Report

Causes of Haze Assessment for US Fish and Wildlife Service Class I Areas in the Central and Eastern United States

Task 2. Aerosol Summary

Sub-Task. Summary of Clean Air Status and Trends Network (CASTNET) atmospheric and dry deposition data for the period 2000-2004

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August 2006

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A. Site locations and characteristics

None of the twelve (12) FWS sites of the IMPROVE network included in this study is collocated with a CASTNET site. For this reason, a set of geographical criteria are developed to associate each FWS site with a CASTNET site. Initially, only CASTNET sites within 100 km away from the FWS site and difference in elevation of no more than 300 meters are considered. As a result, a CASTNET site is obtained for nine FWS sites with distance varied from 25 up to 88 km. For three sites (Breton LA, Wichita Mountain OK and Wolf Island GA) the nearest CASTNET site is chosen (Table 1). For three FWS sites (Breton LA, Okefenokke GA and St. Marks FL), CASTNET site Sumatra (in Florida) is chosen based on the geographical criteria. The background in maps is land cover data obtained from the AVHRR Global land Cover database covering all of North America.

Figure 1 Map showing the location of Fish and Wildlife Service sites (yellow circles) and CASTNET sites (white circles)

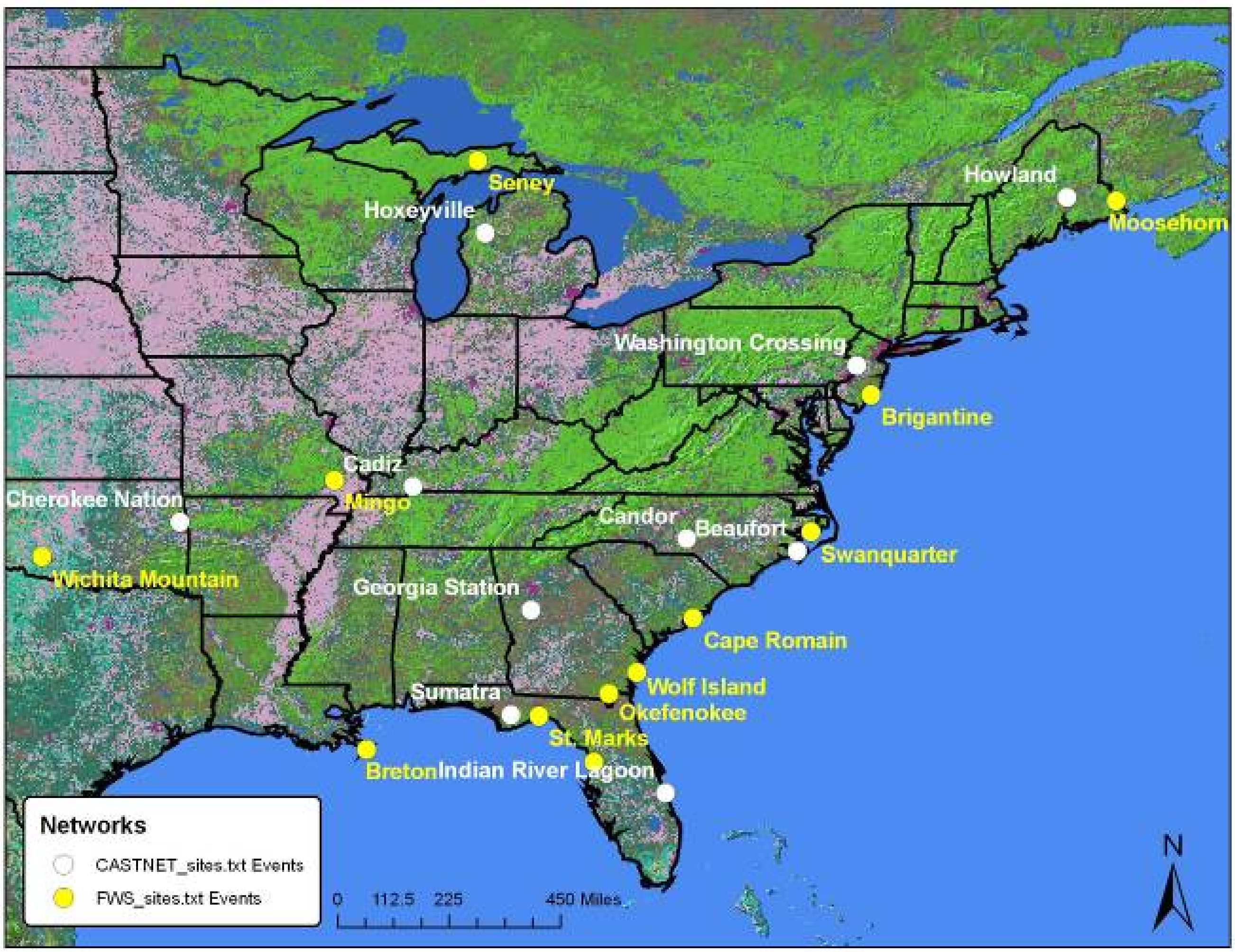


Table 1 FWS and CASTNET sites

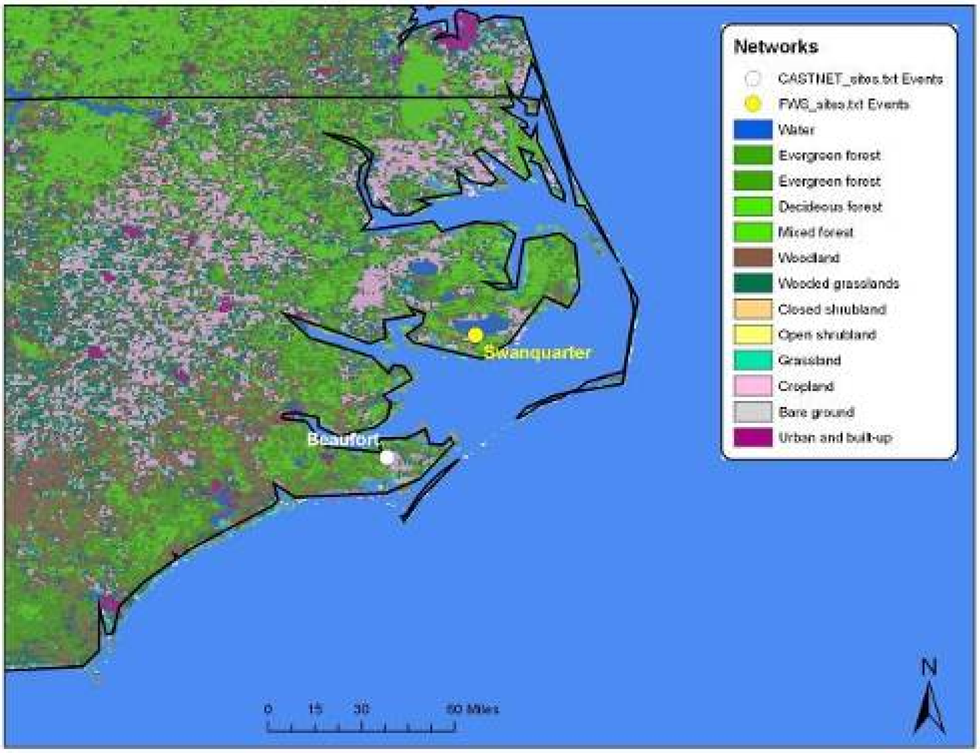
Fish and Wildlife Service site	**Coordinates, Elevation**	**CASTNET site**	**CoordinatesElevation**	**Distances Km / m**
Brigantine NJ	N39.46 W74.45 5	Washington Crossing NJ	N40.31 W74.87 61	31 / 56
Chassahowitzka FL	N28.74 W82.55 4.3	Indian River Lagoon FL	N27.84 W80.45 2	71 / 2
Breton LA	N29.12 W89.20 11	Sumatra FL	N30.11 W84.94 14	131 / 3
Cape Romain SC	N32.94 W79.65 4.7	Candor NC	N35.26 W79.83 198	80 / 193
St. Marks FL	N30.09 W84.16 7.7	Sumatra Fl	N30.11 W84.94 14	25 / 6
Mingo KS	N36.97 W90.14 111.3	Cadiz KY	N36.78 W87.85 189	64 / 78
Moosehorn ME	N45.13 W67.26 77.7	Howland ME	N45.21 W68.70 72.7	35 / 9
Okefenokee GA	N30.74 W82.13 4.8	Sumatra FL	N30.11 W84.94 14	88 / 34
Seney MI	N46.29 W85.95 214.5	Hoxeyville MI	N44.18 W85.74 305	73 / 91
Swanquarter NC	N35.45 W76.20 2	Beaufort NC	N34.88 W76.62 2	23 / 6
Wichita Mt OK	N34.73 98.71 8	Cherokee Nation OK	N35.75 W94.67 299	119 / 294
Wolf Island GA	N31.35 W81.31 2	Georgia Station GA	N33.18 W84.40 270	110 / 268

B. Ambient concentrations and deposition

B.1 Beaufort, North Carolina

Beaufort, North Carolina is used as a surrogate CASTNET site for FWS Swanquarter site in North Carolina. Figure 2 shows the locations of both sites at the end of Pamlico River.

Figure 2 Location of CASTNET Beaufort and FWS Swanquarter sites.



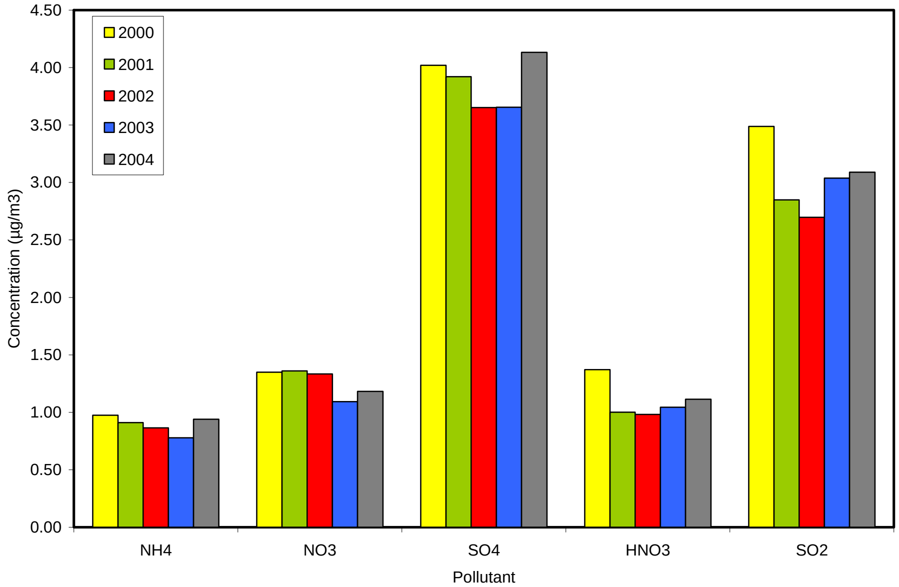
B.1.1 Ambient air

Ambient air concentration levels of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Beaufort, NC during the 2000-2004 period are presented in Table 2.

Table 2 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Beaufort, NC for 2000-2004 periods

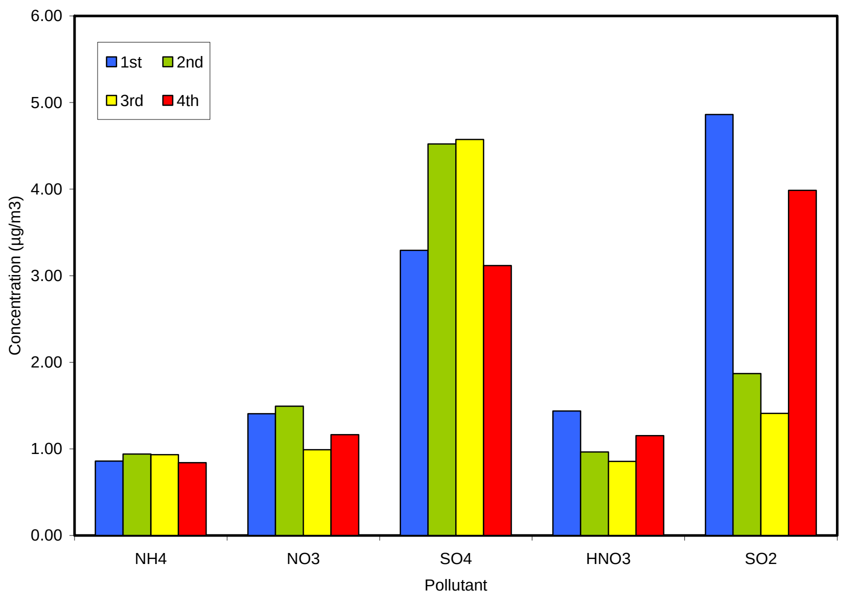
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	0.9	0.2	2.6	0.4
Nitrate (NO_3^-)	1.3	0.1	2.9	0.5
Sulfate (SO_4^{2-})	3.9	0.9	9.9	1.6
Gas Phase				
Sulfur dioxide (SO_2)	3.0	0.4	14.1	2.4
Nitric Acid (HNO_3)	1.1	0.1	4.2	0.7

Figure 3 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 3 concentrations of particulate components (ammonium, nitrate and sulfate) decreased from 2000 to 2003, while a slight increase is observed for 2004. A similar profile but with upward trends in atmospheric concentrations for sulfur dioxide and nitric acid starting in 2003 is plotted.

Figure 4 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for ammonium are measured. The differences are more pronounced for sulfate, while an opposite profile is drawn for sulfur dioxide and nitric acid, with higher concentration being measured during winter.

Figure 5 Scatter plot of NH4 nano-equivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

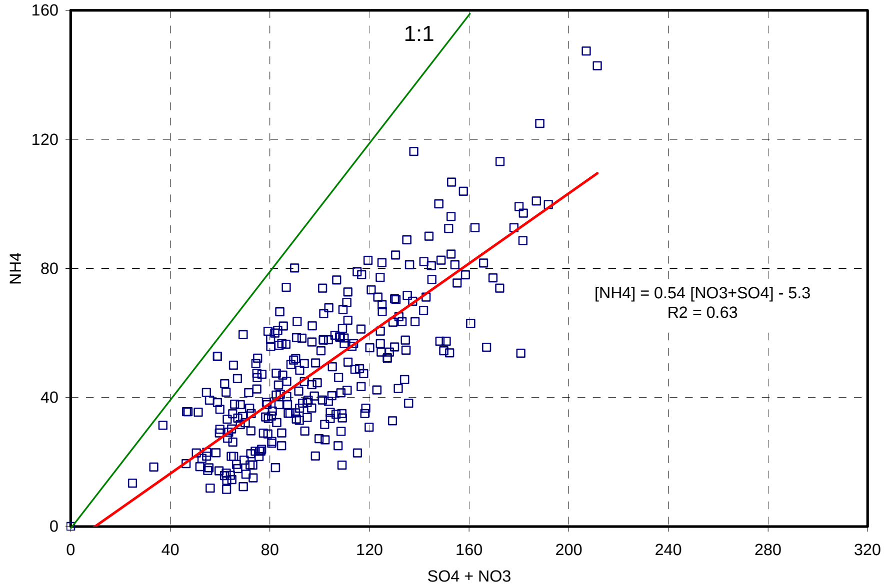


Figure 5 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate (NH_4NO_3) and ammonium sulfate $(NH_4)_2SO_4$). The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 50% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, H_2SO_4 and HNO_3 and minor amounts of NH_4NO_3 and $(NH_4)_2SO_4$, if any.

Table 3 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be more significant during the 2nd and 3rd quarter, while more than 50% of sulfate and nitrate is present in free form (as acids) in the 1st quarter. This may be associated with the seasonal variability of gaseous NH_3, SO_2 and NO_x emissions.

Table 3 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.48	0.61	0.52	0.56	0.53
Intercept	3.7	-12.3	-2.5	-9.2	-3.5
R	0.75	0.86	0.7	0.75	0.82
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.43	0.61	0.68	0.59	
Intercept	8.5	-20.3	-23.3	-2.9	
R	0.66	0.83	0.90	0.78	

B.1.2 Dry Deposition

Deposition rates and total dry deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Beaufort, NC during the 2000-2004 periods are presented in**Error! Not a valid bookmark self-reference.**.

Table 4 Descriptive statistics (g/ha) of dry deposition rate of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Beaufort, NC for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	7	2	21	4	1.4
Nitrate (NO_3^-)	11	1	37	6	0.6
Sulfate (SO_4^{2-})	31	8	82	15	2.8
Gas Phase					
Sulfur dioxide (SO_2)	76	10	366	54	9.6
Nitric Acid (HNO_3)	95	9	363	58	5.3
Total Nitrogen					7.3
Total Sulfur					12.3

As shown in Figure 6 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2), annual dry deposition of particulate ammonium, nitrate and sulphate followed a downward trend from 2000 to 2003 and a slight increase during 2004, which is consistent with their atmospheric concentrations. On the other hand, while SO2 and HNO3 dry deposition was significantly reduced in 2001, it increased progressively from 2002 to 2004.

Figure 6 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2)

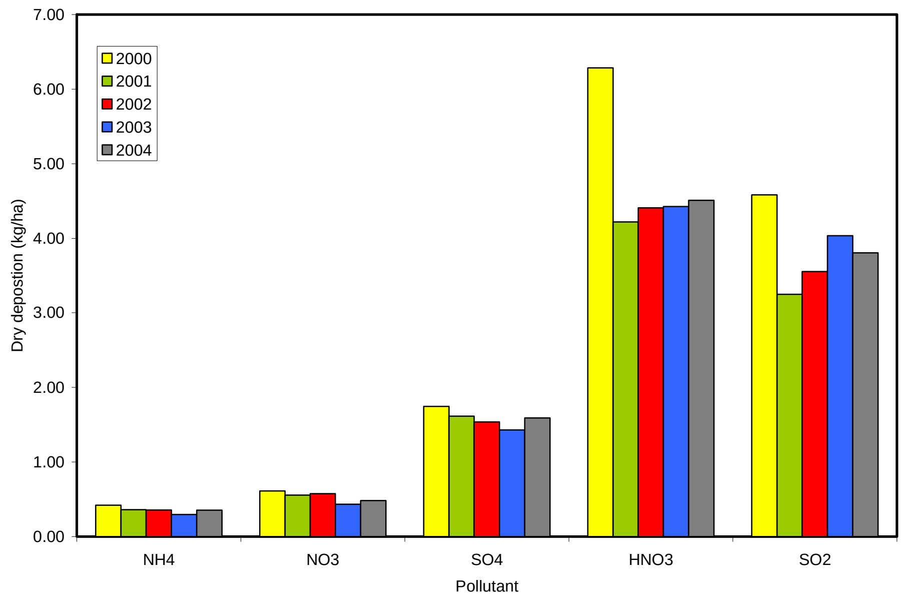
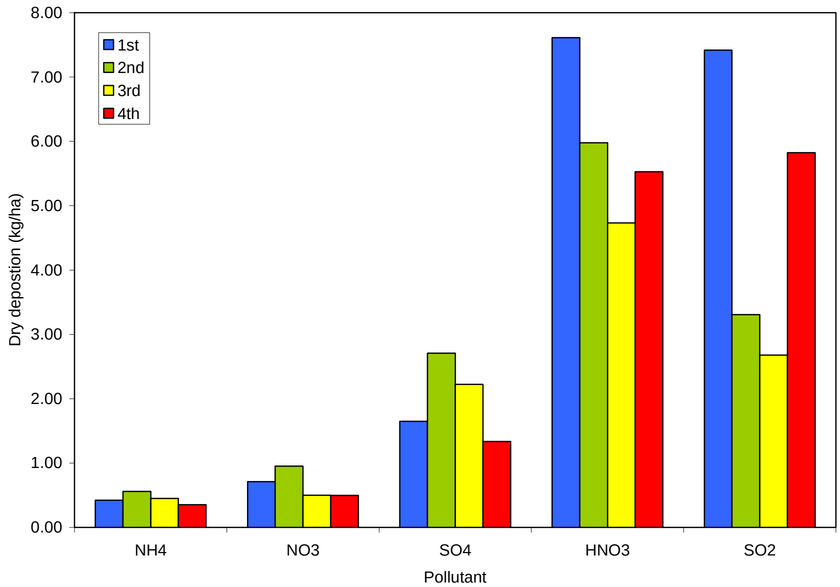
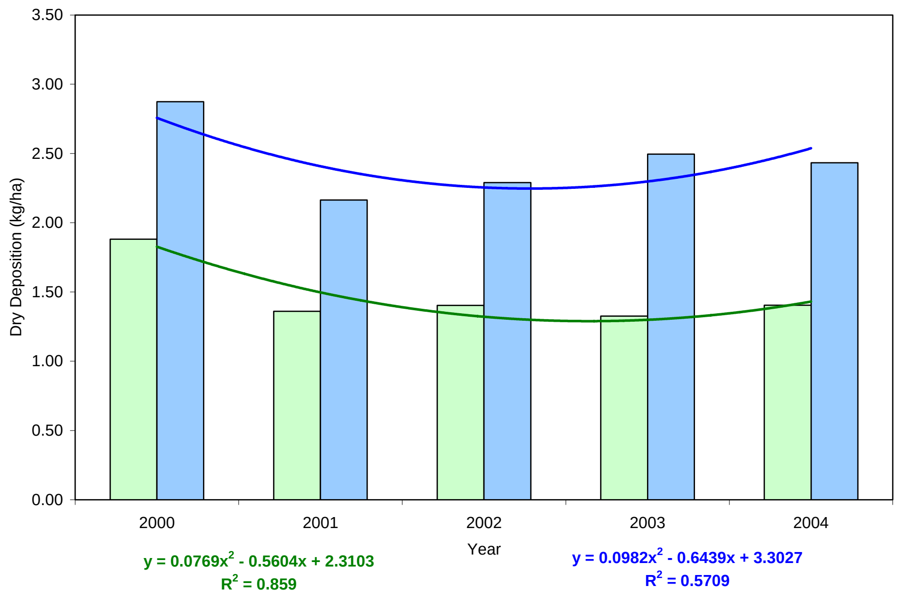


Figure 7 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated components during the warm period (2^{rd} -3^{rd} quarter) and significant input of SO_2 and HNO_3 during the cold period (1^{st} and 4^{th} quarters)

Figure 8 Total nitrogen and sulfur dry deposition at Beaufort during 2000-2004

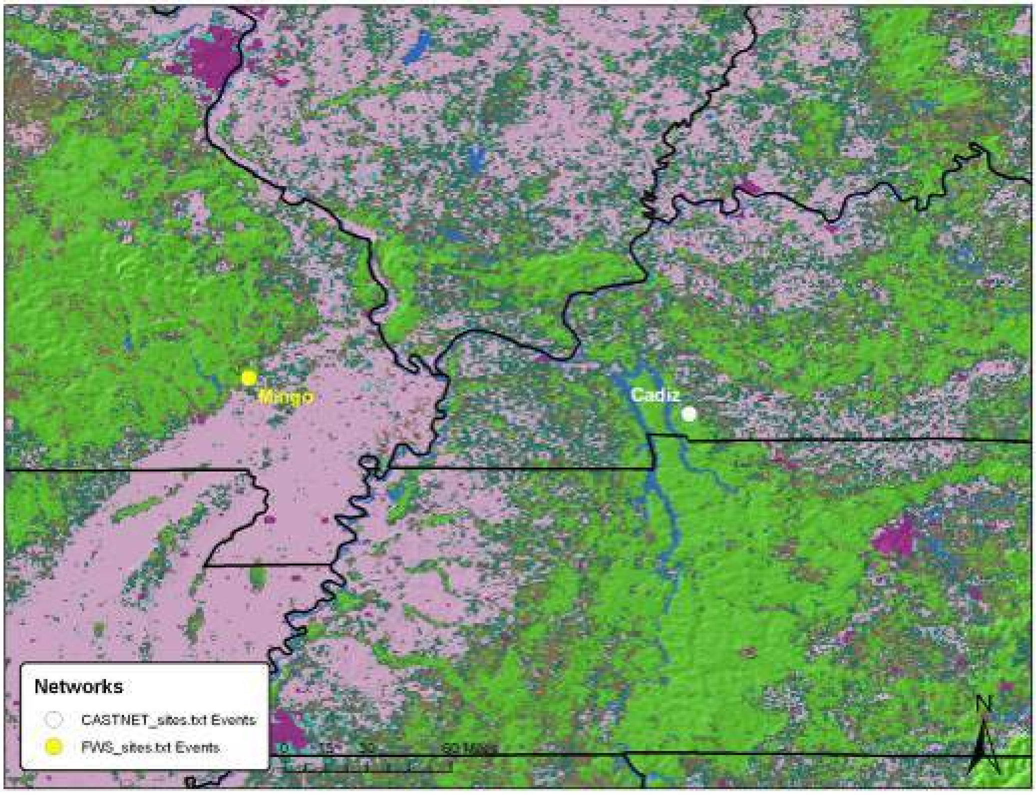


The total amount of nitrogen and sulfur deposited in Beaufort NC, ranged approximately from 1.3 kg/ha to 1.9 kg/ha and from 2.2 kg/ha up to 2.9 kg/ha, respectively. A clear inter-annual pattern is observed for both elements with comparables amounts of dry deposition in 2000 and 2004, providing evidence of an increasing trend during the last two years.

B.2 Cadiz, Kentucky

Cadiz, Kentucky is used as a surrogate CASTNET site for FWS Mingo site in Missouri. Figure 9 shows the locations of both sites.

Figure 9 Location of CASTNET Cadiz site with respect to FWS Mingo site.



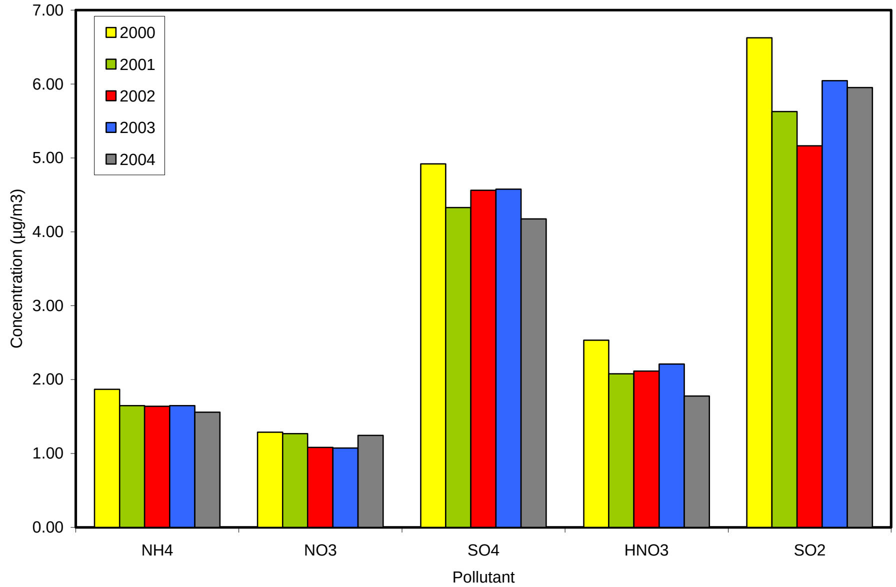
B.2.1 Ambient air

Ambient air concentration levels of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Cadiz, KY during the 2000-2004 period, are presented in Table 5.

Table 5 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Cadiz KY for 2000-2004 periods

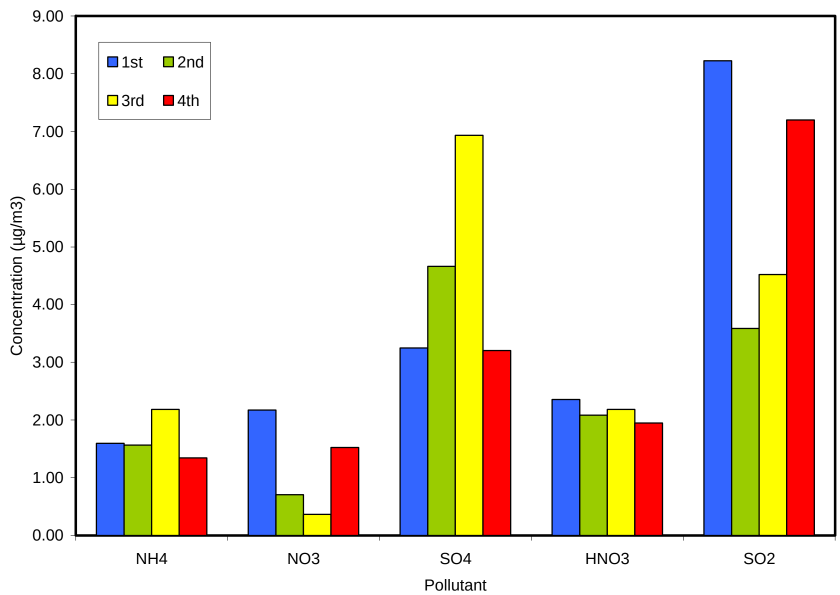
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.7	0.4	4.5	0.7
Nitrate (NO_3^-)	1.2	0.1	6.5	1.2
Sulfate (SO_4^{2-})	4.5	1.2	13.1	2.4
Gas Phase				
Sulfur dioxide (SO_2)	5.9	0.7	24.7	3.5
Nitric Acid (HNO_3)	2.1	0.6	5.1	0.8

Figure 10 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 10 concentrations of particulate components (ammonium, nitrate and sulfate) decreased from 2000 to 2003, while a slight increase is observed for 2004. A similar profile but with upward trends in atmospheric concentrations for sulfur dioxide is plotted.

Figure 11 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate are measured. An opposite profile is drawn for nitrate, sulfur dioxide and nitric acid, with higher concentration being measured during winter.

Figure 12 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

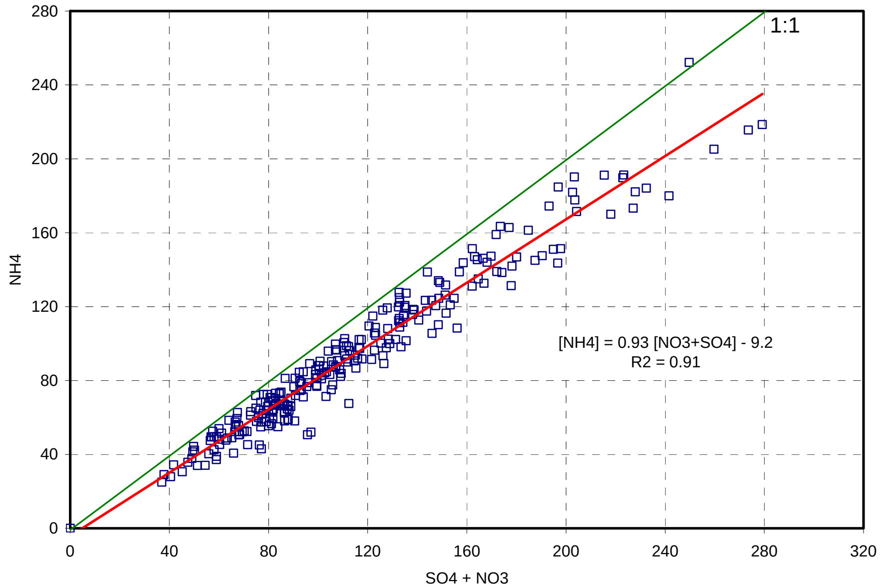


Figure 12 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 93% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 and $(NH_4)_2SO_4$, and minor amounts of H_2SO_4 and HNO_3 if any.

Table 6 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be significant throughout the year, while minor amounts of sulfate and nitrate is present in free form (as acids).

Table 6 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.84	0.83	0.82	0.91	0.87
Intercept	-0.2	0.0	-0.6	-11.8	-6.3
R	0.98	0.97	0.98	0.98	0.98
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	1.02	0.87	0.80	0.92	
Intercept	-16.3	-7.7	1.0	-9.7	
R	0.99	0.95	0.99	0.98	

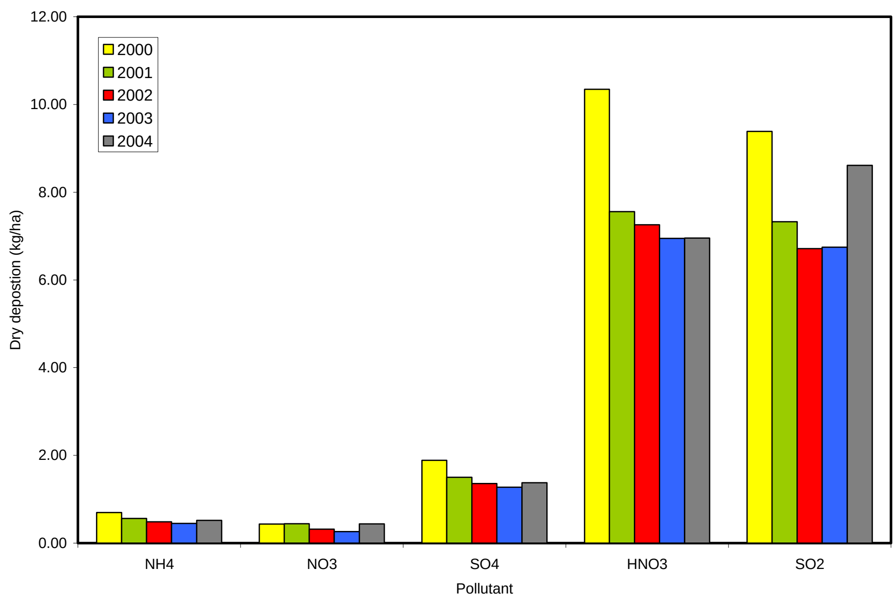
B.2.2 Dry Deposition

Concentrations and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Cadiz, KY during the 2000-2004 period, are presented in **Error! Not a valid bookmark self-reference.**.

Table 7 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Cadiz, KY for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

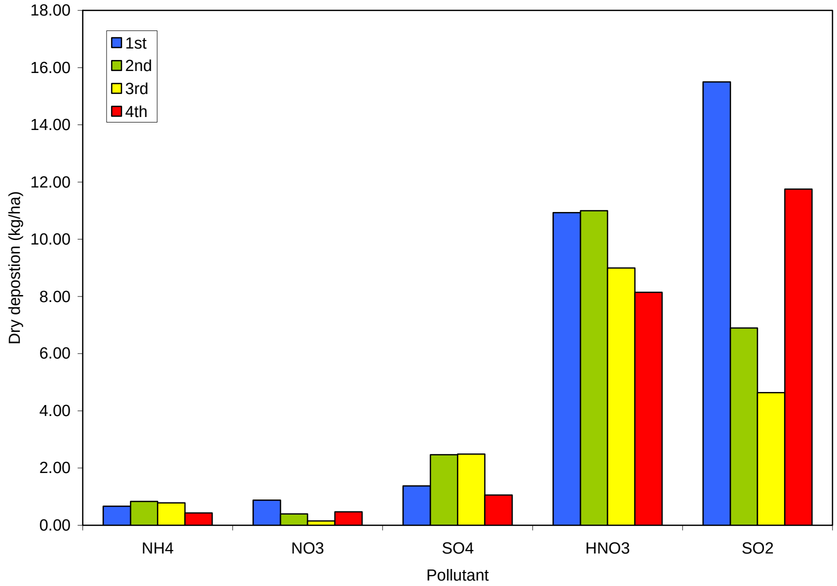
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	12	3	32	6	2.1
Nitrate (NO_3^-)	8	0	46	8	0.4
Sulfate (SO_4^{2-})	32	5	98	19	19.4
Gas Phase					
Sulfur dioxide (SO_2)	167	15	736	106	2.5
Nitric Acid (HNO_3)	168	25	371	60	8.7
Total Nitrogen					11.2
Total Sulfur					21.9

Figure 13 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



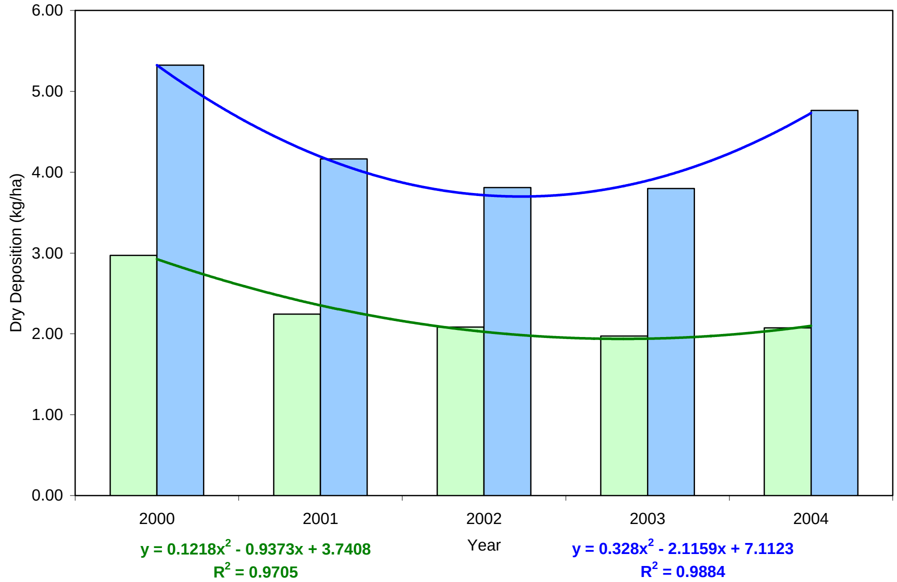
As shown in Figure 13, annual dry deposition of particulate ammonium, nitrate and sulfate followed a downward trend from 2000 to 2003 and a slight increase during 2004, which is consistent with their atmospheric concentrations. On the other hand, while SO2 and HNO3 dry deposition was significantly reduced in 2001, deposited amounts increased progressively from 2002 to 2004.

Figure 14 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate during the warm period (2rd -3rd quarter) and significant input of SO_2 and nitrate during the cold period (1st and 4th quarters)

Figure 15 Total nitrogen and sulfur dry deposition at Cadiz during 2000-2004

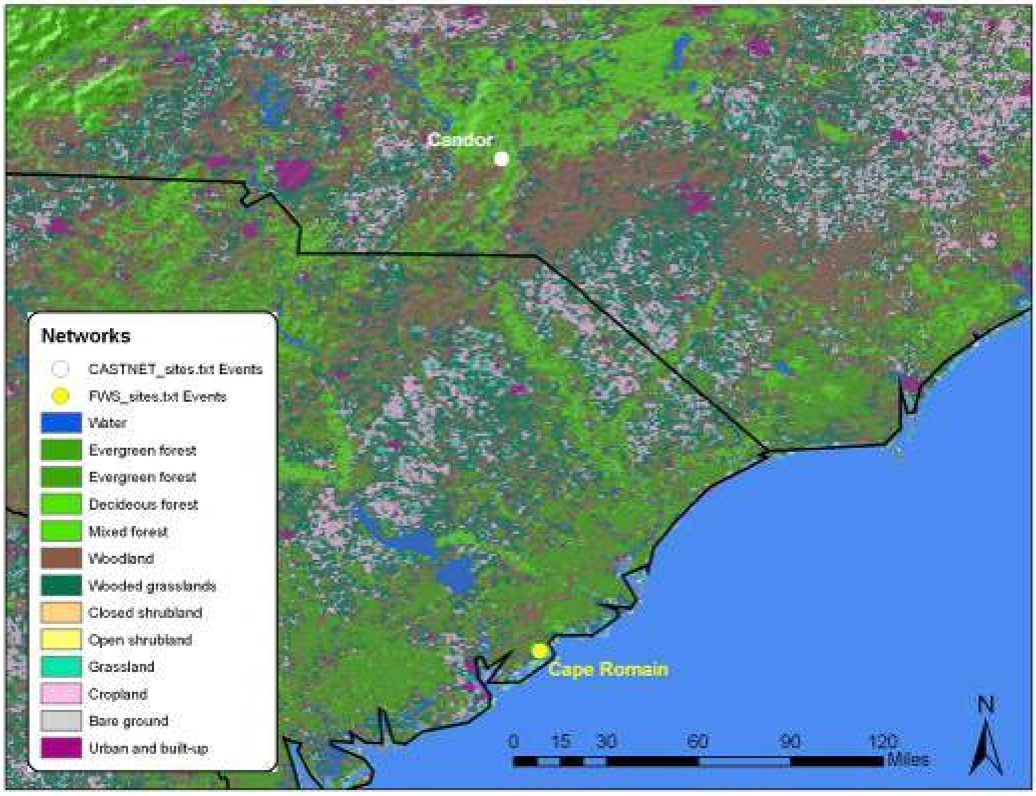


The total amount of nitrogen and sulfur deposited in Cadiz, KY, ranged approximately from 2.0 kg/ha to 3.0 kg/ha and from 4.0 kg/ha up to 5.2 kg/ha, respectively. A clear inter-annual pattern is observed for both elements with comparables amounts of dry deposition in 2000 and 2004, providing evidence of an increasing trend during the last two years.

B.3 Candor, North Carolina

Candor North Carolina is used as a surrogate CASTNET site for FWS Cape Romain site in South Carolina. Figure 16 shows the locations of both sites.

Figure 16 Location of CASTNET Candor and FWS Cape Romain sites.



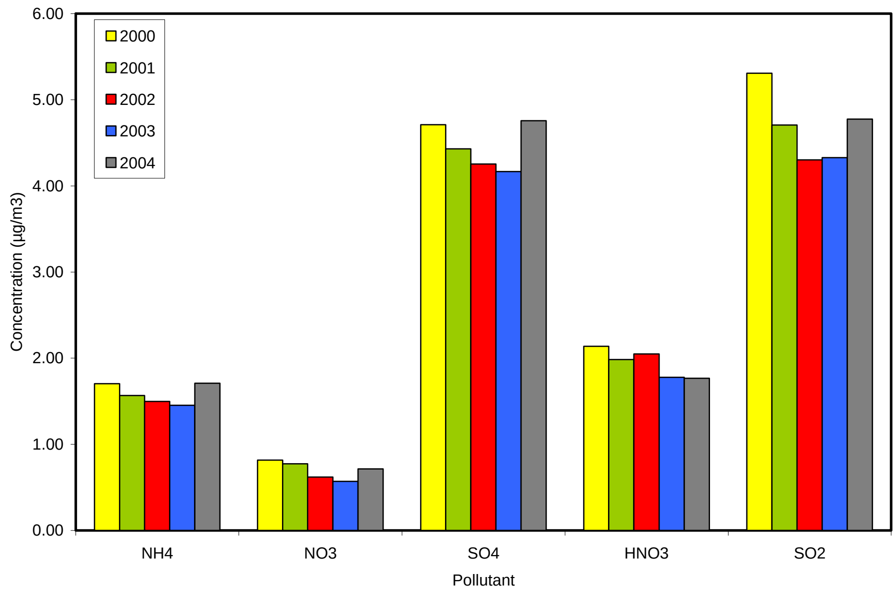
B.3.1 Ambient air

Ambient air concentration levels of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Candor, NC during the 2000-2004 period, are presented in Table 8.

Table 8 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Candor NC for 2000-2004 periods

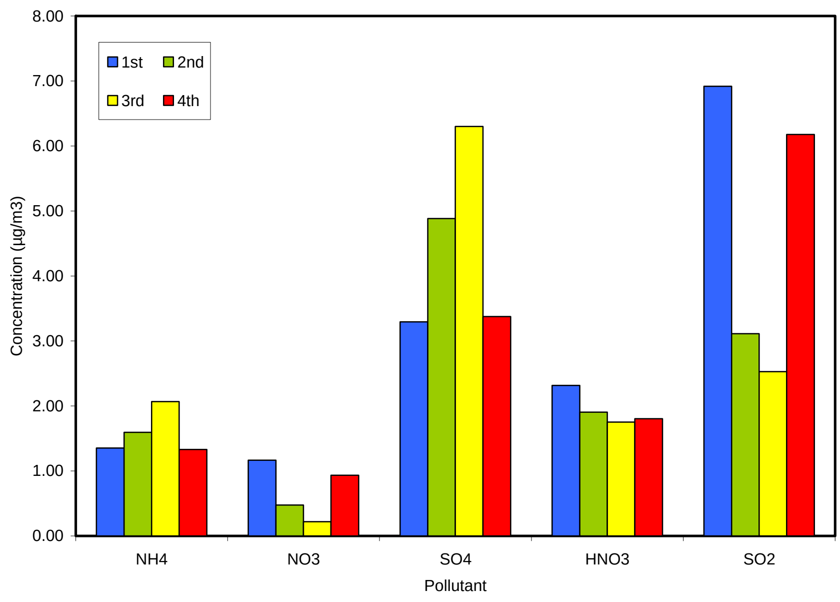
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.6	0.3	6.1	0.7
Nitrate (NO_3^-)	0.7	0.0	1.5	0.6
Sulfate (SO_4^{2-})	4.5	1.2	13.5	2.1
Gas Phase				
Sulfur dioxide (SO_2)	4.7	0.5	21.4	3.0
Nitric Acid (HNO_3)	1.9	0.5	4.4	0.7

Figure 17 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 17, concentrations of particulate components (ammonium, nitrate and sulfate) decreased from 2000 to 2003, while a slight increase is observed for 2004. A similar profile is plotted for atmospheric concentrations of sulfur dioxide and nitric acid.

Figure 18 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2^{nd} (April-June) and 3^{rd} (July-September) for ammonium are measured. The differences are more pronounced for sulfate, while an opposite profile is drawn for sulfur dioxide, nitric acid and nitrate, with higher concentration being measured during winter.

Figure 19 Scatter plot of NH4 nanoequivalents per m3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

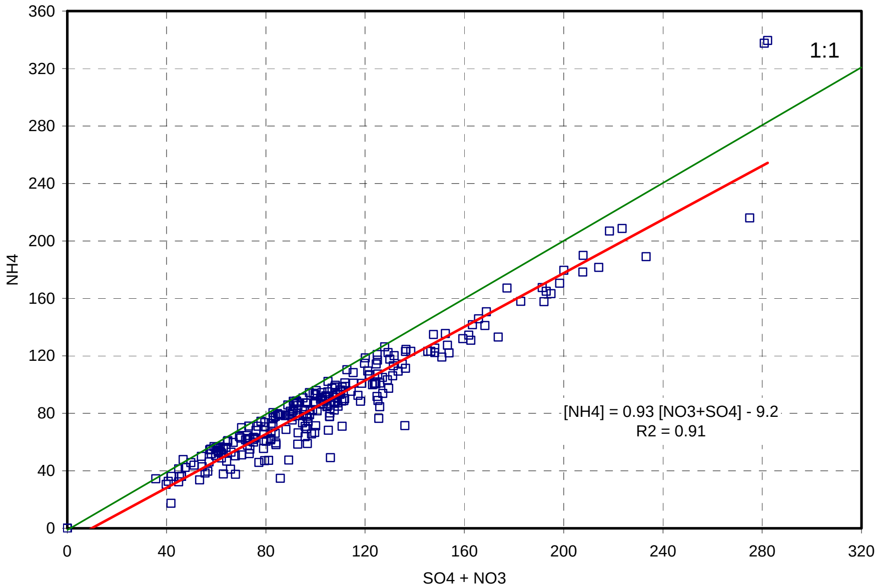


Figure 19 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 93% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 and $(NH_4)_2SO_4$, and minor amounts of H_2SO_4 and HNO_3 if any.

Table 9 shows the inteannual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be more significant during the 2nd and 3rd quarter.

Table 9 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.88	0.85	0.82	0.86	1.10
Intercept	-3.4	-2.0	2.9	-1.7	-26.55
R	0.97	0.98	0.97	0.97	0.96
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.89	0.86	1.02	0.94	
Intercept	-1.9	-5.7	-23.1	-6.8	
R	0.95	0.91	0.96	0.97	

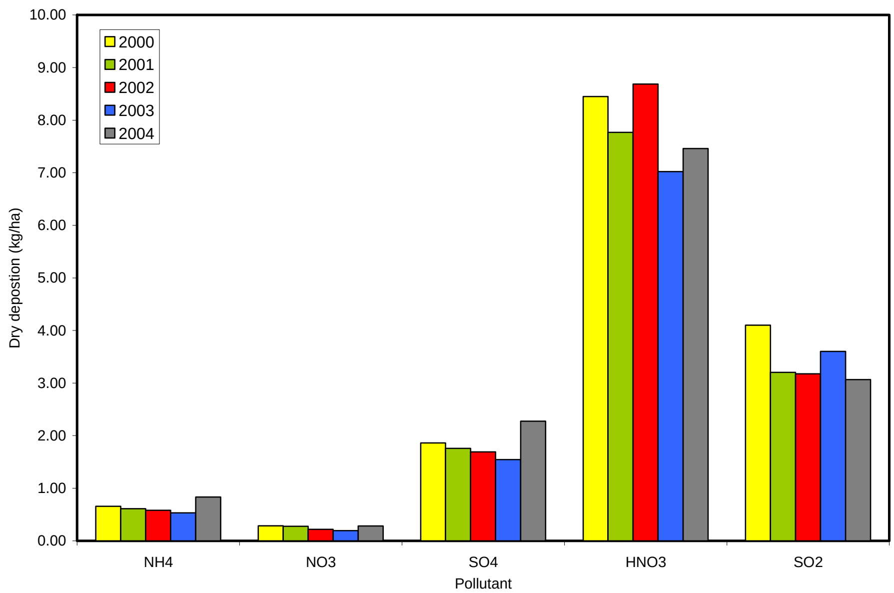
B.3.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Candor NC during the 2000-2004 period, are presented in Table 10.

Table 10 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Candor NC for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

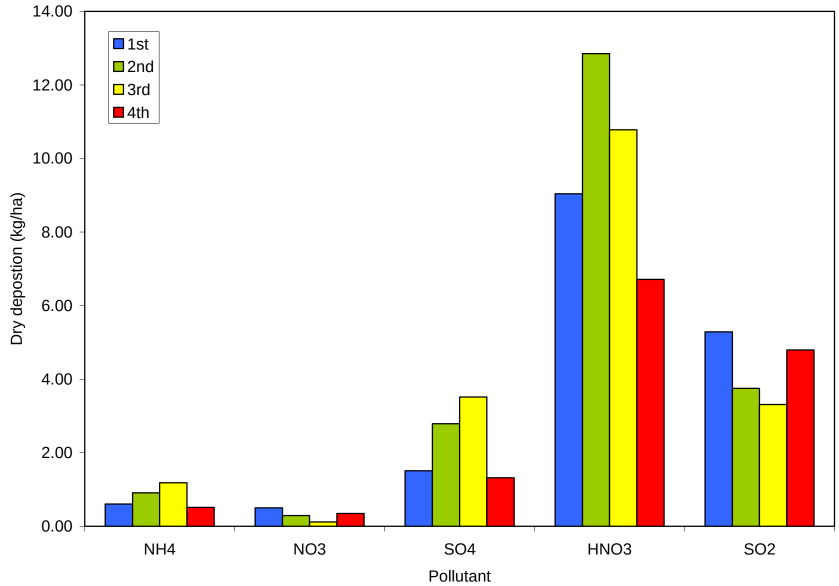
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	13	1	176	12	2.5
Nitrate (NO_3^-)	5	0	26	4	0.3
Sulfate (SO_4^{2-})	36	4	389	30	3.0
Gas Phase					
Sulfur dioxide (SO_2)	67	14	207	32	8.6
Nitric Acid (HNO_3)	155	42	505	69	8.8
Total Nitrogen					11.5
Total Sulfur					11.6

Figure 20 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



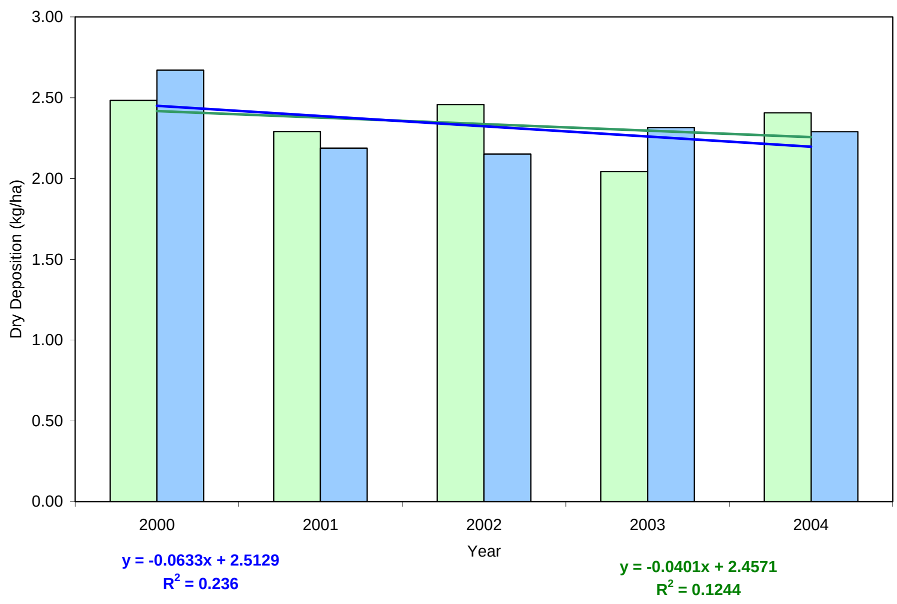
As shown in Figure 20, annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2) the amounts of particulate ammonium, nitrate and sulfate followed a downward trend from 2000 to 2003 and a slight increase during 2004, which is consistent with their atmospheric concentrations. On the other hand, dry deposition rates of SO2 and HNO3 fluctuated during 2000-2004.but indicated a rather downward trend.

Figure 21 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate including nitric acid during the warm period (2^{rd} -3^{rd} quarter) and significant input of SO_2 and nitrate during the cold period (1^{st} and 4^{th} quarters)

Figure 22 Total nitrogen and sulfur dry deposition at Candor during 2000-2004

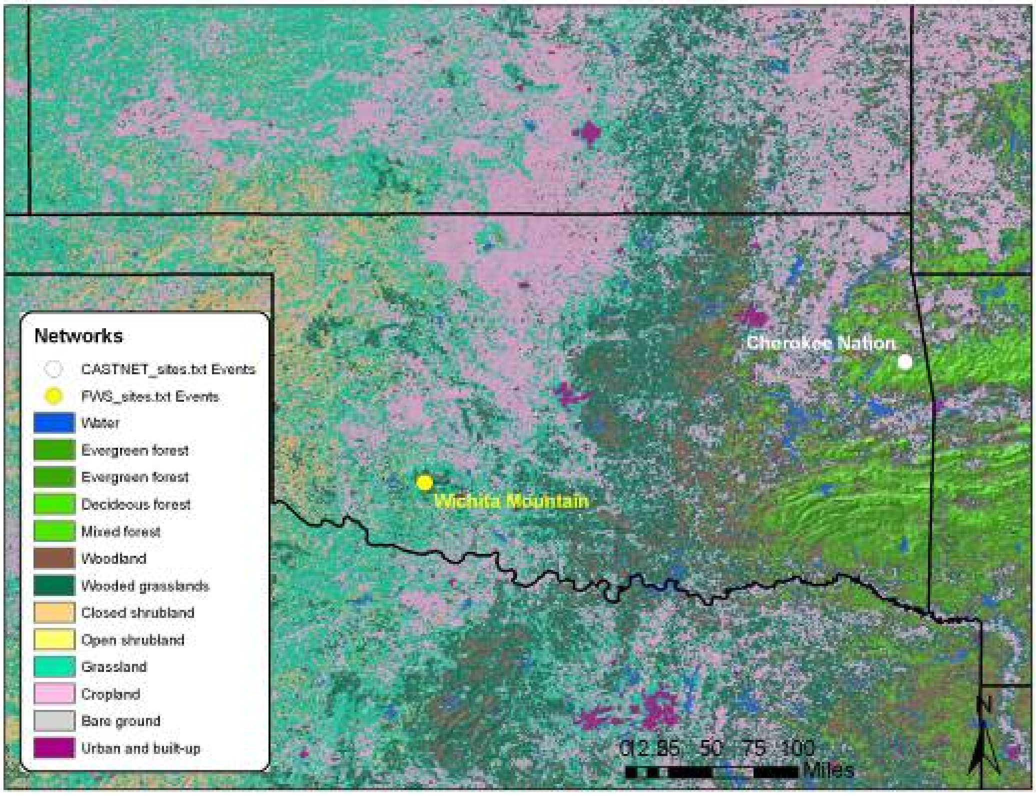


The total amount of nitrogen and sulfur deposited in Candor, ranged approximately from 2.0 kg/ha to 2.5 kg/ha and from 2.2 kg/ha up to 2.7 kg/ha, respectively. A clear inter-annual pattern is observed for both elements with a decreasing trend of dry deposition during the last years.

B.4 Cherokee Nation, Oklahoma

Cherokee Nation in Oklahoma is used as a surrogate CASTNET site for FWS Wichita Mountain site in Oklahoma. Figure 23 shows the locations of both sites.

Figure 23 Location of CASTNET Cherokee Nation and FWS Wichita Mountain sites.



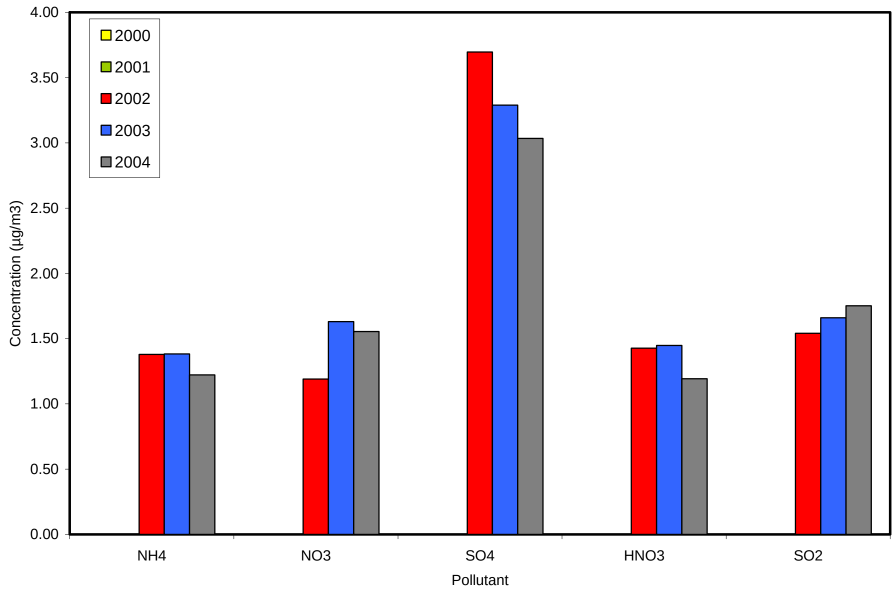
B.4.1 Ambient air

Ambient air concentration levels of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Cherokee Nation, OK during the 2000-2004 period, are presented in Table 11. Ambient air and dry deposition measurements are available from 2002 to 2004.

Table 11 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Cherokee Nation OK for 2000-2004 periods

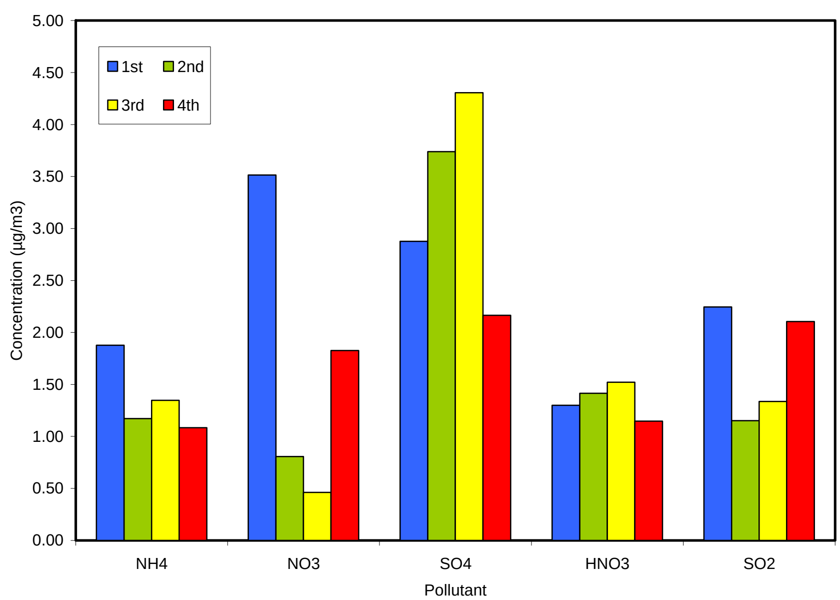
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.3	0.5	3.9	0.6
Nitrate (NO_3^-)	1.5	0.0	7.4	1.5
Sulfate (SO_4^{2-})	3.3	0.8	10.8	1.6
Gas Phase0				
Sulfur dioxide (SO_2)	1.7	0.3	4.7	0.9
Nitric Acid (HNO_3)	1.4	0.3	4.2	0.5

Figure 24 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 24, concentrations of ammonium, sulfate and nitric acid decreased from 2002 to 2004. On the other hand, nitrate and sulfur dioxide concentrations increased progressively.

Figure 25 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate and nitric acid are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate ammonium and nitrate, with higher concentration being measured during winter.

Figure 26 Scatter plot of NH4 nanoequivalents per m3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

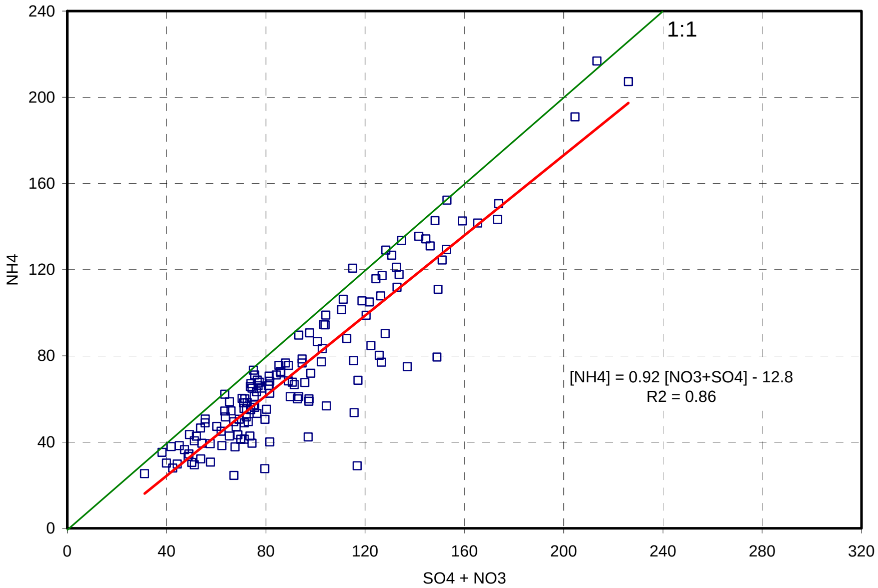


Figure 26 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 93% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 and $(NH_4)_2SO_4$, and minor amounts of H_2SO_4 and HNO_3 if any.

Table 12 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be significant throughout the year although a rathe low slope is computed for spring during the 2nd and 3rd quarter.

Table 12 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope			0.94	0.96	0.97
Intercept			-13.5	-14.3	-8.3
R			0.98	0.94	0.85
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	1.05	0.58	1.00	0.88	
Intercept	-18.3	12.7	-22.5	-5.9	
R	0.97	0.73	0.95	0.94	

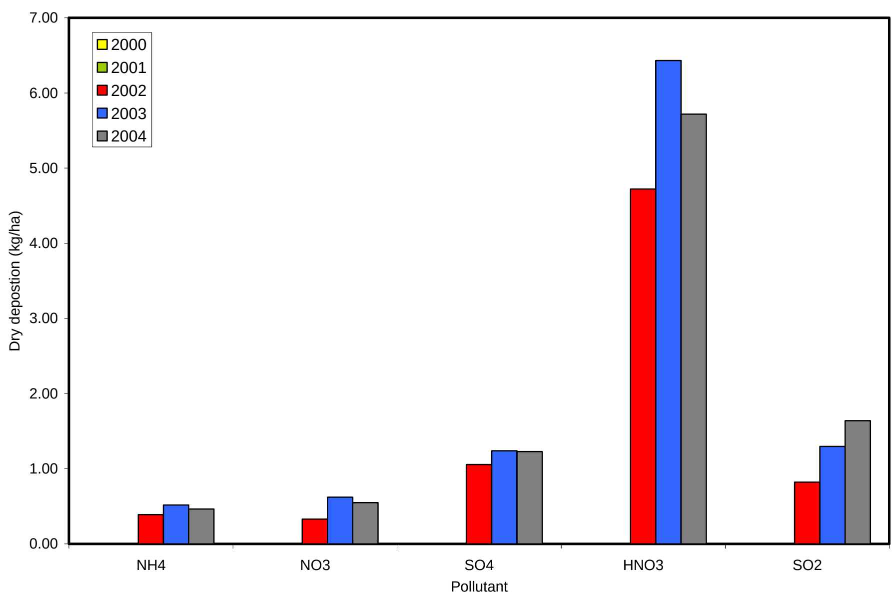
B.4.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Cherokee Nation OK during the 2000-2004 period, are presented in Table 13.

Table 13 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Cherokee Nation OK for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

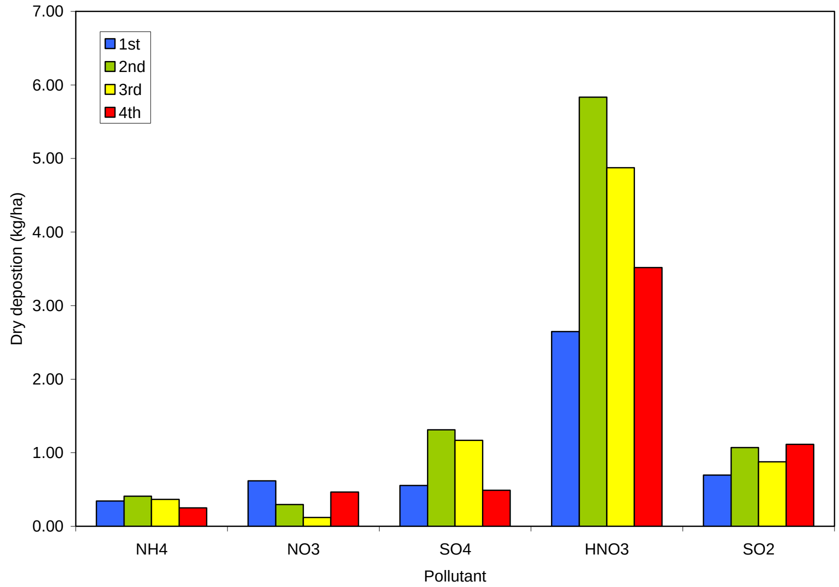
	Mean	**Minimum**	**Maximum**	**Std Deviation**	**Total deposition**
Aerosol					
Ammonium (NH_4^+)	10	2	33	6	1.1
Nitrate (NO_3^-)	11	0	69	11	0.3
Sulfate (SO_4^{2-})	27	5	79	16	1.2
Gas Phase					
Sulfur dioxide (SO_2)	28	8	86	13	1.9
Nitric Acid (HNO_3)	128	36	408	61	3.7
Total Nitrogen					5.2
Total Sulfur					3.1

Figure 27 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



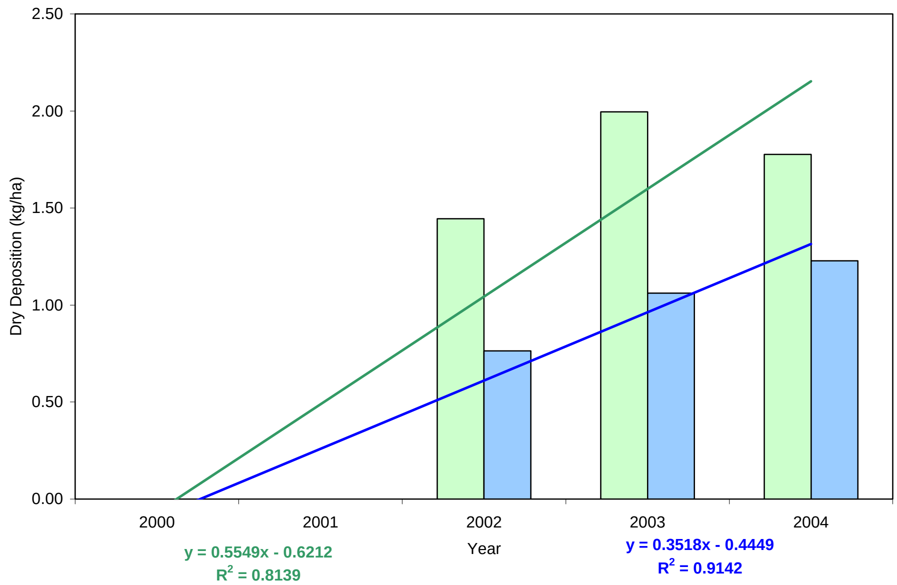
As shown in Figure 20, the amounts of all sulfur- and nitrogen–compounds fluctuated from 2002 to 2004 following an upward trend.

Figure 28 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate including nitric acid during the warm period (2rd -3rd quarter) and significant input of SO_2 and nitrate during the cold period (1st and 4th quarters)

Figure 29 Total nitrogen and sulfur dry deposition at Cherokee Nation during 2000-2004

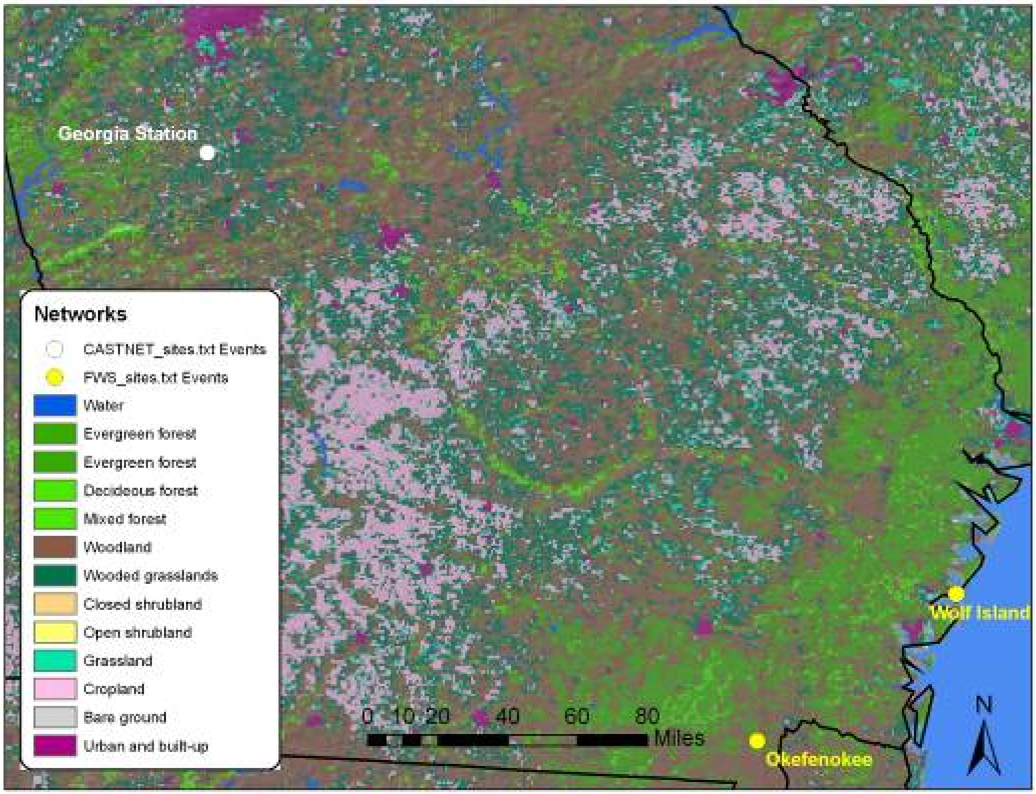


The total amount of nitrogen and sulfur deposited in Cherokee Nation OK, ranged approximately from 1.5 kg/ha to 2.0 kg/ha and from 0.7 kg/ha up to 1.2 kg/ha, respectively. A clear inter-annual pattern is observed for both elements with an increasing trend during the last years.

B.5 Georgia Station, Georgia

Georgia Station in Georgia is used as a surrogate CASTNET site for FWS Wolf Island in Georgia. Figure 37 shows the locations of both sites.

Figure 30 Location of CASTNET Georgia Station and FWS Wolf Island sites.



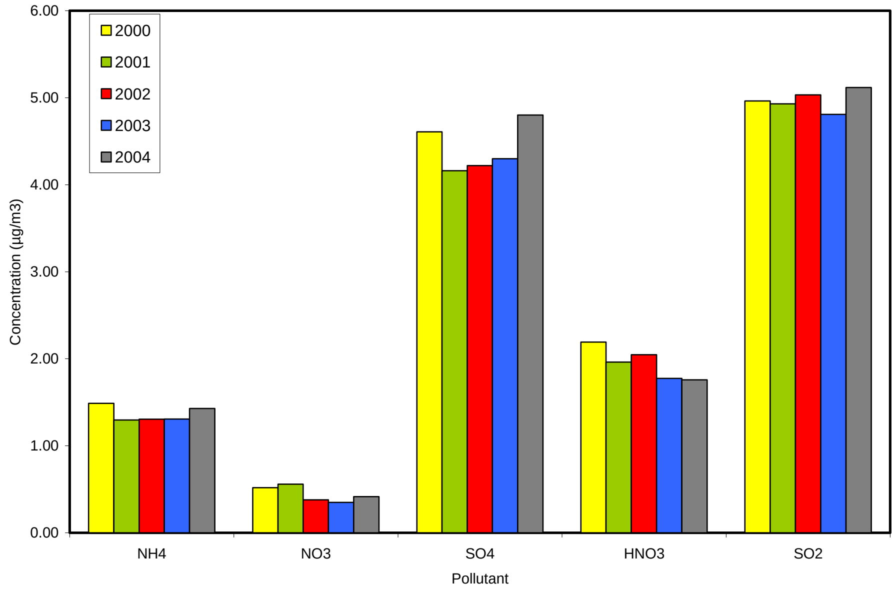
B.5.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Georgia Station during the 2000-2004 period, are presented in Table 14.

Table 14 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Georgia Station for 2000-2004 periods

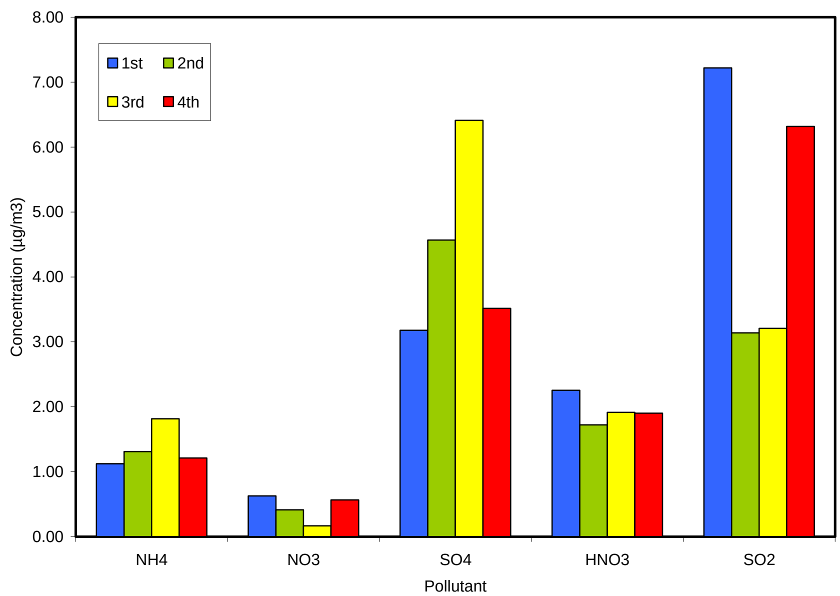
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.4	0.4	4.4	0.6
Nitrate (NO_3^-)	0.4	0.0	1.9	0.4
Sulfate (SO_4^{2-})	4.4	1.4	14.2	2.3
Gas Phase				
Sulfur dioxide (SO_2)	5.0	0.4	24.2	3.1
Nitric Acid (HNO_3)	1.9	0.5	4.7	0.7

Figure 31 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 31, concentrations of all components did not vary since 2000, while for most of them concentrations are increased in 2004.

Figure 32 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate and amoonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher concentration being measured during winter.

Figure 33 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

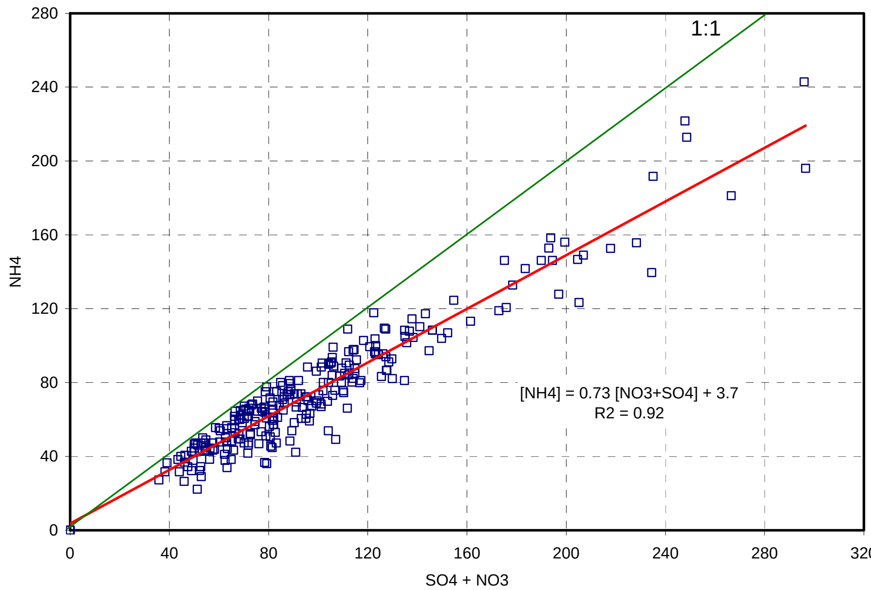


Figure 33 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 75% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 and $(NH_4)_2SO_4$, and minor amounts of H_2SO_4 and HNO_3.

Table 15 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be significant throughout the year although a rather low slope is computed for spring during the 2nd and 3rd quarter.

Table 15 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.82	0.70	0.73	0.70	0.70
Intercept	-1.9	4.8	3.7	6.2	5.1
R	0.97	0.95	0.95	0.96	0.96
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.75	0.69	0.71	0.86	
Intercept	4.5	2.5	4.2	-3.7	
R	0.88	0.91	0.97	0.98	

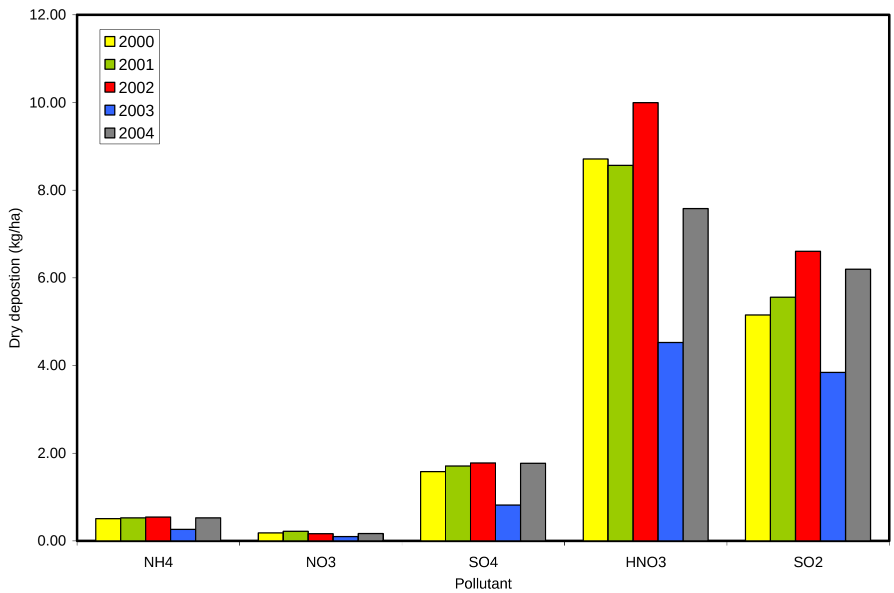
B.5.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Georgia Station during the 2000-2004 period, are presented in Table 16.

Table 16 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Georgia Station for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

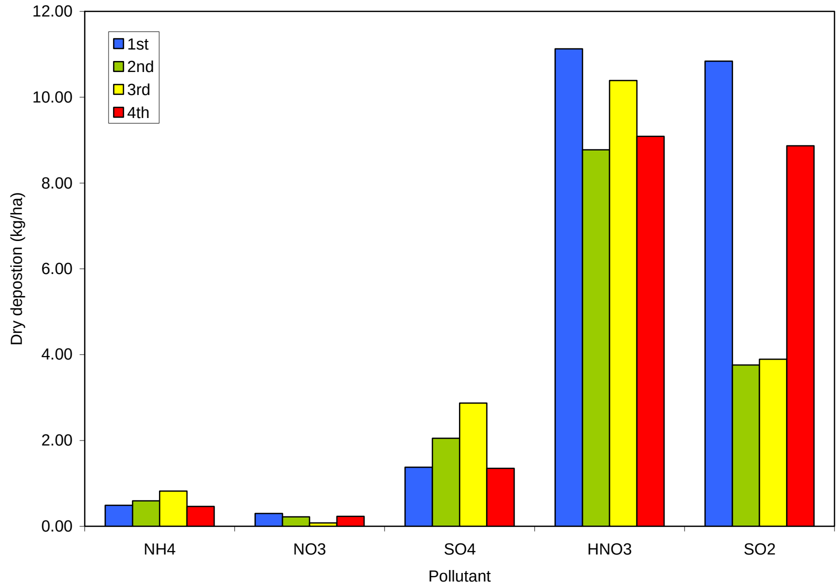
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	10	3	35	5	1.8
Nitrate (NO_3^-)	4	0	22	3	0.2
Sulfate (SO_4^{2-})	33	10	116	20	2.6
Gas Phase					
Sulfur dioxide (SO_2)	119	15	603	84	13.7
Nitric Acid (HNO_3)	172	54	416	64	8.8
Total Nitrogen					10.8
Total Sulfur					16.2

Figure 34 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



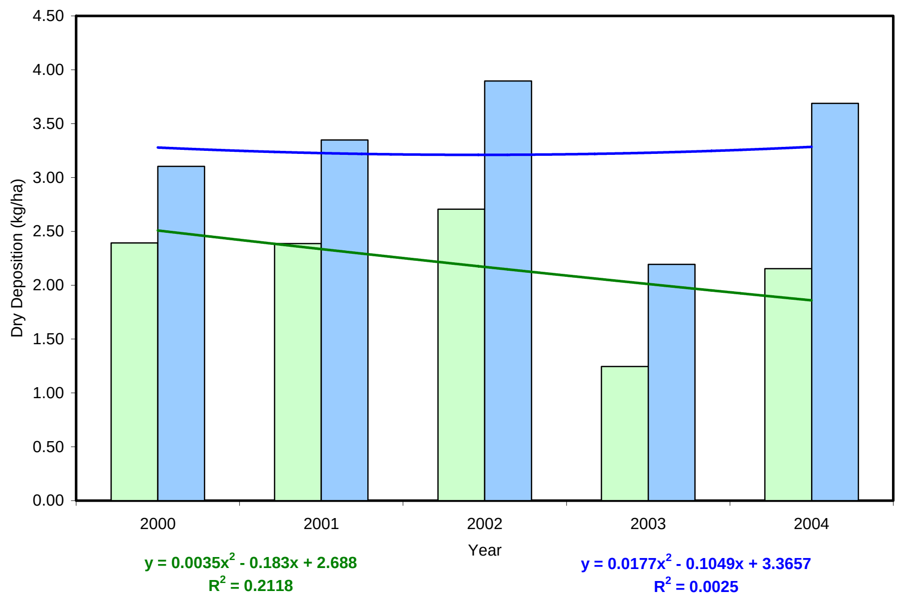
As shown in Figure 34, the amounts of all compounds did not vary significantly from 2000 to 2004 with significantly lower concentrations during 2003.The annual variation indicates a rather increasing trend for all components.

Figure 35 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate including nitric acid during the warm period (2^{rd} -3^{rd} quarter) and significant input of SO_2 and nitrate during the cold period (1^{st} and 4^{th} quarters)

Figure 36 Total nitrogen and sulfur dry deposition at Georgia Station during 2000-2004

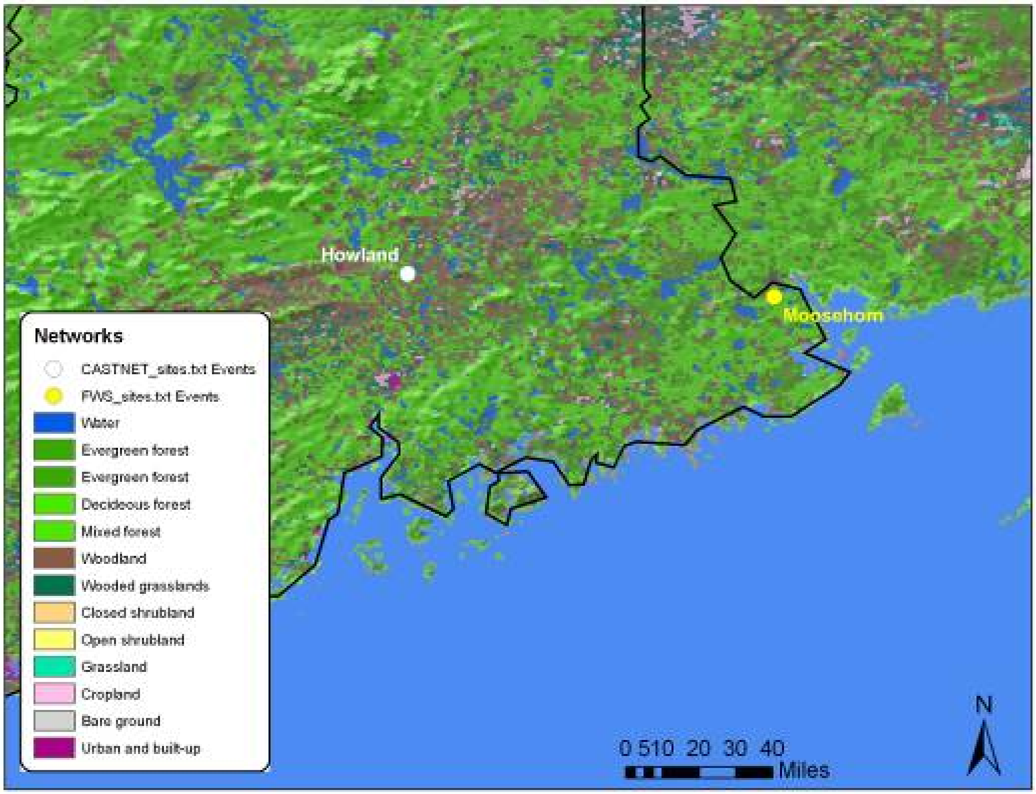


The total amount of nitrogen and sulfur deposited in Georgia Station, ranged approximately from 1.2 kg/ha to 2.7 kg/ha and from 2.2 kg/ha up to 3.9 kg/ha, respectively. Statistically insignificant trends are observed for both elements primarily due to the low deposition during 2003.

B.6 Howland, Maine

Howland in Maine is used as a surrogate CASTNET site for FWS Moosehorn in Maine. Figure 37 shows the locations of both sites.

Figure 37 Location of CASTNET Howland and FWS Moosehorn sites.



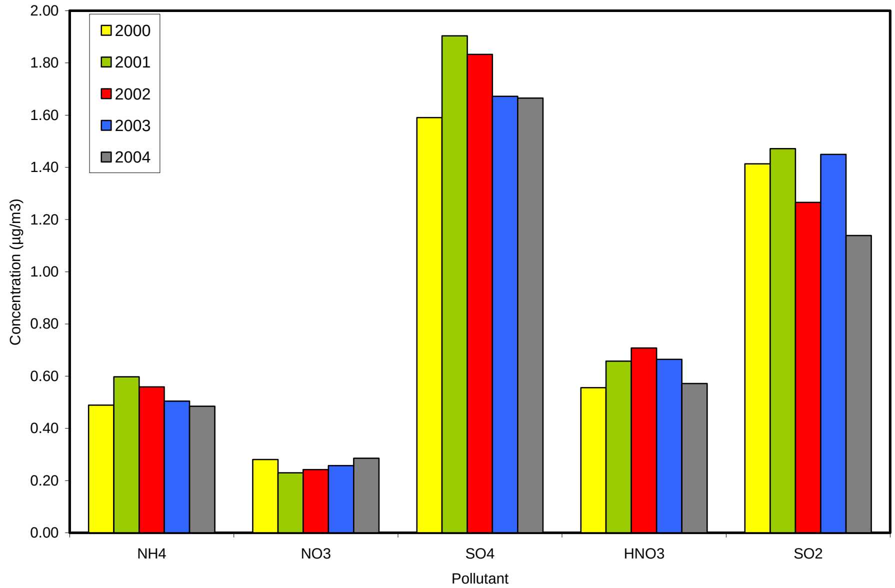
B.6.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Howland during the 2000-2004 period, are presented in Table 17.

Table 17 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Howland for 2000-2004 periods

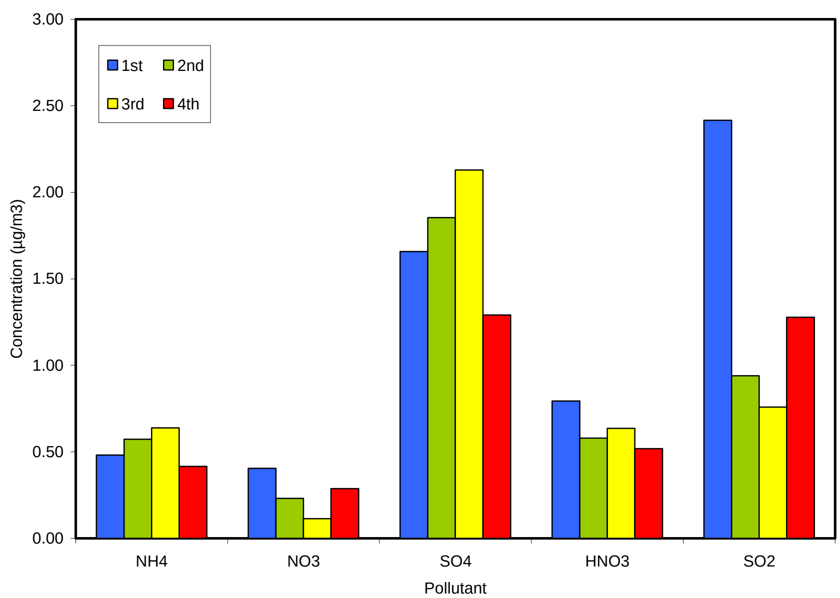
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	0.5	0.1	2.3	0.3
Nitrate (NO_3^-)	0.2	0.0	2.7	0.3
Sulfate (SO_4^{2-})	1.7	0.3	8.5	1.0
Gas Phase				
Sulfur dioxide (SO_2)	1.3	0.2	5.9	1.0
Nitric Acid (HNO_3)	0.6	0.1	2.1	0.3

Figure 38 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 38, concentrations of ammonium, sulfate and sulfur dioxide increase during 2001 followed by decreasing concentrations from 2002 to 2004. Nitrate concentrations increased with comparable levels for 2000 and 2004, while nitric acid increased progressively from 2000 to 2002 followed by a decreasing trend to 2004.

Figure 39 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate and amoonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher concentration being measured during winter.

Figure 40 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

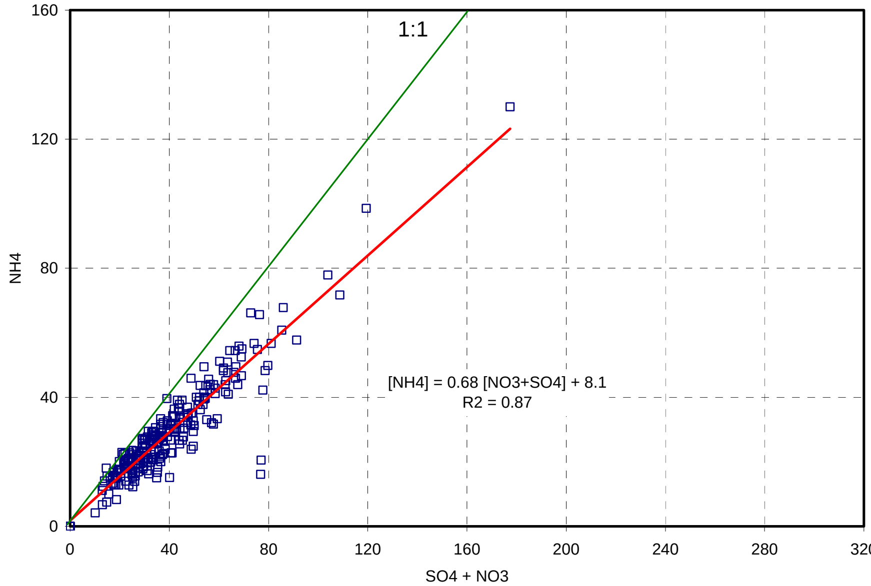


Figure 40 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 68% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, H_2SO_4 and HNO_3. and considerable amounts of NH_4NO_3 and $(NH_4)_2SO_4$,

Table 18 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be significant throughout the year although a rather low slope is computed for spring during the 2nd and 3rd quarter.

Table 18 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.59	0.75	0.70	0.72	0.57
Intercept	5.1	0.8	1.5	0.4	4.5
R	0.82	0.97	0.96	0.97	0.88
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.58	0.72	0.68	0.64	
Intercept	2.7	1.1	4.3	2.9	
R	0.87	0.96	0.97	0.86	

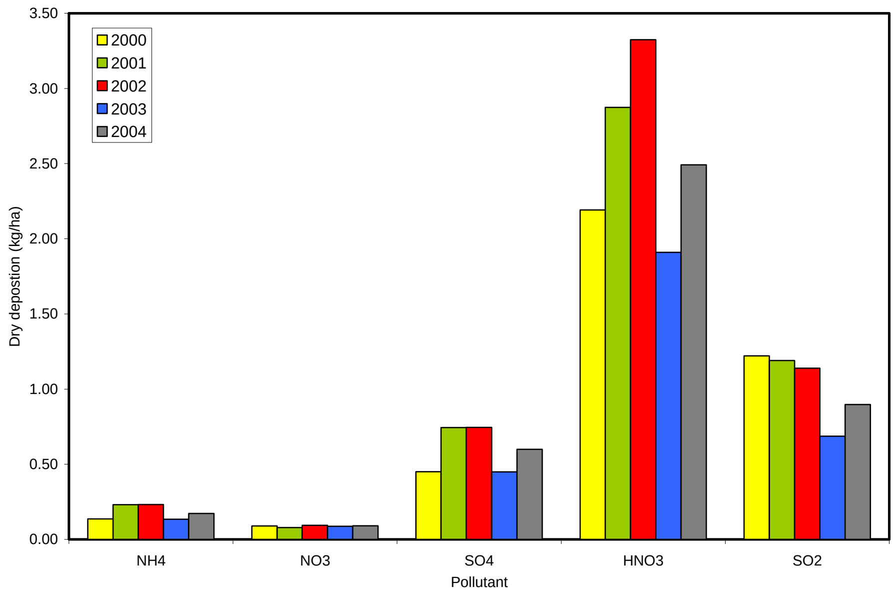
B.6.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Howland during the 2000-2004 period, are presented in Table 19.

Table 19 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Howland for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

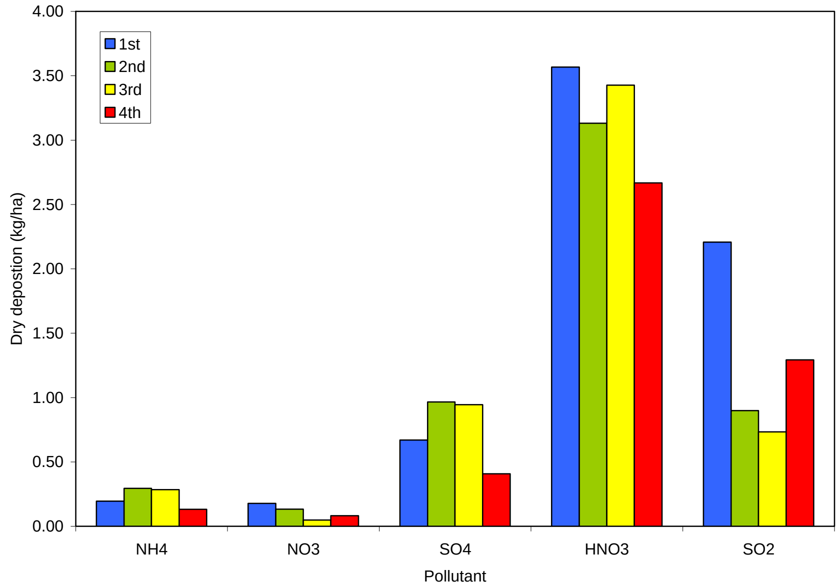
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	4	1	21	3	0.7
Nitrate (NO_3^-)	2	3	38	3	0.1
Sulfate (SO_4^{2-})	13	2	77	10	1.0
Gas Phase					
Sulfur dioxide (SO_2)	22	4	90	15	2.6
Nitric Acid (HNO_3)	55	5	206	33	4.3
Total Nitrogen					5.1
Total Sulfur					3.6

Figure 41 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



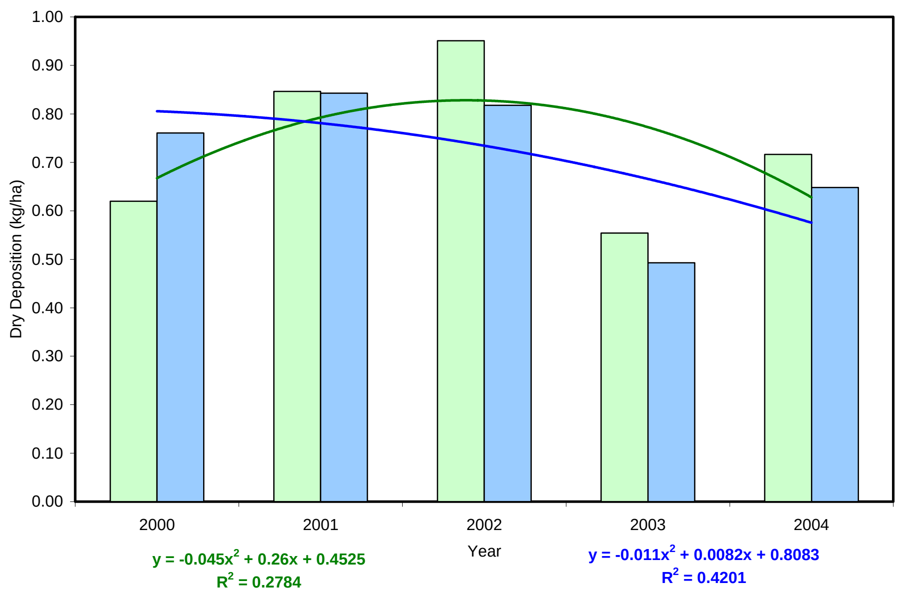
As shown in Figure 41, for all components, lower depositions are estimated during 2003. Overall, an upward trend is observed for ammonium, sulfate and nitric acid, while no significant changes are estimated for nitrate and sulfur dioxide followed a decreasing trend.

Figure 42 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate including nitric acid during the warm period (2^{rd} -3^{rd} quarter) and significant input of SO_2 and nitrate during the cold period (1^{st} and 4^{th} quarters)

Figure 43 Total nitrogen and sulfur dry deposition at Howland during 2000-2004

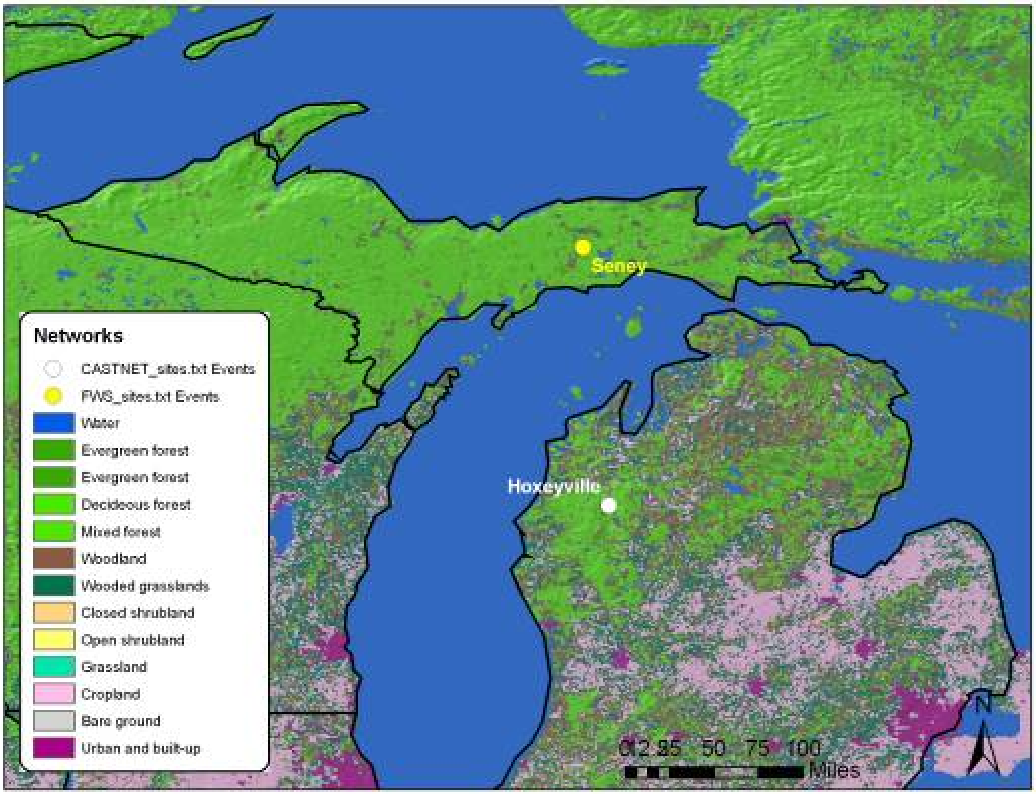


The total amount of nitrogen and sulfur deposited in Howland, ranged approximately from 0.5 kg/ha to 0.9 kg/ha and from 0.5 kg/ha up to 0.8 kg/ha, respectively. While deposition of sulfur and nitrogen increased during 2000-2002, a downward pattern is observed during tha last two years.

B.7 Hoxeyville, Michigan

Hoxeyville in Michingan is used as a surrogate CASTNET site for FWS Seney in Michingan. Figure 44 shows the locations of both sites.

Figure 44 Location of CASTNET Hoxeyville and FWS Seney sites.



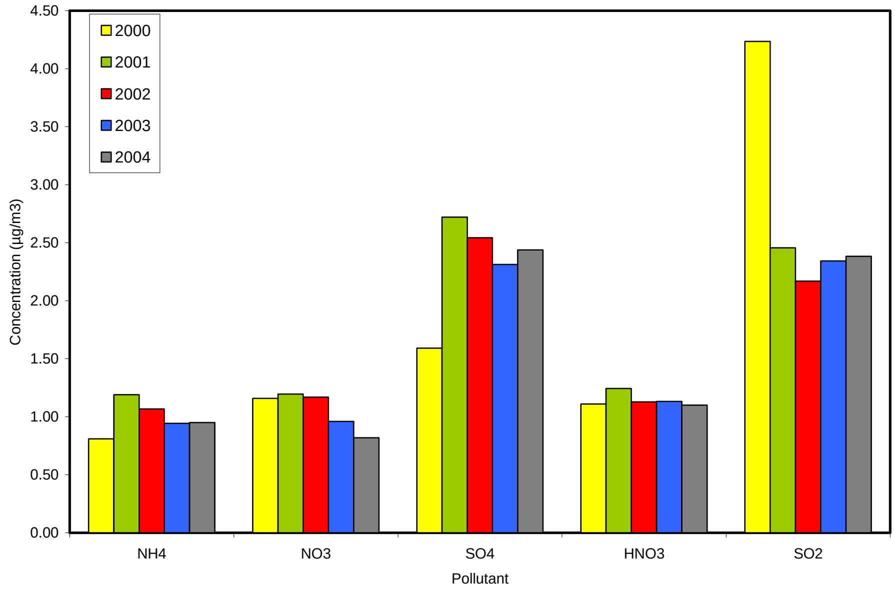
B.7.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Hoxeyville during the 2000-2004 period, are presented in Table 20.

Table 20 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Hoxeyville for 2000-2004 periods

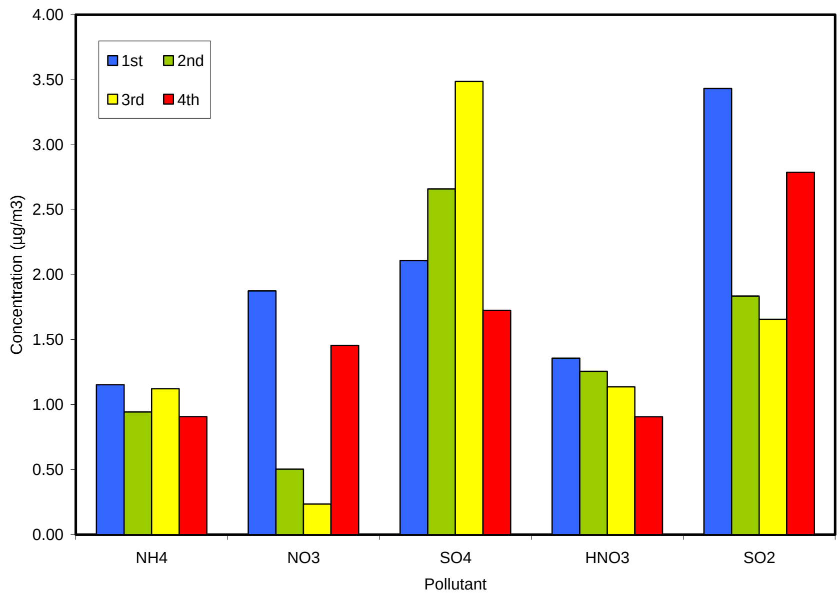
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.0	0.	3.8	0.6
Nitrate (NO_3^-)	1.0	0.0	6.6	1.2
Sulfate (SO_4^{2-})	2.5	0.5	11.4	1.7
Gas Phase				
Sulfur dioxide (SO_2)	2.4	0.2	9.4	1.6
Nitric Acid (HNO_3)	1.1	0.2	3.4	0.6

Figure 45 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 45, concentrations of ammonium, sulfate and sulfur dioxide increase during 2001 followed by decreasing concentrations from 2002 to 2004..

Figure 46 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate and ammonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher concentration being measured during winter.

Figure 47 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

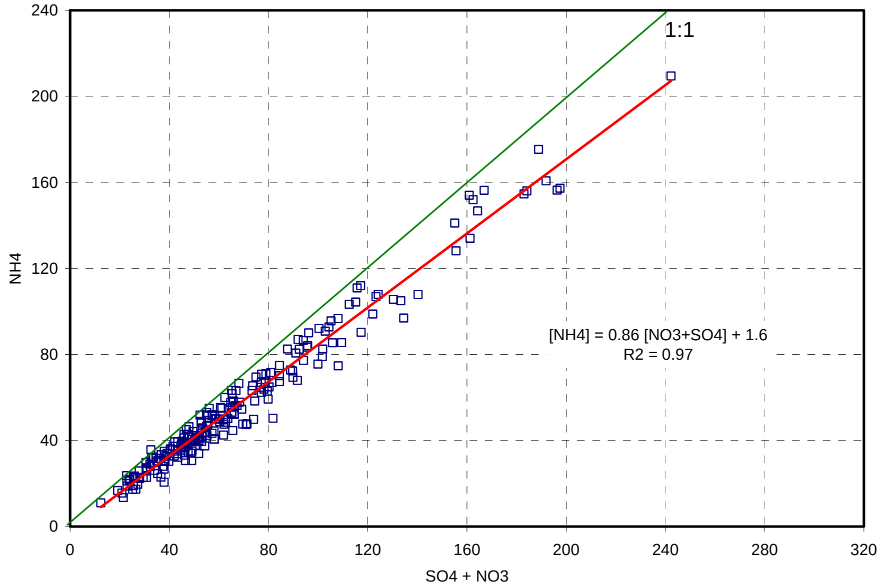


Figure 47 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 85% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 and $(NH_4)_2SO_4$ and minor amounts of H_2SO_4 and HNO_3,

Table 21 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be significant throughout the year although a rather low slope is computed for spring during the 2nd and 3rd quarter.

Table 21 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.94	0.90	0.84	0.84	0.84
Intercept	-3.5	-2.5	0.1	-0.8	-1.4
R	0.95	0.99	0.98	0.99	0.98
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.95	0.81	0.81	0.91	
Intercept	-6.3	0.6	0.6	-4.0	
R	0.99	0.98	0.99	0.98	

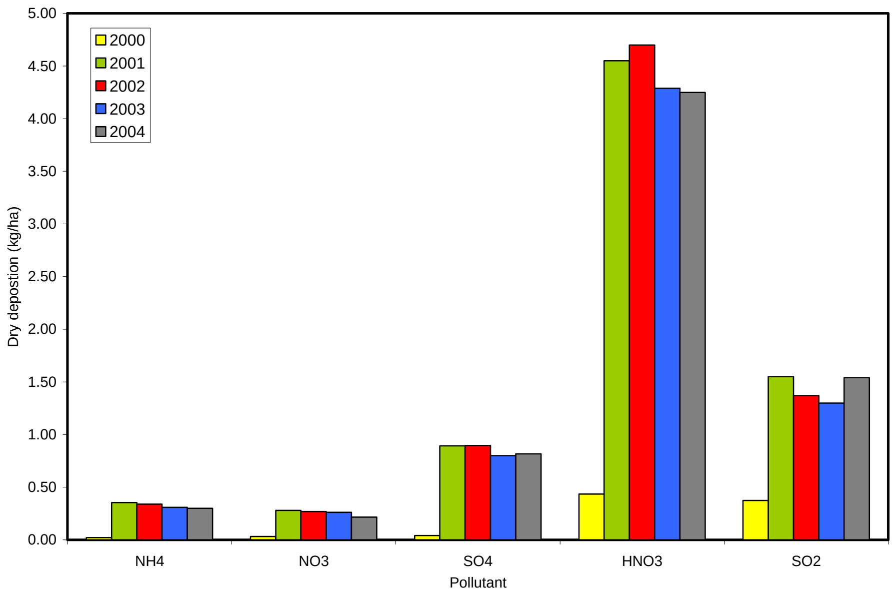
B.7.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Hoxeyville during the 2000-2004 period, are presented in Table 22.

Table 22 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Hoxeyville for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

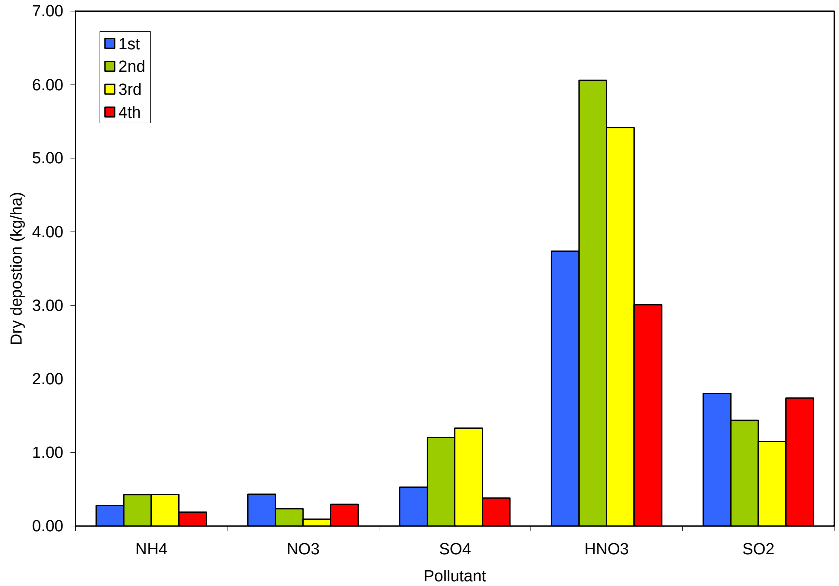
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	6	0	26	4	1.0
Nitrate (NO_3^-)	5	0	34	5	0.2
Sulfate (SO_4^{2-})	16	0	80	14	1.1
Gas Phase					
Sulfur dioxide (SO_2)	28	1	39	17	3.1
Nitric Acid (HNO_3)	84	1	278	53	6.1
Total Nitrogen					7.3
Total Sulfur					4.2

Figure 48 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



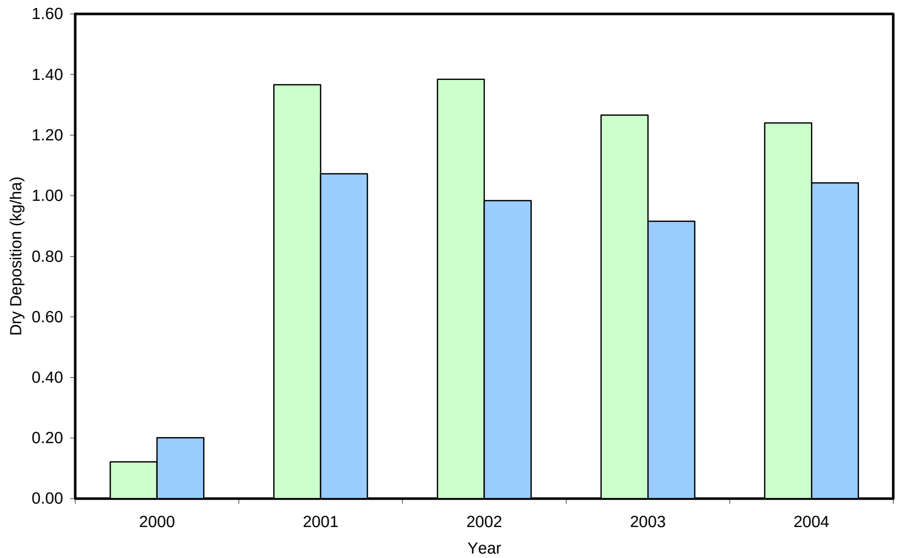
As shown in Figure 48, for all components, a rather downward trend was observed for 2001-2004 period.

Figure 49 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with highest contributions of particle-associated ammonium and sulfate including nitric acid during the warm period (2^{rd} -3^{rd} quarter) and significant input of SO_2 and nitrate during the cold period (1^{st} and 4^{th} quarters)

Figure 50 Total nitrogen and sulfur dry deposition at Hoxeyville during 2000-2004

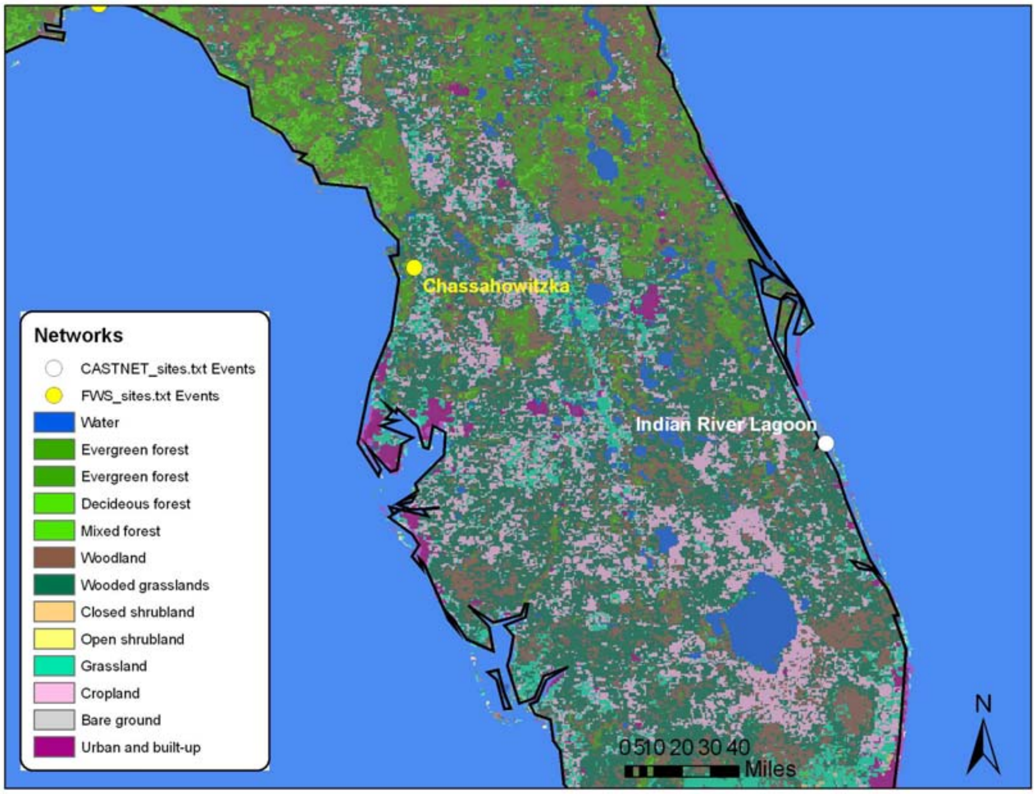


The total amount of nitrogen and sulfur deposited in Hoxeyville, ranged approximately from 0.1 kg/ha to 1.4 kg/ha and from 0.2 kg/ha up to 1.1 kg/ha, respectively. With the exception of 2000, a downward pattern is observed for both elements during the last years.

B.8 Indian River Lagoon, Florida

Indian River Lagoon in Florida is used as a surrogate CASTNET site for FWS Chassahowitzka in Florida. Figure 51 shows the locations of both sites.

Figure 51 Location of CASTNET Indian River Lagoon and FWS Chassahowitzka sites.



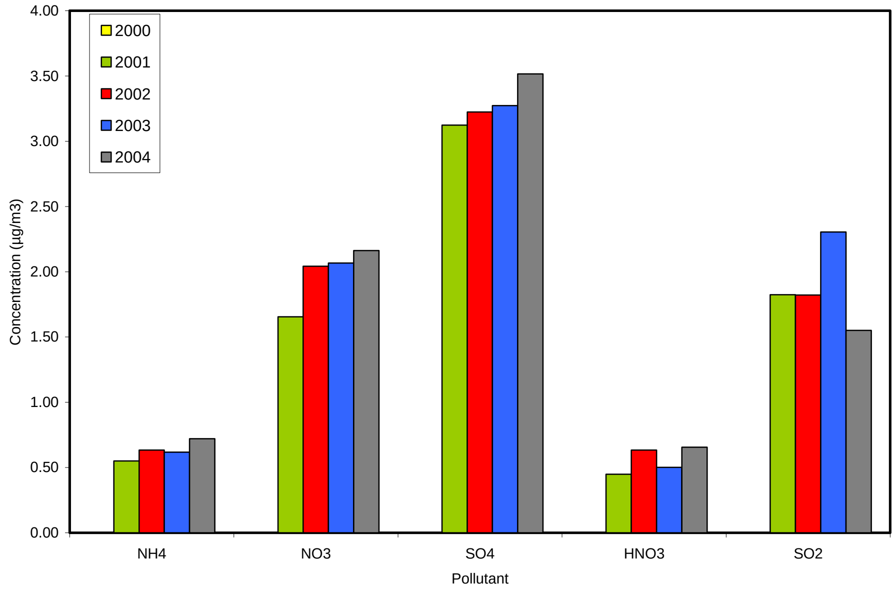
B.8.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Indian River Lagoon during the 2000-2004 period, are presented in Table 23. Measurements of ambient concentration and dry deposition started in 2001.

Table 23 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Indian River Lagoon for 2000-2004 periods

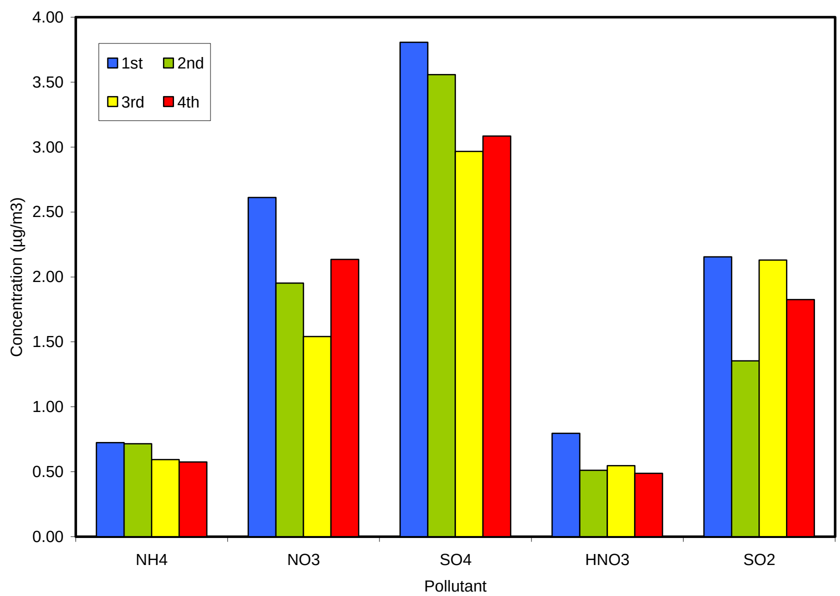
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	0.6	0.2	1.8	0.3
Nitrate (NO_3^-)	2.0	0.7	4.0	0.7
Sulfate (SO_4^{2-})	3.3	1.4	6.8	1.1
Gas Phase				
Sulfur dioxide (SO_2)	1.9	0.3	9.6	1.3
Nitric Acid (HNO_3)	0.6	0.1	2.2	0.5

Figure 52 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 52, concentrations of ammonium, sulfate and sulfur dioxide increase from 2001 to 2004, while concentration of gases varied substantially.

Figure 53 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations for almost all components are measured during winter and fall.

Figure 54 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

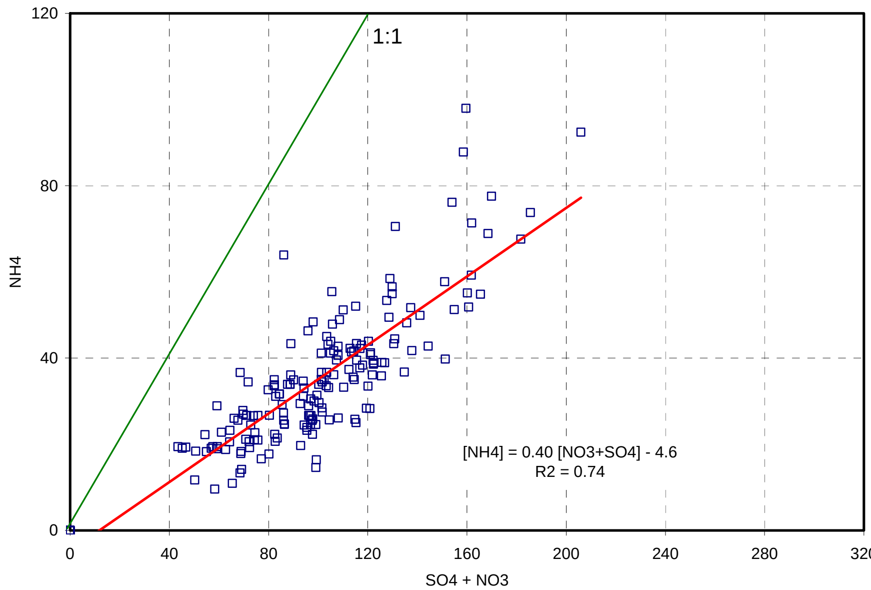


Figure 54 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 40% of nitrate and sulfate, which indicates that the predominant chemical components are H_2SO_4 HNO_3 and $(NH_4)HSO_4$, and minor amounts of NH_4NO_3 $(NH_4)_2SO_4$.

Table 24 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2002 to 2004. Neutralization by ammonia appears to be insignificant throughout the year and for different seasons.

Table 24 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope		0.42	0.40	0.40	0.40
Intercept		-8.2	-4.8	-6.3	-2.6
R		0.86	0.86	0.77	0.90
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.46	0.46	0.41	0.38	
Intercept	-16.3	-9.3	-2.7	-5.4	
R	0.81	0.85	0.89	0.89	

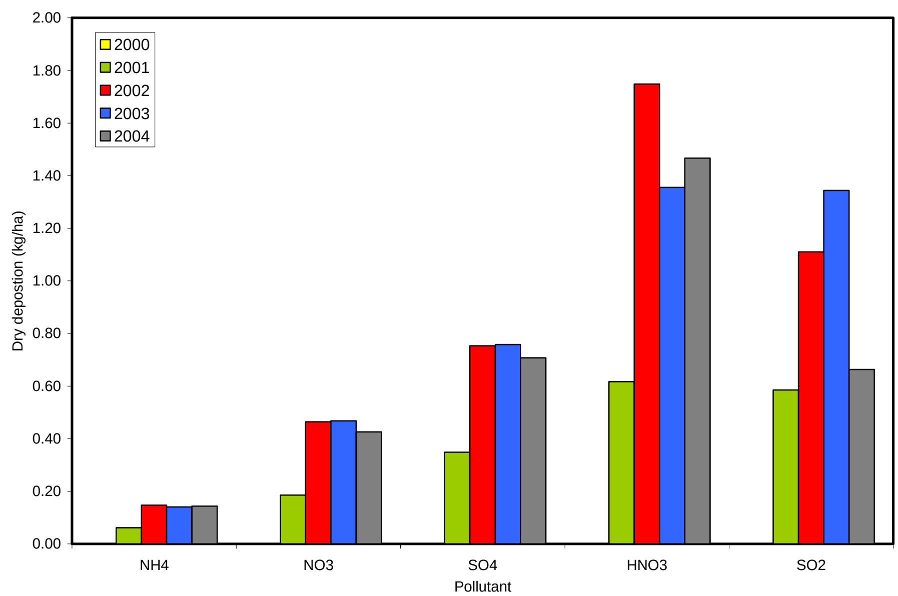
B.8.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Indian River Lagoon during the 2000-2004 period, are presented in Table 25.

Table 25 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Indian River Lagoon for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

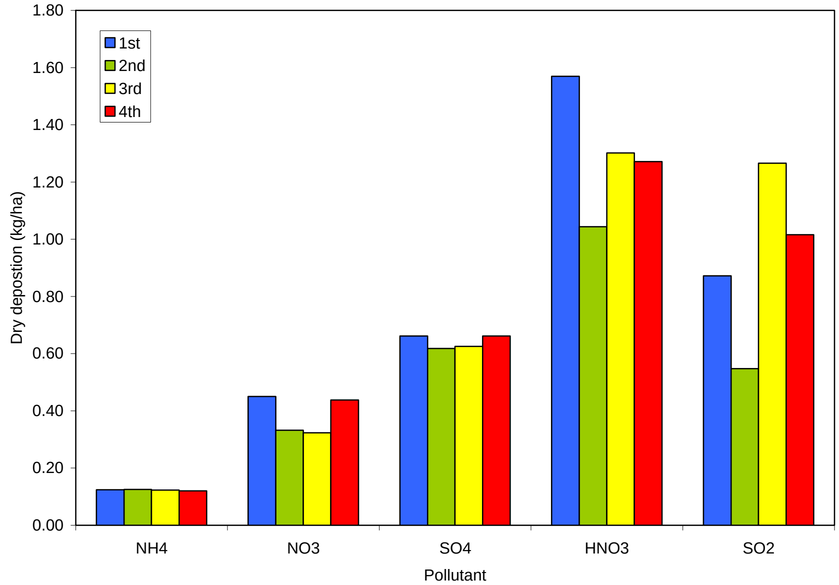
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	3	0	10	2	0.4
Nitrate (NO_3^-)	9	0	28	4	0.3
Sulfate (SO_4^{2-})	15	2	39	7	0.9
Gas Phase					
Sulfur dioxide (SO_2)	22	2	120	15	1.9
Nitric Acid (HNO_3)	30	2	108	24	1.7
Total Nitrogen					2.5
Total Sulfur					2.7

Figure 55 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



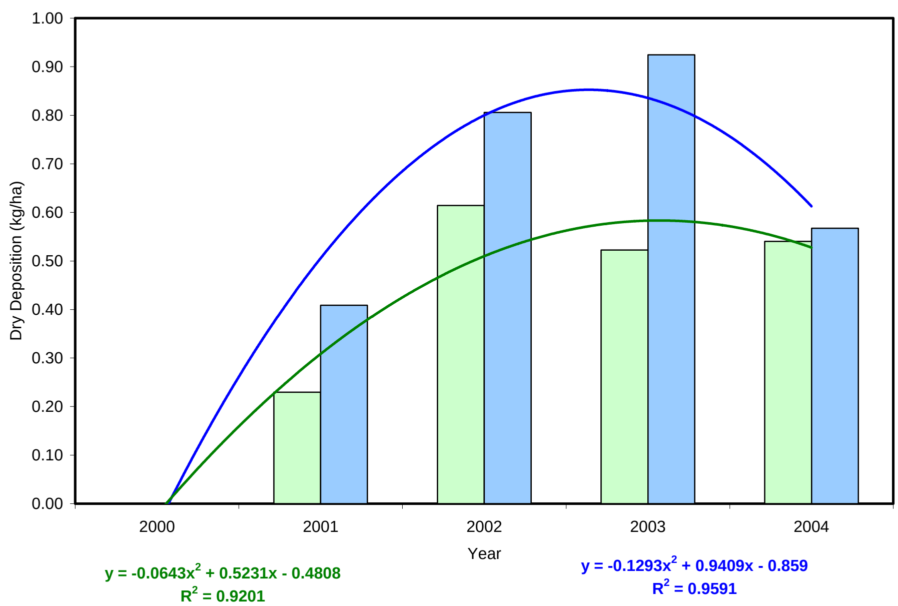
As shown in Figure 55, for all components, a rather downward trend was observed for 2001-2003 period followed by a slight decrease of dry deposition for 2004.

Figure 56 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, dry deposition follows a similar trend as compared to atmospheric concentrations with slightly higher concentrations for almost all components being measured during winter and fall.

Figure 57 Total nitrogen and sulfur dry deposition at Indian River Lagoon during 2000-2004

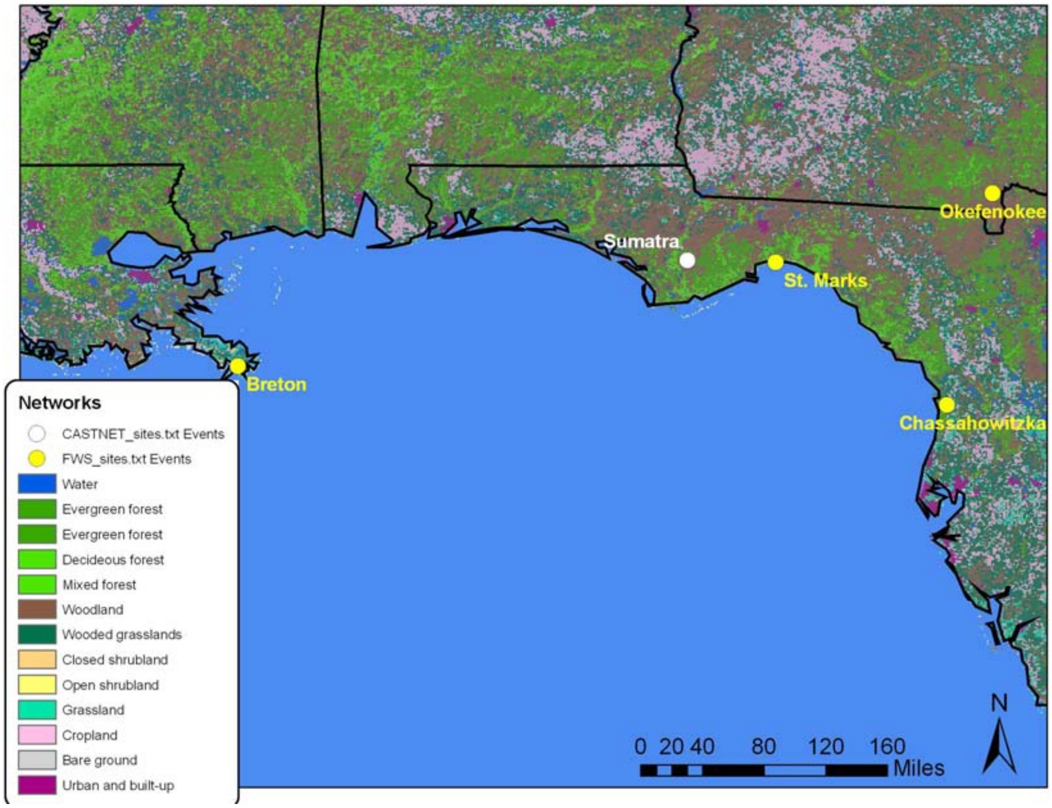


The total amount of nitrogen and sulfur deposited in Indian River Lagoon, ranged approximately from 0.2 kg/ha to 0.6 kg/ha and from 0.4 kg/ha up to 0.9 kg/ha, respectively. Dry deposition increased from 2001 to 2003, while a significant decrease is observed for sulfur in 2004.

B.9 Sumatra, Florida

Sumatra in Florida is used as a surrogate CASTNET site for three FWS sites, namely Breton, Louisiana, St. Marks in Florida and Okefenokee in Georgia. Figure 58 shows the locations of both sites.

Figure 58 Location of CASTNET Sumatra and FWS Breton, St.Marks and Okefenokee sites.



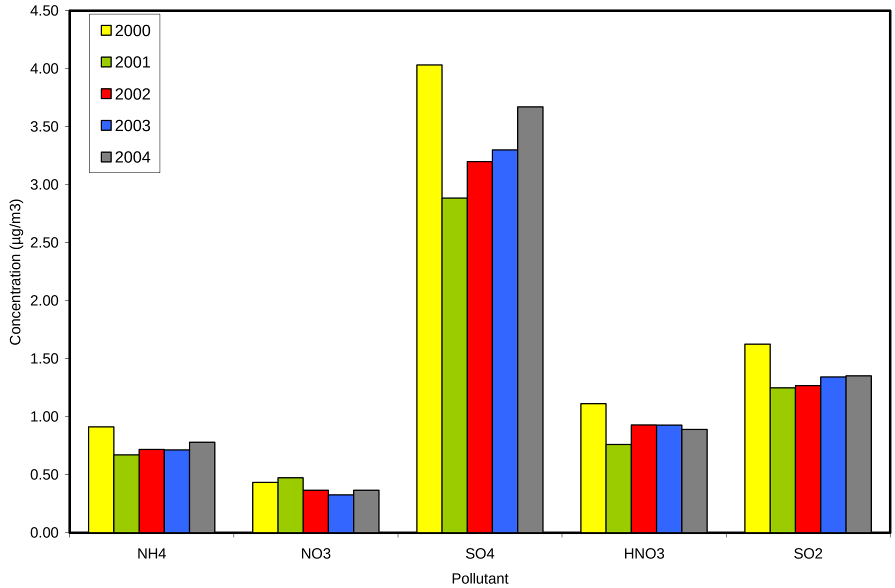
B.9.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Sumatra during the 2000-2004 period, are presented in Table 26.

Table 26 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Sumatra for 2000-2004 periods

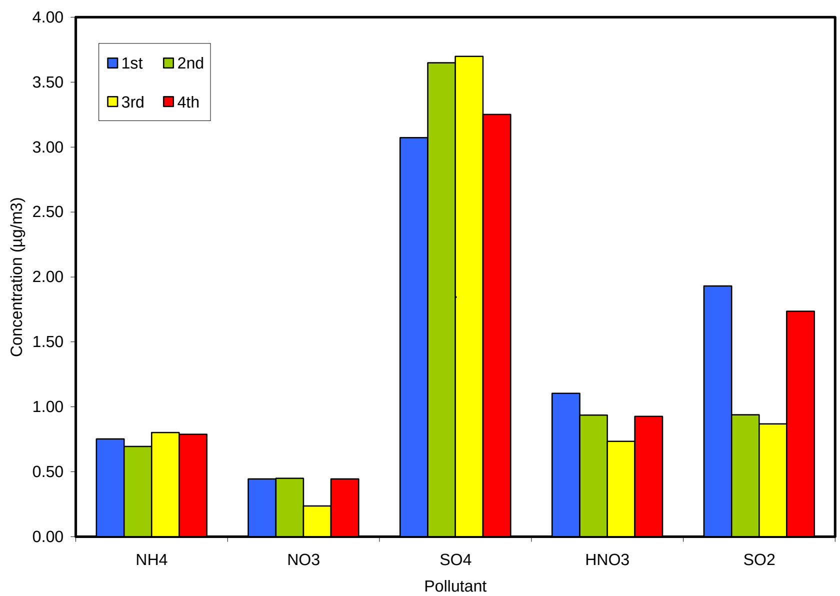
	Mean	**Minimum**	**Maximum**	**Std Deviation**
Aerosol				
Ammonium (NH_4^+)	0.8	0.1	2.9	0.4
Nitrate (NO_3^-)	0.4	0.1	1.5	0.3
Sulfate (SO_4^{2-})	3.4	0.5	11.8	1.7
Gas Phase				
Sulfur dioxide (SO_2)	1.4	0.1	4.3	0.8
Nitric Acid (HNO_3)	0.9	0.1	2.3	0.4

Figure 59 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 59 concentrations of all components followed an increasing trend from 2001 to 2004.

Figure 60 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2[nd] (April-June) and 3[rd] (July-September) for sulfate and ammonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher concentration being measured during winter

Figure 61 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

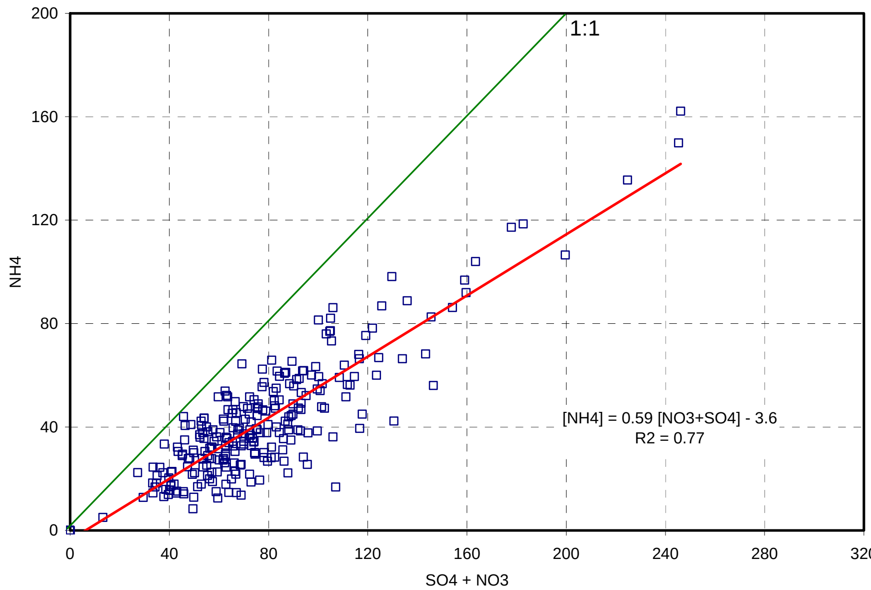


Figure 61 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 60% of nitrate and sulfate, which indicates that the predominant chemical components are H_2SO_4 HNO_3 and $(NH_4)HSO_4$, and minor amounts of NH_4NO_3 $(NH_4)_2SO_4$.

Table 27 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2000 to 2004. Neutralization by ammonia appears to be moderately significant throughout the year and for different seasons.

Table 27 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.60	0.49	0.45	0.65	0.66
Intercept	-4.3	3.5	7.0	-8.8	-10.9
R	0.90	0.78	0.72	0.90	0.92
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.52	0.54	0.62	0.64	
Intercept	4.3	-5.9	-5.8	-4.3	
R	0.67	0.80	0.97	0.90	

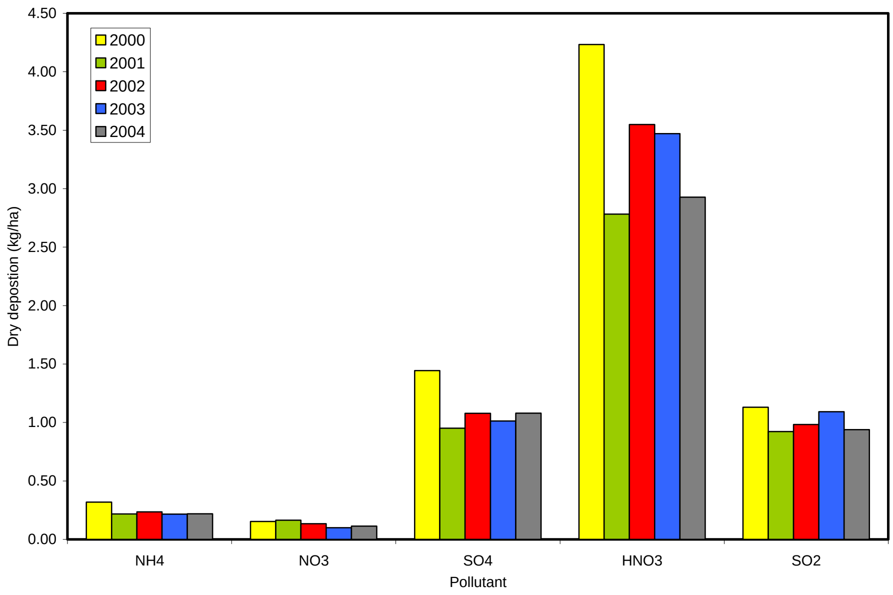
B.9.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Sumatra during the 2000-2004 period, are presented in Table 28.

Table 28 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Sumatra for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

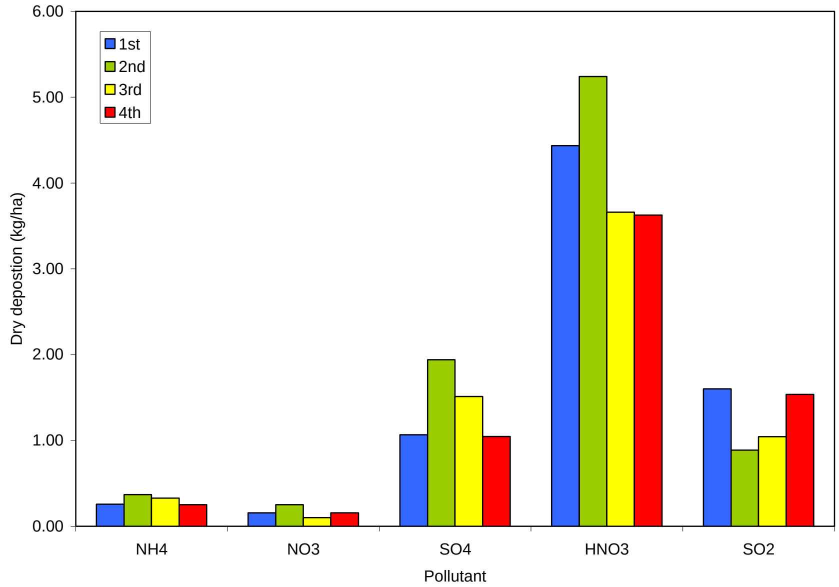
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	5	0	19	3	0.9
Nitrate (NO_3^-)	3	0	15	3	0.2
Sulfate (SO_4^{2-})	23	1	68	13	1.9
Gas Phase					
Sulfur dioxide (SO_2)	21	3	67	12	2.5
Nitric Acid (HNO_3)	69	4	202	36	5.7
Total Nitrogen					6.7
Total Sulfur					4.4

Figure 62 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



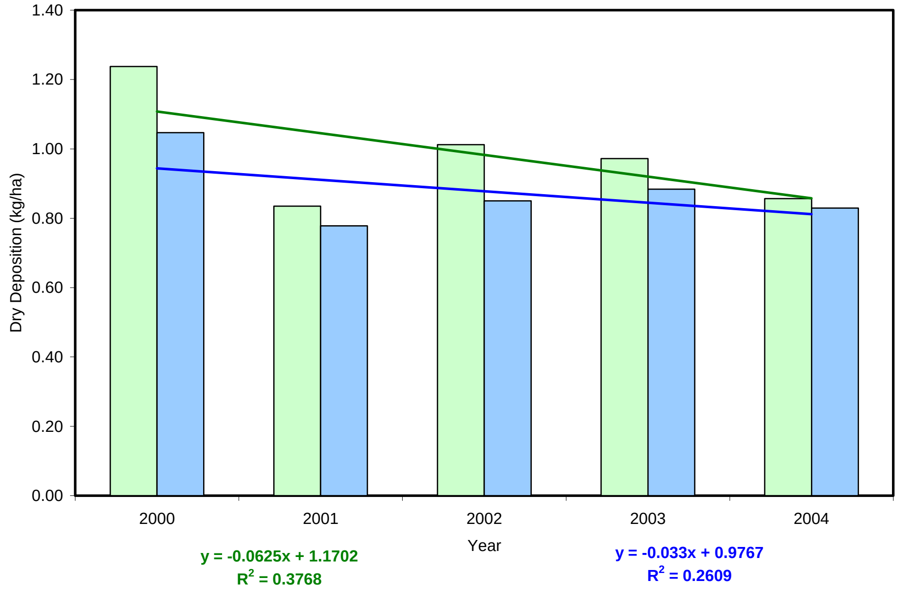
As shown in Figure 62, for all components, a rather downward trend was observed for 2001-2003 period followed by a slight decrease of dry deposition for 2004.

Figure 63 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, slightly higher deposition rates during the 2nd (April-June) and 3rd (July-September) for sulfate and ammonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher deposition being measured during winter

Figure 64 Total nitrogen and sulfur dry deposition at Sumatra during 2000-2004

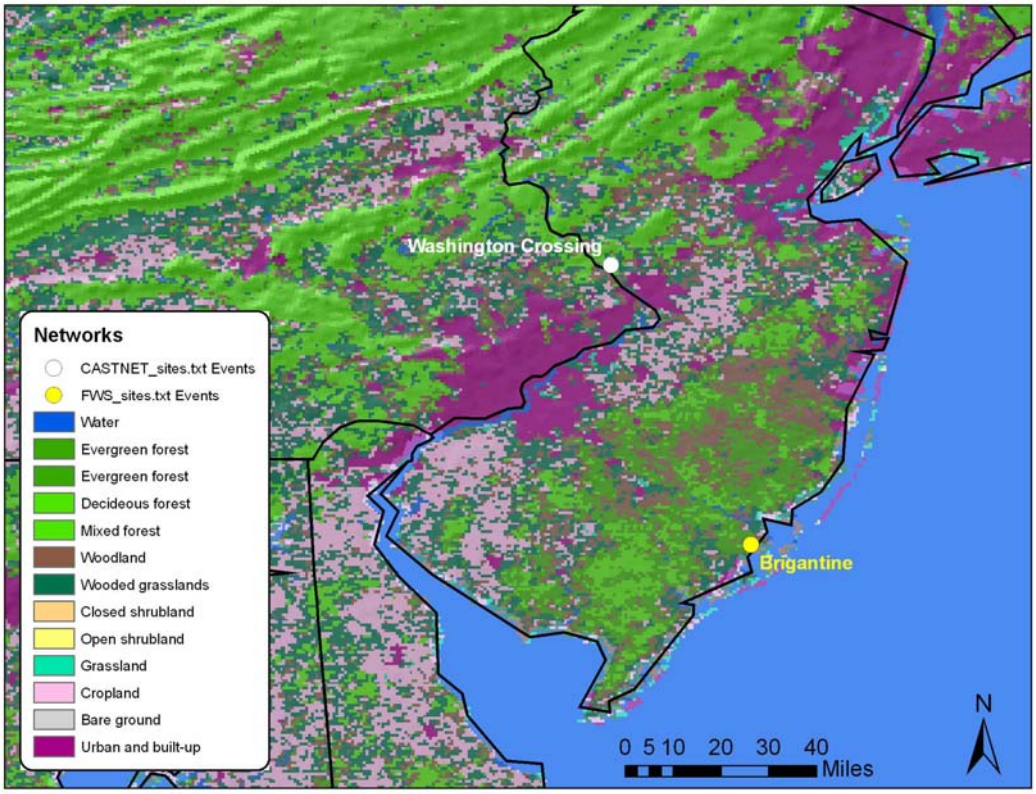


The total amount of nitrogen and sulfur deposited in Sumatra, ranged approximately from 0.9 kg/ha to 1.2 kg/ha and from 0.8 kg/ha up to 1.0 kg/ha, respectively. Dry deposition decreased from 2000 to 2003 for both elements,

B.10 Washington Crossing, New Jersey

Washington Crossing in New Jersey is used as a surrogate CASTNET site for FWS Brigantine New Jersey. Figure 65 shows the locations of both sites.

Figure 65 Location of CASTNET Sumatra and FWS Breton, St.Marks and Okefenokee sites.



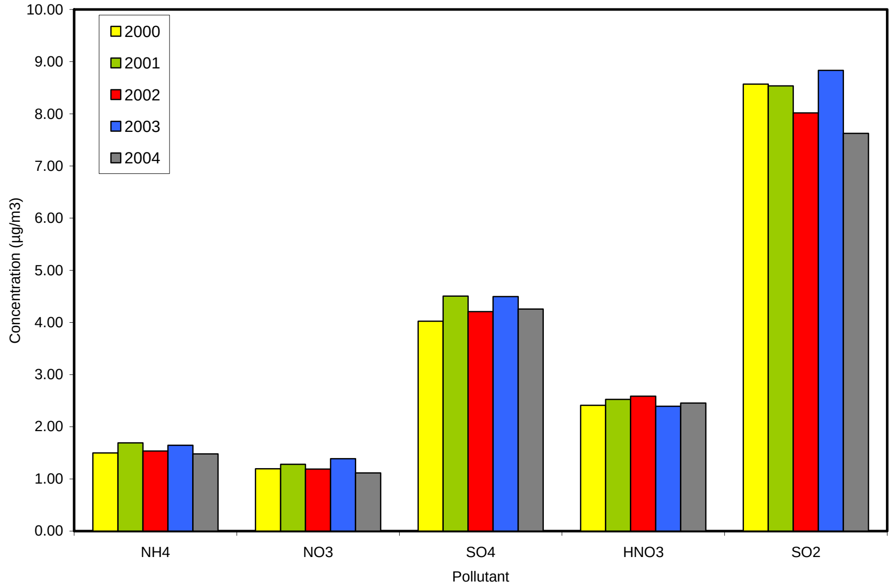
B.10.1 Ambient air

Ambient air concentration levels of particle-asociated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid for Washington Crossing during the 2000-2004 period, are presented in Table 29.

Table 29 Descriptive statistics (in µg/m^3) of atmospheric particulate ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Washington Crossing for 2000-2004 periods

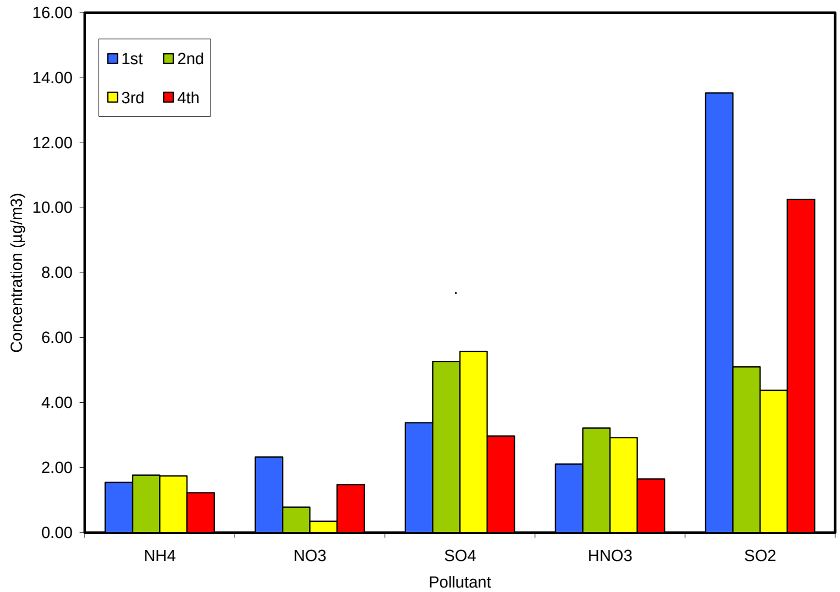
	Mean	Minimum	Maximum	Std Deviation
Aerosol				
Ammonium (NH_4^+)	1.6	0.3	4.6	0.7
Nitrate (NO_3^-)	1.2	0.1	5.9	1.1
Sulfate (SO_4^{2-})	4.3	0.9	13.4	2.2
Gas Phase				
Sulfur dioxide (SO_2)	8.3	1.4	31.1	5.4
Nitric Acid (HNO_3)	2.5	0.5	5.9	1.1

Figure 66 Mean annual concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



As shown in Figure 66 concentrations of all components fluctuated for different years with no clear trends for the monitoring period.

Figure 67 Mean quarterly concentrations of particulate ammonium (NH4), nitrate (NO3) and sulfate (SO4) and gaseous nitric acid (HNO3) and sulfur dioxide (SO2)



On a seasonal basis, slightly higher concentrations during the 2nd (April-June) and 3rd (July-September) for sulfate and ammonium are measured. An opposite profile is drawn for sulfur dioxide, nitric acid and particulate nitrate, with higher concentration being measured during winter

Figure 68 Scatter plot of NH4 nanoequivalents per m^3 vs. the sum of NO_3 and SO_4 nano-equivalents per m^3

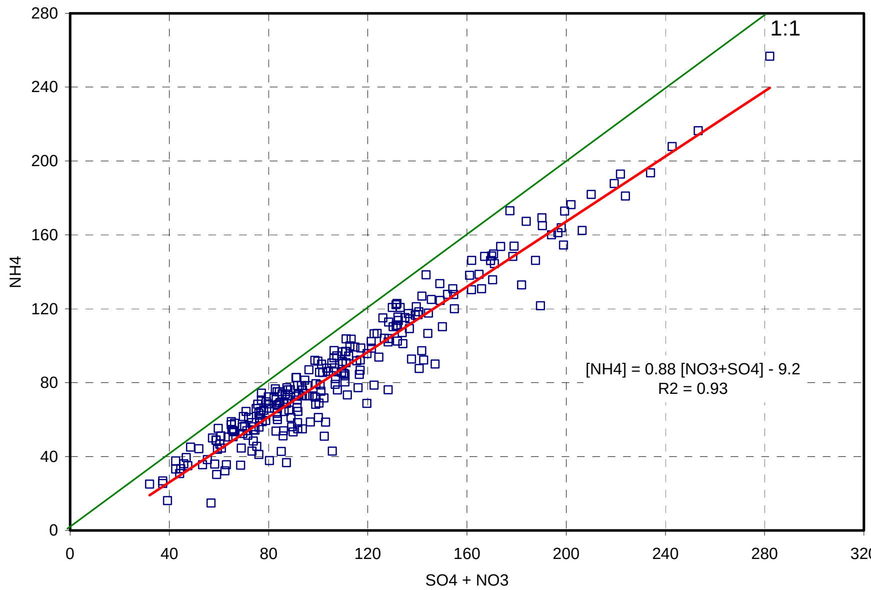


Figure 61 shows the relationship between the sum of sulfate and nitrate vs. ammonium (in n-equivalents/m^3). The green line indicates that both nitrate and sulfate are fully neutralized by ammonium in the forms of ammonium nitrate and ammonium sulfate. The slope of the regression indicates that ammonium concentrations are able to neutralize approximately 88% of nitrate and sulfate, which indicates that the predominant chemical components are $(NH_4)HSO_4$, NH_4NO_3 $(NH_4)_2SO_4$ and minor amounts of. H_2SO_4 and HNO_3

Table 30 shows the inter-annual and seasonal variation of the relationships between ammonium, and nitrate and sulfate. No significant variability is computed from 2000 to 2004. Neutralization by ammonia appears to be significant throughout the year and for different seasons.

Table 30 Annual and quarterly variation of regression coefficients of atmospheric NH4 (as dependent variable) and the sum of NO3 and SO4 (as independent variable).

Year	**2000**	**2001**	**2002**	**2003**	**2004**
Slope	0.85	0.86	0.84	0.96	0.89
Intercept	-4.8	-4.3	-4.2	-19.9	-12.8
R	0.96	0.98	0.96	0.96	0.97
Quarter	**1st**	**2nd**	**3rd**	**4th**	
Slope	0.88	0.92	0.90	0.82	
Intercept	-8.8	-14.2	-13.1	-2.4	
R	0.94	0.6	0.98	0.94	

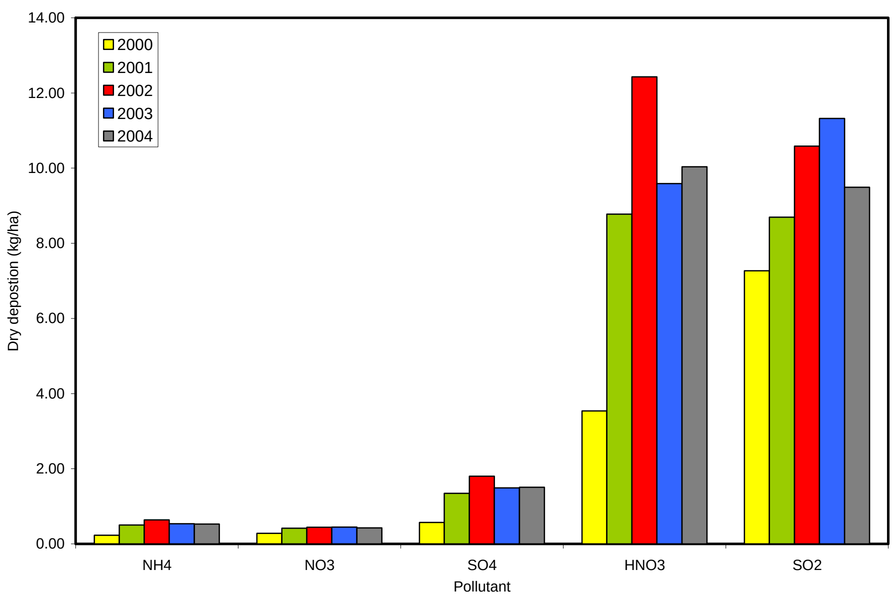
B.10.2 Dry Deposition

Deposition rates and total deposition of particle-associated ammonium, nitrate, sulfate and gas-phase sulfur dioxide and nitric acid, and total nitrogen and sulfur for Washington Crossing during the 2000-2004 period, are presented in Table 31.

Table 31 Descriptive statistics (g/ha) of dry deposition of ammonium, nitrate, sulfate and, sulfur dioxide and nitric acid in Washington Crossing for 2000-2004 periods and total nitrogen and sulfur deposition (kg/ha) for the entire period

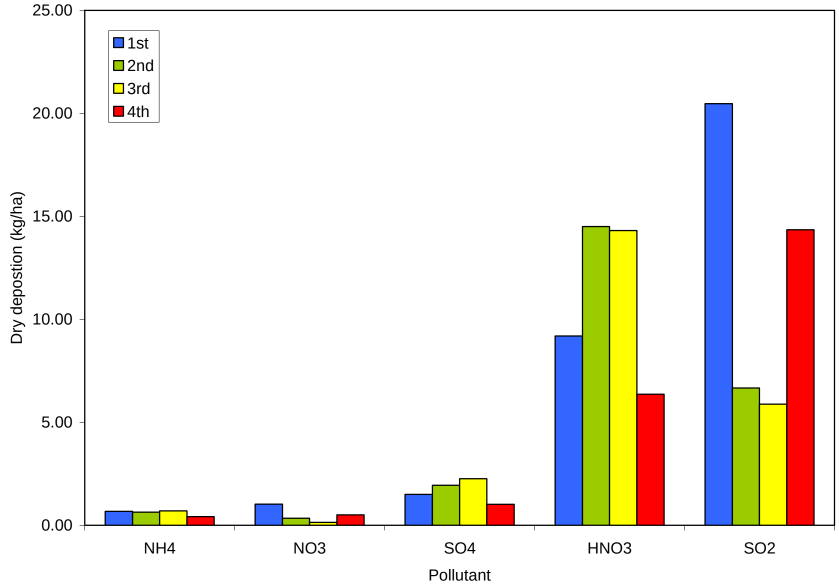
	Mean	Minimum	Maximum	Std Deviation	Total deposition
Aerosol					
Ammonium (NH_4^+)	11	2	37	6	1.9
Nitrate (NO_3^-)	9	0	41	8	0.5
Sulfate (SO_4^{2-})	31	4	112	19	2.2
Gas Phase					
Sulfur dioxide (SO_2)	215	29	708	134	23.7
Nitric Acid (HNO_3)	202	26	591	118	14.8
Total Nitrogen					17.1
Total Sulfur					25.9

Figure 69 Annual dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



As shown in Figure 69, for all components, a strong increasing trend is observed for 2001-2003 period followed by a slight decrease of dry deposition for 2004.

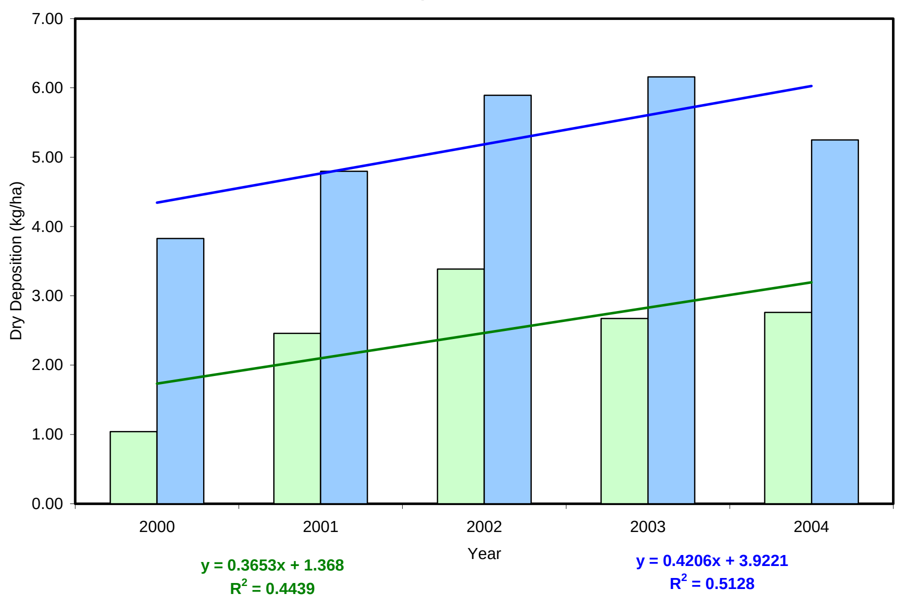
Figure 70 Quarterly dry deposition of ammonium (NH4), nitrate (NO3), sulfate (SO4), nitric acid (HNO3) and sulfur dioxide (SO2).



On a seasonal basis, slightly higher deposition rates during the 2nd (April-June) and 3rd (July-September) for sulfate and ammonium and nitric acid are measured. An opposite profile is drawn for sulfur dioxide and particulate nitrate, with higher deposition being measured during winter

Figure 71 Total nitrogen and sulfur dry deposition at Washington Crosiing during 2000-2004

Figure 72



The total amount of nitrogen and sulfur deposited in Washington Crossing, ranged approximately from 1.0 kg/ha to 3.4 kg/ha and from 3.8 kg/ha up to 6.2 kg/ha, respectively. Dry deposition decreased from 2000 to 2004 for both elements,