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Annual wet and dry deposition of sulfur and nitrogen in the snowy range, Wyoming

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Abstract

The collocation of three national networked programs NADP, EPA's CASTNET, and the Forest Service's IMPROVE Module A, within a few hundred meters of each other in the pristine Medicine Bow forest of Wyoming has made it possible to assess the total amount of sulfur and nitrogen deposition, both wet and dry for this alpine/subalpine ecosystem. Additional sites within a few kilometers add spatial depth to this study. Wet deposition assessed using NADP data accounts for 1 to a little over 3 kg ha^{-1} yr^{-1} for both nitrogen and sulfur; however, annual trends for the two species differ. Dry deposition assessed using both CASTNET (a.k.a. NDDN) and IMPROVE (for sulfur) indicates 1–2 kg ha^{-1} yr^{-1} for nitrogen but less than 1 kg ha^{-1} yr^{-1} for sulfur. The overall trend of wet plus dry for nitrogen has been downward from 5 kg ha^{-1} yr^{-1} in 1989 to 3.6 kg ha^{-1} yr^{-1} in 1994, while varying between 2 and under 4 kg ha^{-1} yr^{-1} for sulfur. This paper introduces the sites and presents the three programs and the analysis approach. Spatial comparisons between sites are investigated. Weekly data are analyzed from three NADP sites, separated horizontally 6.8 and 2.4 km and vertically 430 and 98 m from the highest elevation site. The site comparisons demonstrate that winter season data requires careful analysis due to the vagaries of inefficient precipitation collection during high winds and snow fall.

Keywords: Nitrogen deposition; Sulfur deposition; Wet deposition; Dry deposition; Atmospheric deposition

1. Introduction

This paper summarizes data specific to nitrogen and sulfur deposition collected at three national program site: (1) the Environmental Protection Agency (EPA) Clean Air Status and Trends Network (CASTNET) (a.k.a. National Dry Deposition Network: NDDN) Brooklyn Lakes site (http://www.epa.gov/acidrain/castnet/); (2) the National Park Service (NPS) – United States Dept. of Agriculture Forest Service (USDA FS) Interagency Monitoring of Protected Visual Environments (IMPROVE) Module A (predecessor program: SFU: stacked filter unit) (http://crocker.ucdavis.edu/CNL/RESEARCH/AQG4.htm); and (3) the National Atmospheric Deposition Program National Trends Network (NADP/NTN) (http://nadp.sws.uiuc.edu/) programs conducted at the USDA FS Glacier Lakes Ecosystem Experiment Site (GLEES) in the Snowy Range mountains, WY.

Determining atmospheric deposition, particularly dry deposition, is complicated by the inability to measure dry deposition directly (Hicks et al., 1991) and by the variety of measurement protocols used. Wet deposition is also problematic (Erisman et al., 1994); however, the adoption of NADP/NTN protocols for most monitoring programs within the US has helped to simplify data analysis. Three closely located NADP sites are compared to address

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spatial representativeness. Spatial representativeness for dry deposition is partially addressed by comparing the sulfur concentration results from two NDDN locations with IMPROVE results. Although IMPROVE program protocols were designed to address visibility and not deposition, the similarity in species monitored makes it useful for this study.

2. Methods

2.1. Sites

The data presented here are taken from five sites in Wyoming within or in close proximity of the USDA Forest Service's GLEES area in the Snowy Range of the Medicine Bow National Forest, WY about 65 km west of Laramie, WY. The GLEES complex is described by Musselman (1994). The major tree species in the GLEES forest are *Picea engelmannii* (48%), *Abies lasiocarpa* (48%), and *Pinus contorta* (4%) The Snowy Range (SR) NADP site, (WY00), is located southwest of West Glacier Lake in Snowy Range of Wyoming at an elevation of 3286 m which is also the approximate tree line elevation for the forest. Samples have been collected at this site from 1986 to the present. The second NADP site, Nash Fork (NF), (WY96), is located 6.8 km southeast of the SR site at an elevation of 2856 m. Samples were collected at Nash Fork from 1987 to September, 1992. In September 1992, this site was relocated and given a new name and calcode. The relocated NADP site, Brooklyn Lake, (BL), (WY95), is 2.4 km southeast of the SR site at an elevation of 3188 m. Samples have been collected at this site from Sept. 1992 to the present.

GLEES area CASTNET NDDN monitoring commenced in July 89 at an open dry meadow site near Centennial, WY (CNT169) 12.2 km southeast of GLEES. During August 1991 CNT169 was relocated to GLEES in an open-sloped dry meadow location 138 m west-southwest of the Brooklyn Lake tower IMPROVE site and approximately 50 m south–southeast of the Brooklyn Lake NADP site (WY95). The USDA IMPROVE site, located about 130 m east of BL NADP WY95, was not moved during the period of this study. Table 1 gives the location and relative horizontal distance from the Snowy Range NADP site (for a site map see Musselman, 1994). The three 'Brooklyn' sites are all within 140 m horizontal distance of each other.

2.2. Wet deposition

NADP was established in 1978 as a long-term atmospheric deposition monitoring network (CSU, 1991; Sisterson, 1991) Today the program has approximately 200 rural stations in the United States, which monitor wet deposition. The main goal of the program is to determine the spatial patterns and temporal trends in chemical deposition to support studies of the impact of chemical deposition on aquatic and terrestrial ecosystems. Weekly precipitation samples are collected in a 'wet only' NADP bucket (i.e. only open during precipitation events) from each site and sent to the Central Analytical Laboratory (CAL) for analysis. A weighing 'rain' gage is also used in the NADP protocol at each site to accurately determine the weekly precipitation amount. The samples are analyzed for SO_4^{2-}, NO_3^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^{2+}, NH_4^+, H^+, pH, and conductivity.

The site comparisons, limited to SO_4^{2-}, NO_3^- and NH_4^+ in this study, were completed for each year (1987–1994) using the weekly data at each site. In all cases, the comparisons are done relative to the Snowy Range site. Although measured NADP/NTN does not report weekly deposition, it reports rather seasonal and yearly depositions. The validity of the weekly samples, however, is determined by NADP/NTN and a sample may be considered invalid for a number of reasons, including contamination, short or long sampling period (< 6 days or > 8 days), and lab error. Large precipitation differences between the study sites greatly effected the site comparisons between concentration and deposition values. Hence a 'reduced' or filtered dataset, established by removing the data whenever the precipitation

Table 1
Monitoring sites, program and location

Name	Program	Elevation	Latitude	Longitude	Distance from SR
SR: Snowy Range (WY00)	NADP	3286 m	41°22′34″	106°15′34″	0.0 km
NF: Nash Fork (WY96)	NADP	2856	41°20′25″	106°11′27″	6.8
BL: Brooklyn Lake (WY95)	NADP	3188	41°21′53″	106°14′27″	2.4
CNT-BL 169: Brooklyn Lake	CASTNET (NDDN)	3180	41°21′50″	106°14′27″	2.5
CNT169: Centennial	CASTNET (NDDN)	2591	41°18′30″	106°09′10″	12.2
BT: Brooklyn Tower	IMPROVE	3186	41°21′53″	106°14′15″	2.6

Table 2
Number of weekly data points in each dataset

Year	Comparison sites	Valid dataset	Reduced dataset
1987	SR-NF	31	24
1988	SR-NF	22	17
1989	SR-NF	34	25
1990	SR-NF	27	22
1991	SR-NF	41	29
1992a	SR-NF	28	23
1992b	SR-BL	3	3
1993	SR-BL	31	30
1994	SR-BL	27	24

at a site was either less than 3 mm or whenever the precipitation difference, P_d, between two sites was greater than 100%, was prepared for this study comparison.

$$P_d = 100\left(\frac{P_{SR} - P_i}{P_{SR}}\right), \quad (1)$$

where P_{SR} is the precipitation measured at SR, and P_i is the precipitation measured at either NF or BL. Table 2 shows the number of resulting data points used for each year for the valid dataset and the reduced dataset.

The weekly deposition values for this study were calculated using the weekly concentration and precipitation values reported by NADP:

$$D_i = C_i \frac{P_i}{100}, \quad (2)$$

where D_i is the deposition of sample i in kg ha^{-1} wk^{-1}, C_i is the concentration of sample i in mg l^{-1}, and P_i is the precipitation amount of sample i in mm. Extensive regression analyses were made between the SR and other two sites by grouping the weekly data into separate years and seasons as well as for the whole data set. The regression results are briefly discussed below; however summary site comparisons for this paper are presented as difference plots. The differences in the concentration and deposition between two sites ((SR-NF); and (SR-BL)) were calculated using the reduced weekly dataset. The resulting differences were analyzed in two ways: sorting the data by year and sorting the data by month. Using the data sorted by year or month, the average differences and standard deviations were also calculated for each species and each year or month.

2.3. Dry deposition

2.3.1. EPA CASTNET (NDDN)

NDDN was established by EPA to obtain routine weekly ambient concentrations and dry deposition measurements of O_3, SO_4^{2-}, NO_3^-, NH_4^+, SO_2, and HNO_3 and meteorological parameters. Weekly filter pack samples are exchanged on Tuesday mornings in coordination with NADP protocol. NDDN was expanded to incorporate similar programs and is now called CASTNET although the NDDN sites are still named so. The dry deposition component of NDDN is the three-stage serial filter pack: first a 47 mm Teflon filter (Zefluor, 1 μm) for SO_4^{2-} and NO_3^- aerosol; second a 47 mm nylon filter (Nylasorb, 1 μm) for HNO_3 and third a K_2CO_3-impregnated cellulose (Whatman no. 41) filter for SO_2. Ambient air from 10 m height is continuously drawn through the filter pack at 1.5 l min^{-1}. Field and laboratory procedures are provided by ESE (1990a, b).

Dry deposition is a two step analysis: step 1 measure the ambient concentration, c, of the pollutant species of interest; step 2 models the applicable deposition velocity

Table 3
Annual wet deposition (kg ha^{-1} yr^{-1}) and SR precipitation (cm) as measured by NADP

Year	SR NH_4^+	NF NH_4^+	SR NO_3^-	NF NO_3^-	SR SO_4^{2-}	NF SO_4^{2-}	SR precip. (cm)
1986	0.38	0.00	3.35	0.06	3.46	0.09	65
1987	0.78	0.65	5.48	4.28	5.24	3.73	87
1988	0.57	0.36	6.08	3.25	8.65	4.28	119
1989	1.93	0.73	9.73	4.48	9.72	4.00	127
1990	1.56	0.90	9.95	4.90	6.39	3.35	130
1991	1.14	0.74	8.38	4.57	6.40	3.40	125
1992[a]	1.69	0.71	9.43	3.30	6.79	2.62	108
		BL NH_4^+		BL NO_3^-		BL SO_4^{2-}	
1993	1.48	1.18	8.44	8.02	8.41	6.77	142
1994	1.14	0.77	5.94	5.06	4.58	3.5	83

[a]Combined NF and BL locations.

(Bytnerowicz et al., 1987; Schmel, 1984; Wyers and Duyzer, 1997), V_d, and multiplies it by c to determine deposition, F_c, (or vertical flux) Eq. (3) (Zeller and Hehn, 1996)

$$F_c = cV_d. \tag{3}$$

The deposition velocity, provided within the CASTNET protocol is a multiple function of chemical species, atmospheric turbulence, vegetation, canopy structure, etc.

2.3.2. IMPROVE

The IMPROVE measurement program was designed and implemented to address visibility (background, visibility impairment species and trends) for federal land management and regulatory agencies (Malm et al., 1994; Sisler, 1996). Basically an IMPROVE site can have up to four separate modules operated in parallel: module A, a Teflon filter, for fine particulate ($\leqslant 2.5$ μm diameter) mass; module B, a denuded nylon filter, for sulfate (SO_4^{2-}) and nitrate (NO_3^-) ($\leqslant 2.5$ μm); module C, a quartz filter, for organic and light absorbing carbon (C) ($\leqslant 2.5$ μm); and module D for particulate mass $\leqslant 10.0$ μm diameter. Sample measurements are made through 24-h continuous filtering on Wednesday and Saturday of each week. The 2.5 μm cutoff is achieved using a size selective inlet in addition to a cyclone separator. Sample flow rate is held at 22.7 l min^{-1} using a critical orifice. The IMPROVE program was established in 1992 and replaced the earlier stacked filter unit (SFU) protocol originally fielded in 1979. At sites where the fine particulate module A replaced the SFU, fine mass and sulfur monitoring records are considered closely comparable to IMPROVE and are analyzed as single records. At GLEES, SFU measurements commenced at the Brooklyn Lake tower site, a 30 m diameter opening in the GLEES forest (Zeller and Hehn, 1996) (BRLA = BL) on 01 Mar 91 and ended 31 July 93; IMPROVE module a measurements commenced 31 July 93 to the present time. For this study the sulfur analysis results of Module A was used to estimate sulfate concentration by assuming that all of the collected sulfur was sulfate.

Table 4
Coefficients of determination (r^2) for seasonal (and annual) data between NADP sites

Species	1987 to Sept. 1992 (SR-NF) concentration	deposition	Sept. 1992 to 1994 (SR-BL) concentration	deposition
NH_4^+	0.66 (0.76)	0.28 (0.63)	0.36	0.69
NO_3^-	0.71 (0.83)	0.09 (0.57)	0.23	0.21
SO_4^{2-}	0.70 (0.70)	0.20 (0.60)	0.21	0.53

The comparison results given below were obtained by assuming that the IMPROVE concentration average of the Wednesday 24 h sample and the Saturday 24 h sample would be the closest quantity to the 7 day (168 h) concentration average measured by the NDDN sampler within the same sampled week. Neither a deposition velocity analysis nor sulfate deposition analysis was made for the IMPROVE site.

3. Results and discussion

3.1. Wet deposition

Table 3 gives the annual wet deposition amounts for SO_4^{2-}, NO_3^- and NH_4^+ and the SR annual precipitation. Note that the values for 1992 are the combined NF (Jan. to Sept.) and BL (Sept. to Dec.). The SO_4^{2-}, NO_3^- and NH_4^+ values in Table 3, are low compared to values measured in the northeast US where SO_4^{2-} exceeded

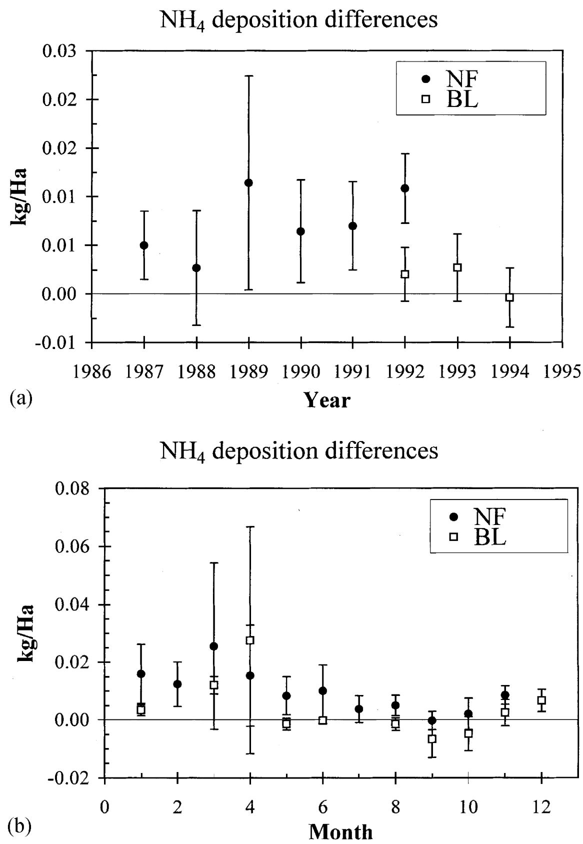

Fig. 1. The average and standard deviation of weekly wet NH_4^+ deposition difference between Snowy Range (SR) and Nash Fork (NF) or Brooklyn Lake (BL) NADP sites: (a) by year; and (b) by month.

40 kg ha^{-1} yr^{-1}, NO_3^- 25 kg ha^{-1} yr^{-1} and NH_4^+ 5 kg ha^{-1} yr^{-1} (Irving, 1991). There is no corrrelation between deposition and precipitation.

For each year and species, the NF and BL weekly, seasonal and annual data were linearly regressed against the SR data. Linear trendlines (slopes and offset) were calculated including coefficients of determination (correlation squared) r^2 values. Correlation, slope and offset results varied widely from species to species and from year to year as well as within the same species between years. Correlation values improved when the weekly data were consolidated from annual weekly to yearly winter averages, yearly summer averages, and yearly averages. As an example, Table 4 gives the r^2 values for the combined multi-year seasonal data for both site comparisons. The annual total comparisons (seven data points) are also included in Table 4 for SR vs. NF only as there were only two annual points for SR vs. BL. In the case of Table 4, r^2 values are higher for NF concentrations than for NF deposition and vice versa for BL. Also r^2 values are higher for NF annual comparisons. We expected higher r^2 values at BL for both concentration and deposition because it is closer to SR; however, we suspect that the smaller data sample (11 seasons for NF; 5 seasons for BL) is affecting the SR vs. BL r^2 values.

The site comparison also demonstrated two distinct seasons: winter and summer defined by the averaged precipitation type: winter period – snow; summer period – rain. The summer season extended from 1 May through 30 Sept. (although complete snow pack melt above 3150 m does not typical occur until late June), while the winter season extends from 1 Oct. through 30 April. Precipitation amounts measured at NF were typically 70% of SR during the summer and dropped to 50% during winter and at BL they were 90% year round compared to SR. Since wet deposition measurements are precipitation quantity dependent, they account for the weaker deposition correlations as compared to concentration. Measured concentrations were often higher at the lower sites but deposition is always higher at SR compared to NF.

The average weekly difference between sites and their standard deviations are shown in Figs. 1–3 as functions of year (a: values plotted at end of year) and month (b). (Note that Figs. 1–3 are weekly difference statistics while

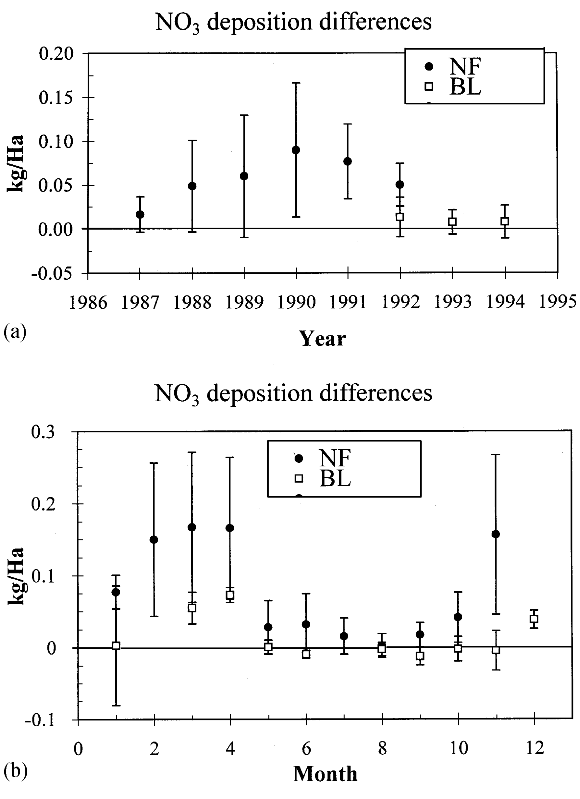

Fig. 2. The average and standard deviation of weekly wet NO_3^- deposition difference between Snowy Range (SR) and Nash Fork (NF) or Brooklyn Lake (BL) NADP sites: (a) by year; and (b) by month.

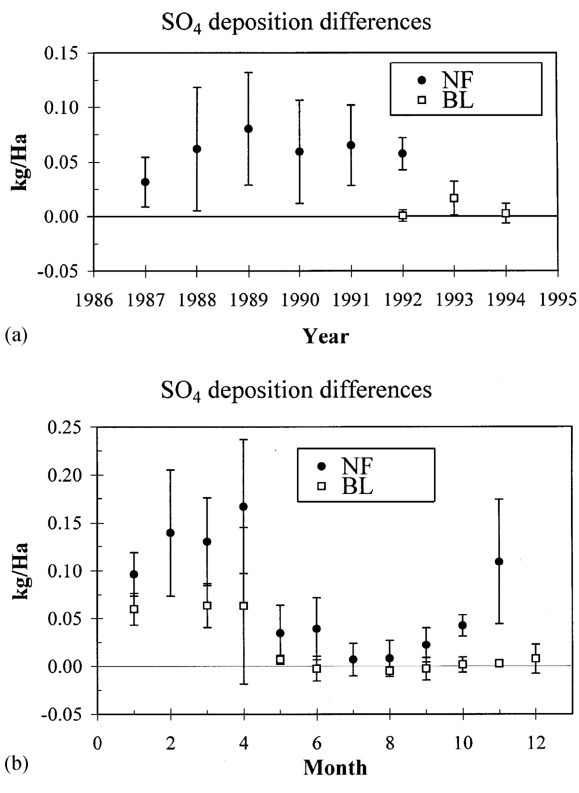

Fig. 3. The average and standard deviation of weekly wet SO_4^{2-} deposition difference between Snowy Range (SR) and Nash Fork (NF) or Brooklyn Lake (BL) NADP sites: (a) by year; and (b) by month.

Table 3 provides total annual deposition from which total annual differences might be calculated.) Figs. 1a, 2a and 3a consistently demonstrate that deposition is higher at SR, the highest elevation site, and that the difference is greater for NF compared to BL. Also the two-season phenomena, attributed to precipitation type, is clearly seen for each species in Figs. 1b, 2b and 3b. Standard deviations of the differences are also substantially smaller during the summer season. Although there is high weekly variance, the average annual deposition at SR for all three species is about 1% greater than the total annual depositions at the other two sites.

3.2. Dry deposition

As discussed above, Eq. (3), the determination of dry deposition is a measurement plus modeling exercise. Fig. 4 is a scatter plot of the weekly sulfate concentration data measured by IMPROVE (average of Wed. + Sat.) and by NDDN, for 93 Sep.–95 Sep. The coefficient of determination in Fig. 4, 0.73, is very good considering that IMPROVE was originally designed to measure visibility parameters and that it samples only 2/7ths of the period sampled by NDDN. Coefficient of determinatfion for the SFU comparison with NDDN at BL from 91 Aug. to 93 Sept. was 0.63. The linear regression slopes in both comparisons were 0.63, indicating that relative to the GLEES area, IMPROVE was able to determine about 63% of the sulfate measured by NDDN 138 m distance. The r^2 value for the period, 89 Oct.–91 Aug. when NDDN was located in Centennial, WY, (15 km southeast of BT) was 0.37 and the regression slope was 0.54. The IMPROVE measurements were made in a forested opening while the NDDN measurements were always made in open dry meadows; hence, it is possible that if the IMPROVE measurements were also taken in the open the

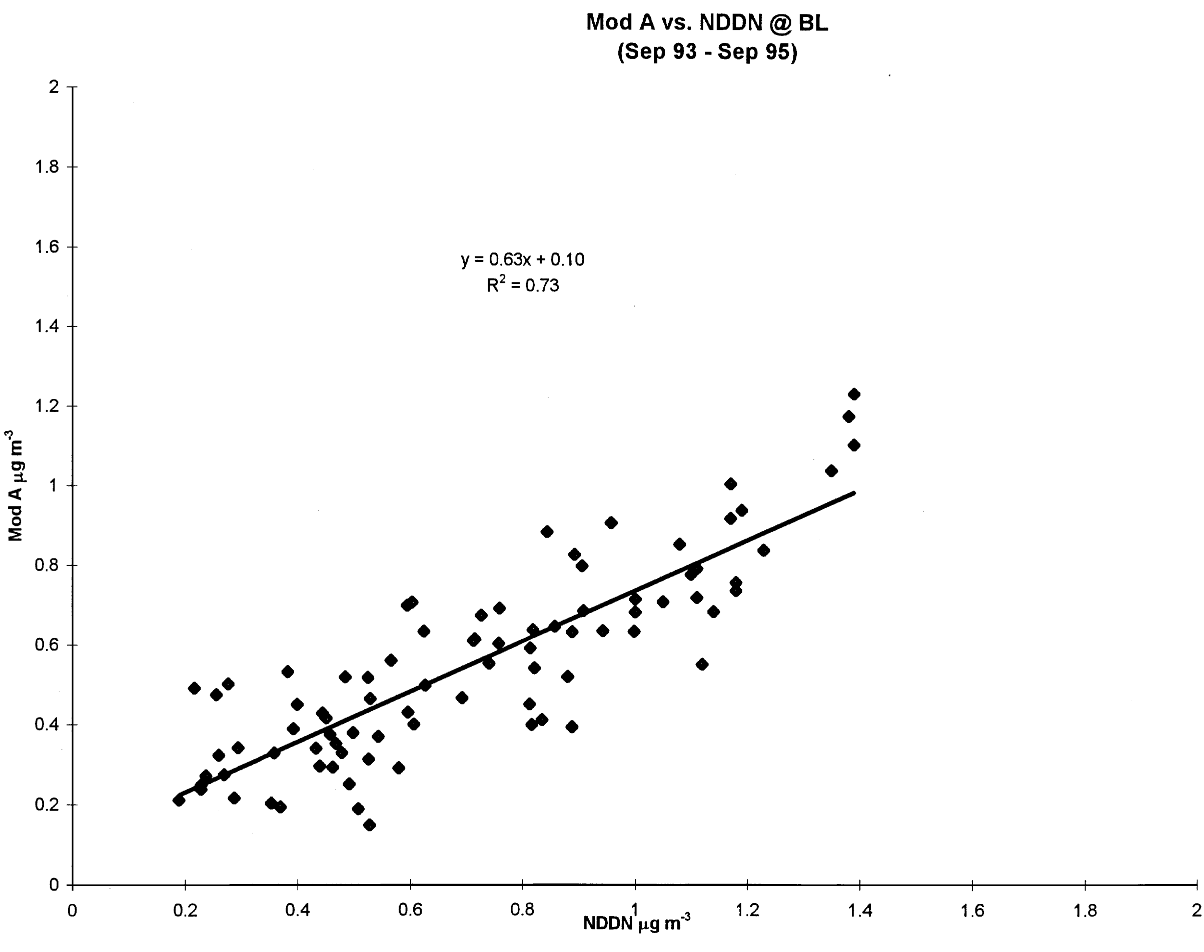

Fig. 4. Scatter diagram of IMPROVE (Module A weekly value estimated from 2 to 24 h samples) SO_4^{2-} concentration vs. NDDN SO_4^{-2} concentration.

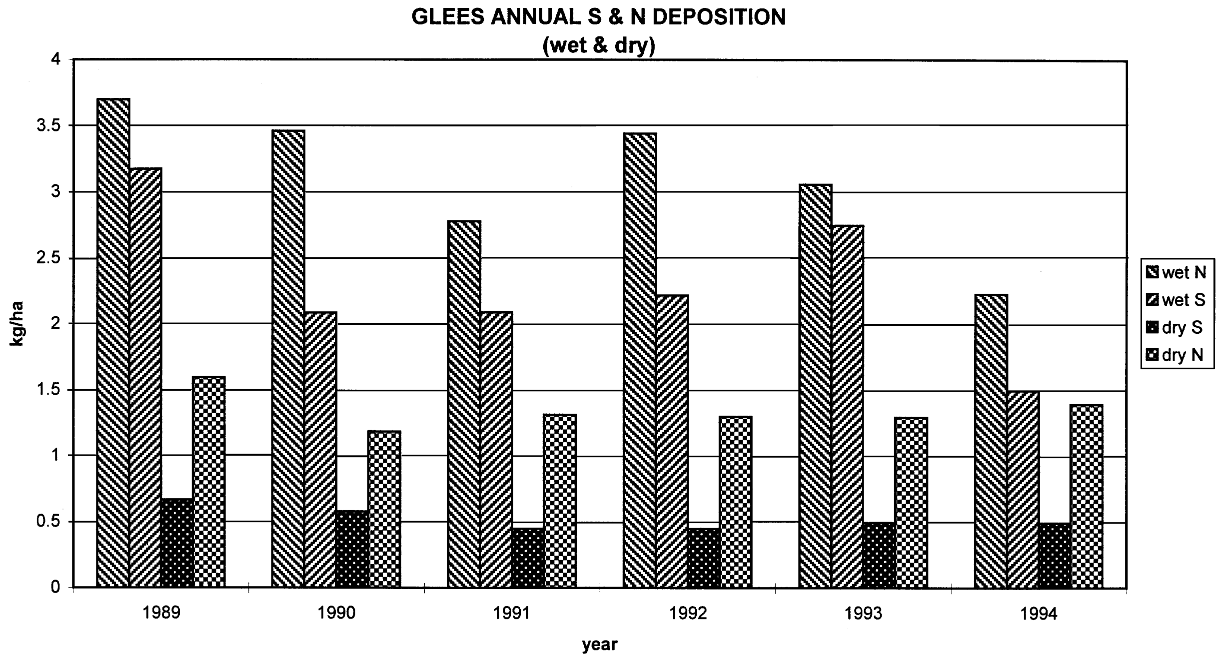

Fig. 5. Total annual wet and dry nitrogen and sulfur deposition at GLEE for 1989–1994.

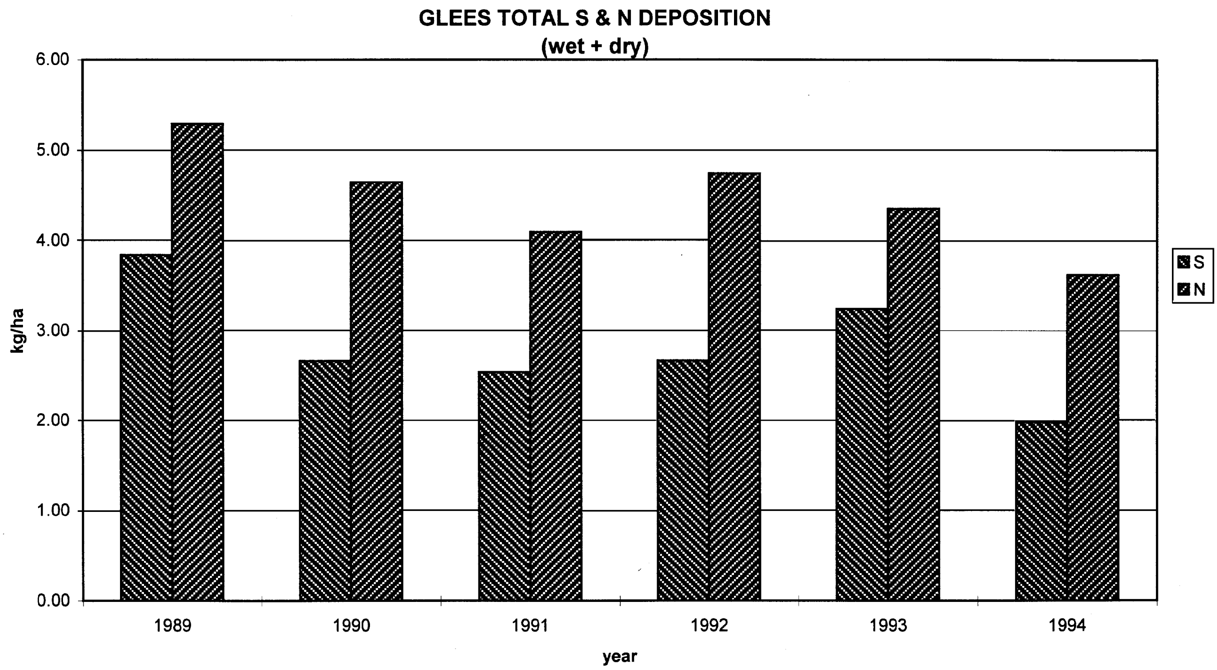

Fig. 6. Total wet plus dry nitrogen and sulfur deposition at GLEE for 1989–1994.

regression slope may become larger than 0.63. Table 5 gives the annual dry deposition results in kg ha^{-1} yr^{-1} of using the NDDN data. HNO_3, accounts for the greatest contribution to dry nitrogen deposition. The annual dry deposition does not appear to vary much from year to year, nor between the two sites. Unfortunately simultaneous measurements were not possible. Compared to Table 5 dry deposition values for NO_3

Table 5
Annual dry deposition (kg ha^{-1} yr^{-1})

Year	SO_4^{2-}	SO_2	NH_4^+	NO_3^-	HNO_3
Centennial:					
1989	0.63	0.92	0.26	0.14	6.10
1990	0.71	0.69	0.19	0.13	4.51
1991[a]	0.56	0.53	0.22	0.14	4.98
Brooklyn:					
1992	0.64	0.47	0.22	0.13	4.93
1993	0.57	0.62	0.19	0.10	5.06
1994	0.59	0.59	0.19	0.14	5.45
1995	0.47	0.53	0.19	0.10	5.76
1996	0.65	0.55	0.24	0.19	6.54

[a]Combined Centennial, WY and BL locations.

and SO_4 in Eastern Europe (e.g. 0.3–1.2 and 1.5–3.2 kg ha^{-1} yr^{-1}) are higher (Ruijgrok et al., 1995; Zeller et al., 1997a,b) as is also true for the Eastern US (CSU, 1992).

3.3. *Total sulfur and nitrogen deposition*

Fig. 5 shows the annual wet and dry sulfur and the annual wet and dry nitrogen deposition at the GLEES area for the years, 1989–1994, for which complete data sets were available. Fig. 6 shows the total wet plus dry sulfur and the total wet plus dry nitrogen deposition. Wet deposited nitrogen and sulfur were calculated from SO_4^{2-}, NO_3^- and NH_4^+ values at SR based on molecular weight. Dry deposited nitrogen and dry deposited sulfur were calculated from the CNT169-BL SO_4^{2-}, NO_3^-, SO_2, NH_4^+ and HNO_3 values based on molecular weight. Although different sites were used for wet and for dry deposition, based on the small average annual differences shown in Figs. 1, 2 and 3, and the similarity in concentrations at separate dry sites in Table 5, the authors claim that Fig. 6 presents a reasonable characterization of the nitrogen and sulfur deposition in the surrounding GLEES subalpine area.

4. Conclusion

Deposition results from separate sites within the Medicine Bow forest of Wyoming have been analyzed for spatial representativeness and to assess total annual atmospheric nitrogen and sulfur deposition. The results indicate that both wet and dry deposition at nearby sites are not comparable on a weekly basis; however, on an annual basis results are similar and show higher deposition at higher elevations. Wet deposition assessed using NADP data accounts for 1 to a little over 3 kg ha^{-1} yr^{-1} for both nitrogen and sulfur; however, annual trends for the two species differ. Dry deposition assessed using CASTNET (NDDN) demonstrates 1 to 2 kg ha^{-1} yr^{-1} for nitrogen and on the average accounts for 31% of the total nitrogen deposition. Dry sulfur deposition is less than 1 kg ha^{-1} yr^{-1} and accounts for 19% of the average total sulfur deposition. There are no evident annual dry deposition trends. Except for 1992, the limited 6 yr trend of wet plus dry deposition for nitrogen has been slightly downward from 5 kg ha^{-1} yr^{-1} in 1989 to 3.6 kg ha^{-1} yr^{-1} in 1994, while varying between 2 and slightly under 4 kg ha^{-1} yr^{-1} for sulfur.

Comparisons between NDDN and IMPROVE sulfur concentrations suggest that the IMPROVE data are compatible. With additional research into expanding the use of IMPROVE data, it may prove useful for dry atmospheric deposition assessments.

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