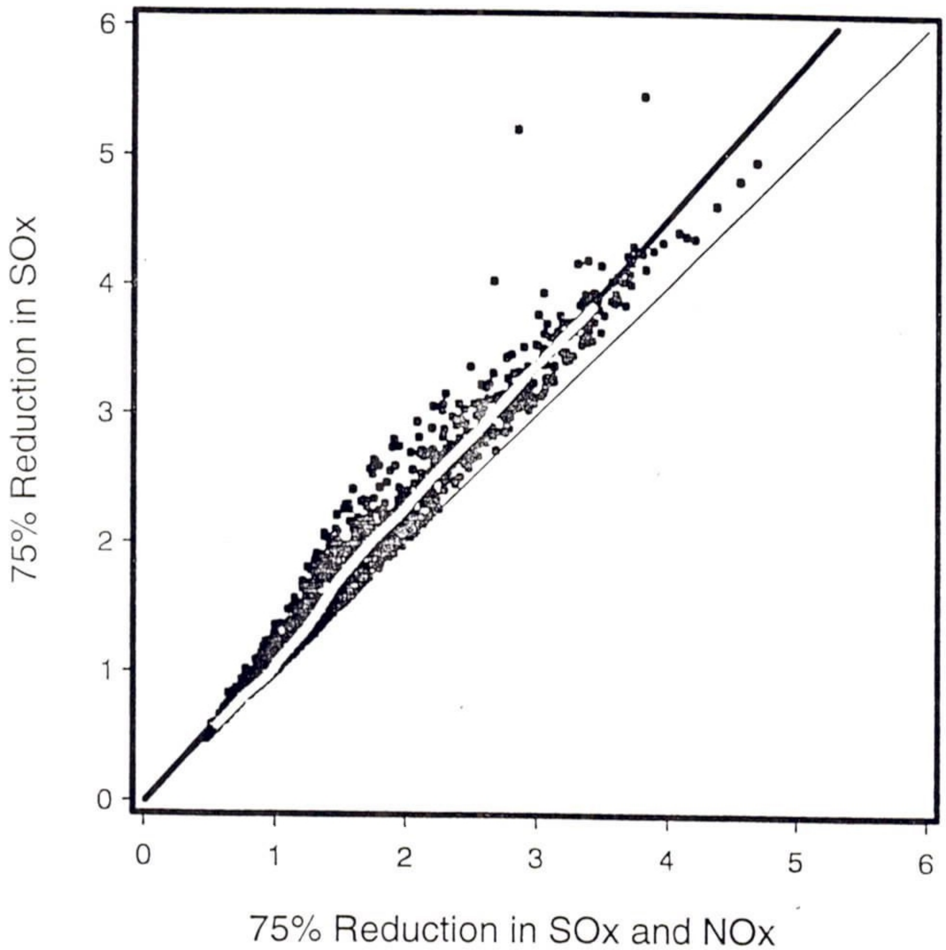
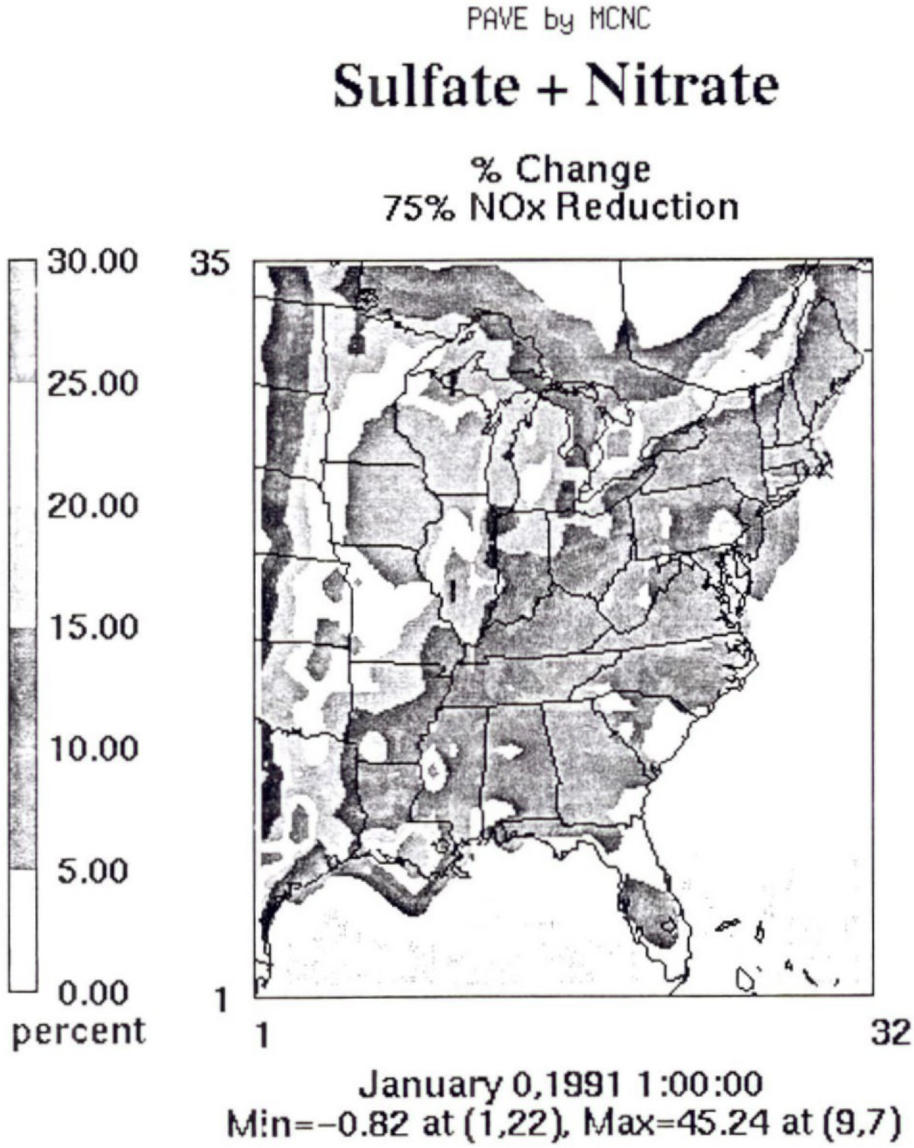
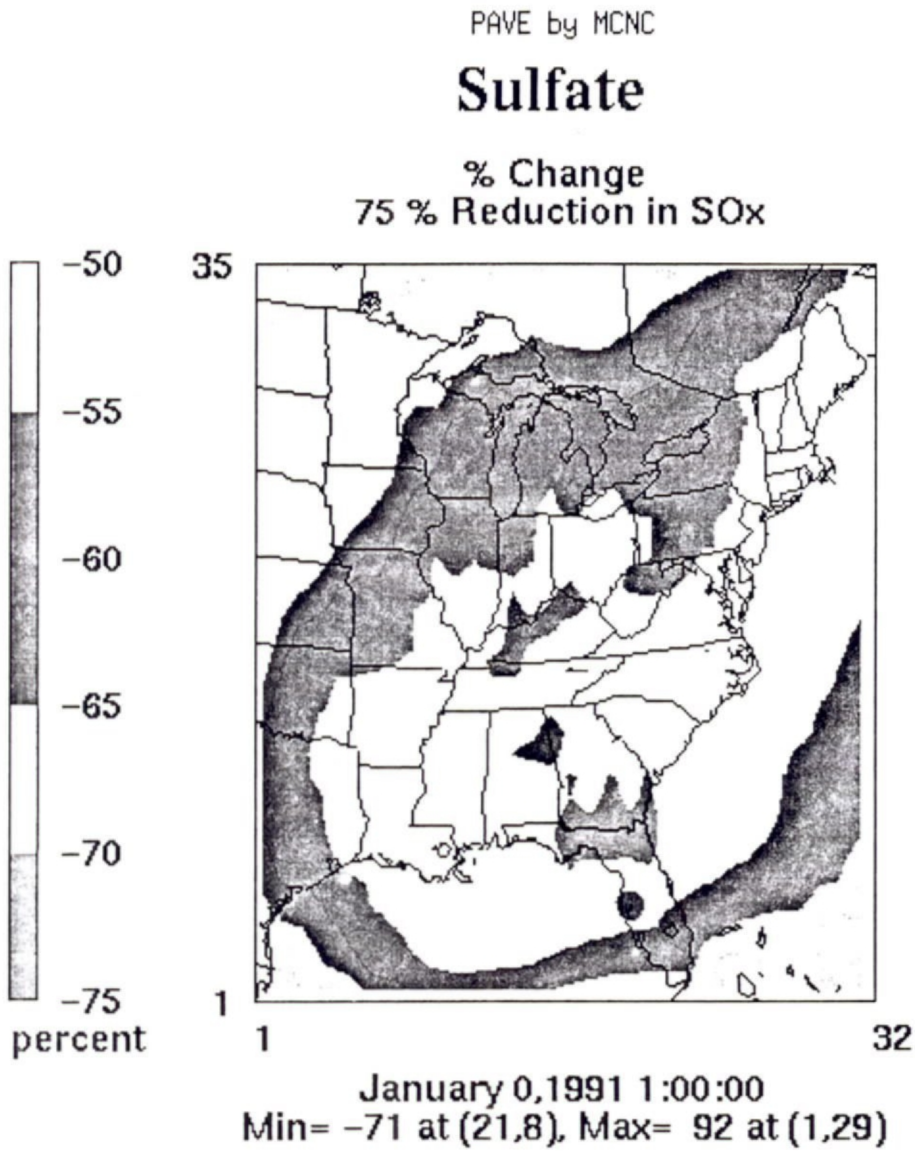


Figure 8



PAVE by MCNC
Sulfate + Nitrate
% Change
75% NOx Reduction
30.00
25.00
20.00
15.00
10.00
5.00
0.00
percent
35
1
1
32
January 0,1991 1:00:00
Min=-0.82 at (1,22), Max=45.24 at (9,7)

Figure 9



PAVE by MCNC
Sulfate
% Change
75 % Reduction in SOx
-50
-55
-60
-65
-70
-75
percent
35
1
1
32
January 0,1991 1:00:00
Min= -71 at (21,8), Max= 92 at (1,29)

Figure 10

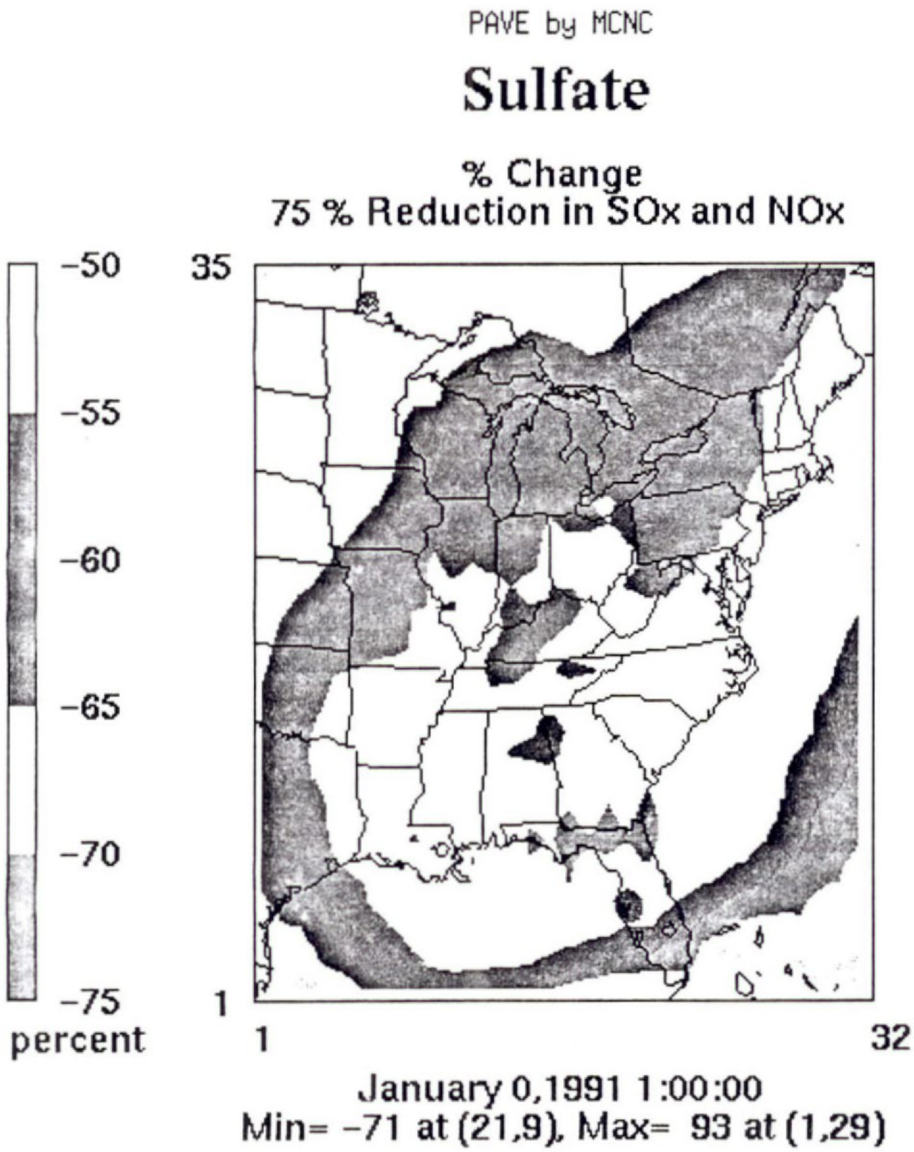

PAVE by MCNC
Sulfate
% Change
75 % Reduction in SOx and NOx
-50
-55
-60
-65
-70
-75
percent
35
1
1
32
January 0,1991 1:00:00
Min= -71 at (21,9), Max= 93 at (1,29)

Figure 11
Sulfate Concentrations (ug/m3)

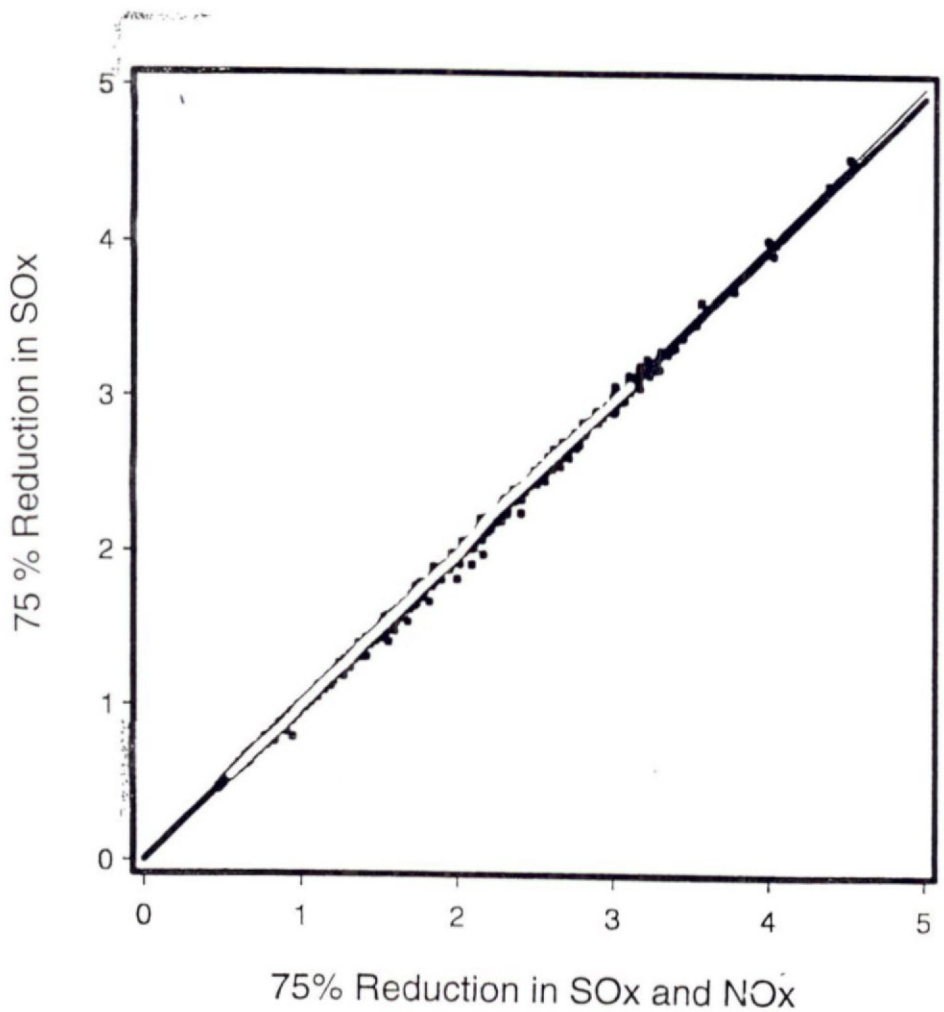


Figure 12

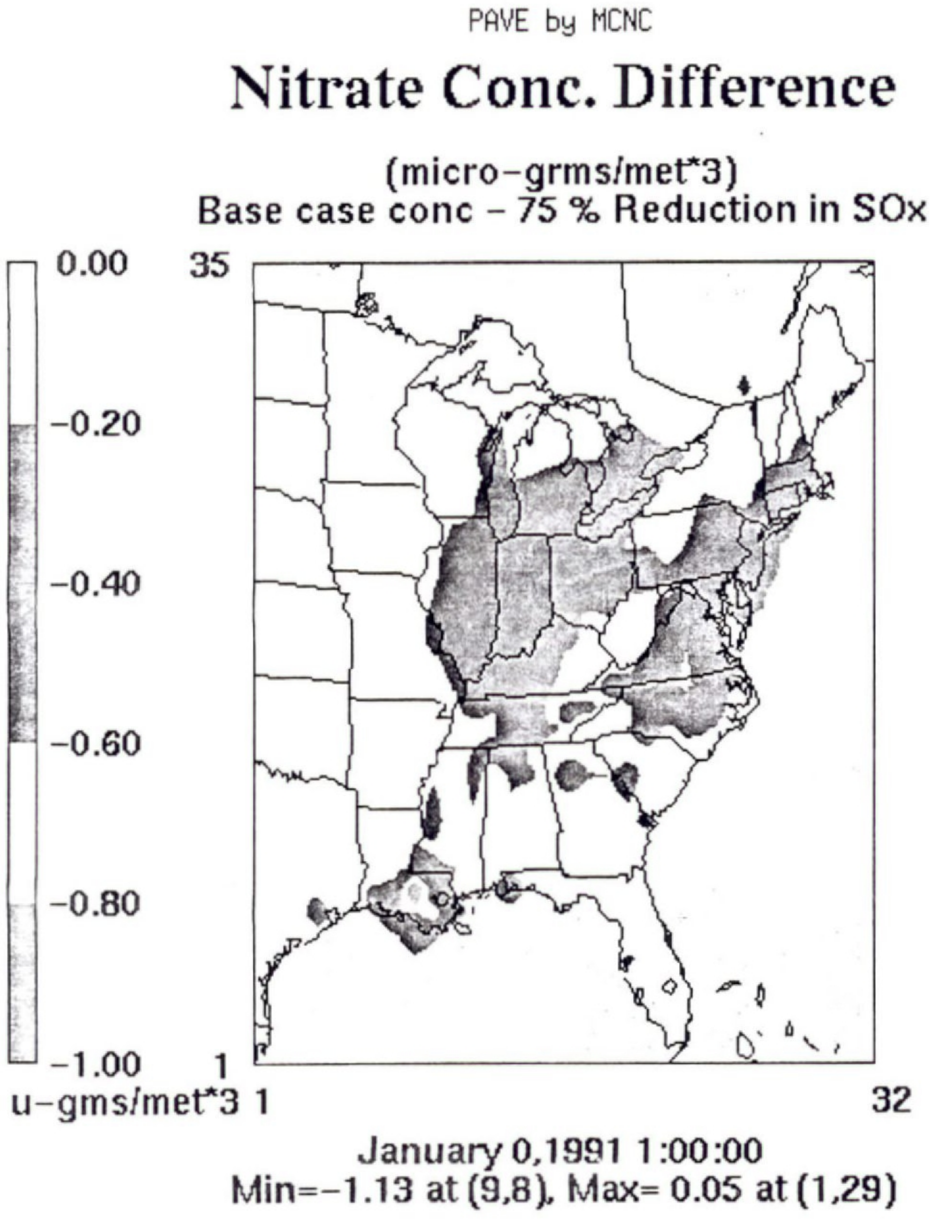

PAVE by MCNC
Nitrate Conc. Difference
(micro-grms/met*3)
Base case conc – 75 % Reduction in SOx
0.00
-0.20
-0.40
-0.60
-0.80
-1.00
u-gms/met*3
35
1
1
32
January 0,1991 1:00:00
Min=-1.13 at (9,8), Max= 0.05 at (1,29)

Figure 13

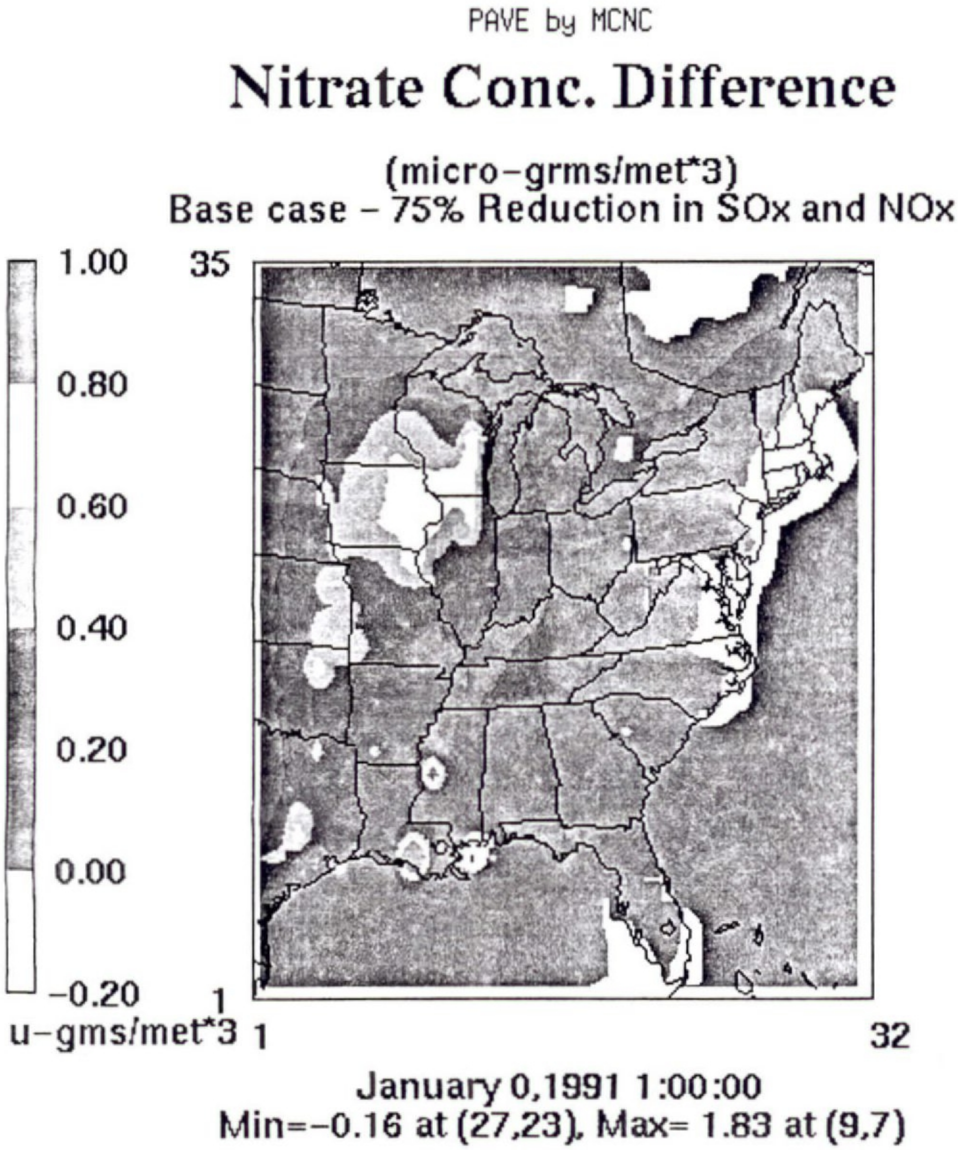

PAVE by MCNC
Nitrate Conc. Difference
(micro-grms/met*3)
Base case - 75% Reduction in SOx and NOx
1.00
0.80
0.60
0.40
0.20
0.00
-0.20
u-gms/met*3
35
1
1
32
January 0,1991 1:00:00
Min=-0.16 at (27,23), Max= 1.83 at (9,7)

Figure 14
Nitrate Concentrations (ug/m3)

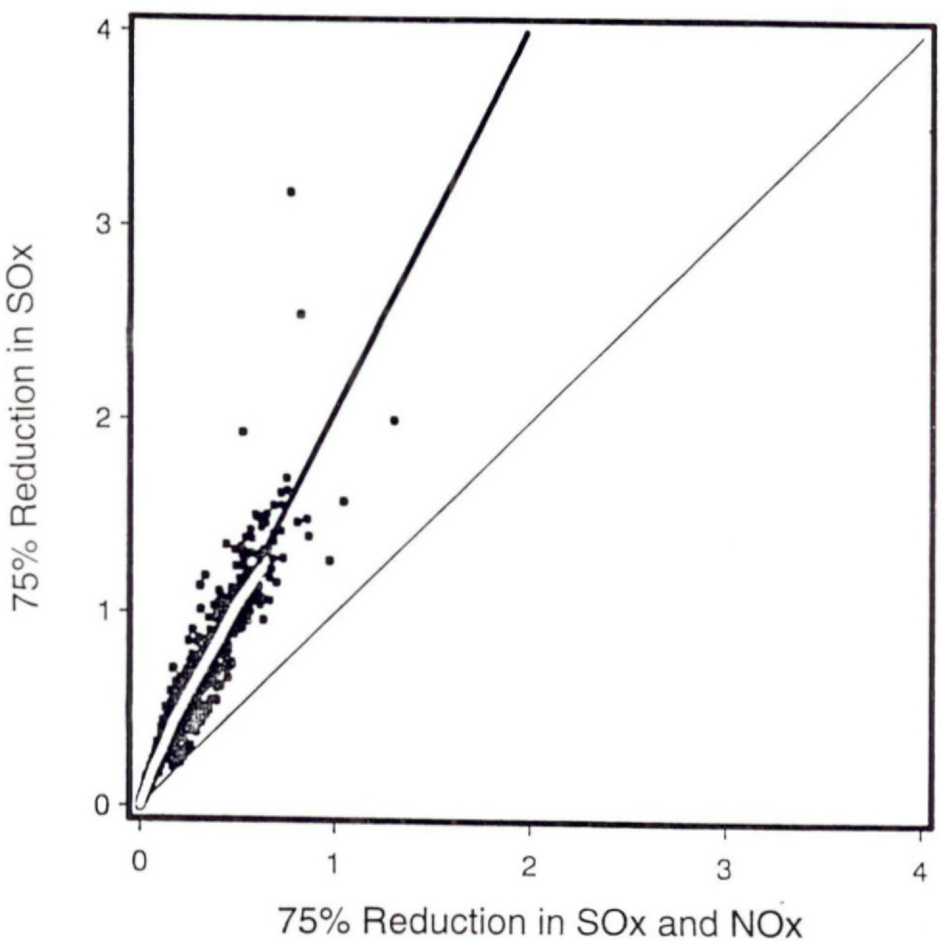
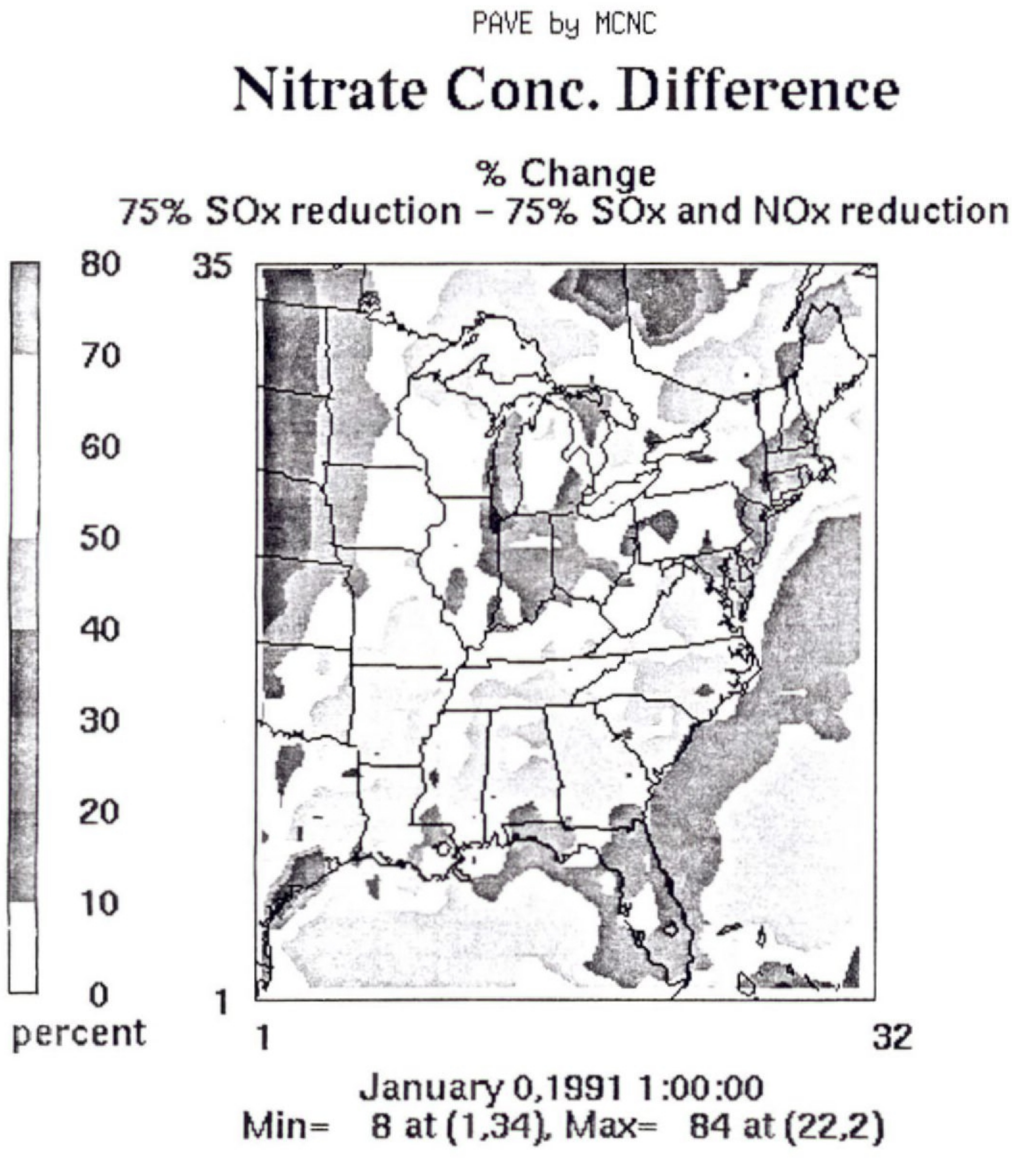


Figure 15


PAVE by MCNC
Nitrate Conc. Difference
% Change
75% SOx reduction - 75% SOx and NOx reduction
80
70
60
50
40
30
20
10
0
percent
35
1
1
32
January 0,1991 1:00:00
Min= 8 at (1,34), Max= 84 at (22,2)