

Strontium isotope studies of atmospheric inputs to forested watersheds in New Mexico

JAMES R. GOSZ & DOUGLAS I. MOORE

Biology Department, University of New Mexico, Albuquerque, NM 87131, USA

Key words: strontium, isotopes, atmospheric, inputs, flux, forest

Abstract. Stable isotopes of strontium provide a unique quantification of ecosystem processes because organisms do not differentiate between them. For landscapes with contrasting geologies, these isotopes can identify atmospheric source material from local weathered material. This study quantified the input of strontium, distribution within the ecosystem, canopy capture versus leaf leachate, canopy loss, and Sr increment in biomass from an atmospheric origin. Forest ecosystems were studied along an elevational gradient in New Mexico. Spruce forests had a much greater capacity for capturing atmospheric Sr than aspen forests; however, aspen contained more total atmospheric Sr in their tissues because of greater uptake rates and the ability to utilize atmospheric deposited Sr before the initiation of the aspen forest. This technique has excellent capabilities for estimating the relative importance of atmospheric and weathering inputs to certain ecosystems.

Introduction

The use of natural, stable isotopes to evaluate pattern and process in biological systems has become very popular. Normally, ratios of stable isotopes of an element are used to characterize a rate process because of the ability of organisms to discriminate against the heavier isotopes of low atomic weight elements (Bender 1968; Siegler et al. 1976). Graustein (1981), Graustein & Armstrong (1983) and Gosz et al. (1983) reported on the potential for using isotopes of strontium to quantify atmospheric inputs of material to a study area in New Mexico. These isotopes are valuable because organisms are unable to differentiate among the isotopes of this heavy element and differences in ratios of $^{87}\text{Sr}/^{86}\text{Sr}$ among samples reflect different geological sources for those samples. In situations where material transported into an ecosystem through the atmosphere is from a geological substrate different from that in the ecosystem, the potential exists for quantifying that atmospheric input. Graustein and Armstrong 1983, and Gosz et al. 1983 reported on studies using this technique during the growing

season for aspen and conifer forests. This paper extends those analyses with more intensive sampling of aspen and conifer forests as well as over a complete year allowing quantification of strontium budgets for these two contrasting ecosystems in New Mexico.

Detectable amounts of strontium (Sr) and rubidium (Rb) occur in most igneous, metamorphic, and sedimentary rocks. Rubidium is important because it has a naturally occurring, radioactive isotope (^{87}Rb) which decays to a stable isotope of strontium (^{87}Sr). Rubidium is most commonly found as a trace element in potassium-bearing minerals resulting in an enrichment of ^{87}Sr in those minerals over time. Thus, granite typically has a high $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, whereas limestone has a low ratio. The study area in New Mexico is ideal because the granitic mountains are like "islands" in the surrounding "sea" of lowland sedimentary materials. Air masses passing over the lowland areas transport the sediment until the mountains and their vegetation result in its deposition. The low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio signature of this transported material allows it to be identified, quantified, and traced through the ecosystem in which it was deposited. Most importantly, the isotope ratio allows quantification of aerosol material collected by the canopy between rain events which is subsequently washed off enriching the throughfall beneath the canopy. Without such a technique it is difficult to identify material adhering onto the outside of the vegetation surface (aerosol collection) from that leached out of the vegetation.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the vegetation tissue reflects the isotopic ratio of Sr taken up by roots from the soil. The uptake is a mixture of Sr originating from the two end member sources; in this case these are atmospheric inputs and bedrock weathering products. A mixing equation can be used to identify the proportion of each source present in the plant uptake. A convention established by Graustein and Armstrong (1983) expresses $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in relation to an arbitrary local standard. In their case this standard was the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for the precipitation at the Santa Fe Airport (0.7088) because it was both the lowest ratio measured and characterized atmospheric inputs from outside the study area. This ratio also will be used for this study to allow comparisons. Also, the convention we will use identifies relative differences rather than absolute. Because the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of a mixture is not a linear combination of the two end members, $^{87}\text{Sr}/(^{87}\text{Sr} + ^{86}\text{Sr})$ is the proper term to express the Sr composition ratio. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are converted to a $\Delta^{87}\text{Sr}$ which expresses a relative change from that of the local standard via the following formula:

$$\Delta^{87}\text{Sr} = [^{87}\text{Sr}/(^{87}\text{Sr} + ^{86}\text{Sr})_{\text{SAMPLE}} - ^{87}\text{Sr}/(^{87}\text{Sr} + ^{86}\text{Sr})_{\text{STANDARD}}] \times 10,000$$

(a)

Then the mixing ratio will have the form:

$$X = \frac{\Delta^{87}\text{Sr}_M - \Delta^{87}\text{Sr}_B}{\Delta^{87}\text{Sr}_A - \Delta^{87}\text{Sr}_B} \quad (\text{b})$$

Where X is the proportion of component A in the mixture; M refers to the mixture while A and B refer to end members. If component B is the atmospheric source, represented by the local standard (0.7088), then $\Delta^{87}\text{Sr}_B = 0$ and:

$$X = \frac{\Delta^{87}\text{Sr}_M}{\Delta^{87}\text{Sr}_A} \quad (\text{c})$$

Mixing ratios, applied to $\Delta^{87}\text{Sr}$ of spruce and aspen vegetation, provide relative contributions of atmospheric Sr and weathered Sr. Low $\Delta^{87}\text{Sr}$ (nearer 0) reflects higher relative contribution of atmospheric Sr while high $\Delta^{87}\text{Sr}$ indicates greater incorporation of Sr from a weathering origin. Mixing ratios applied to throughfall provide a means of calculating relative contribution from atmospheric inputs (precipitation and dry deposition) as opposed to Sr leached from the canopy foliage. In this case the throughfall is the mixture (M), the $\Delta^{87}\text{Sr}$ within foliage tissue is component A and component B is the atmospheric source (material adhering to the outside of the foliage).

Study area

The study area is in the Sangre de Cristo Mountain Range of New Mexico (Sante Fe National Forest). The elevational gradient (2365 m–3734 m) produces a gradient of temperature, humidity, wind, and precipitation resulting in a variety of communities ranging from pinon-juniper to alpine tundra. Disturbances, such as fire, have resulted in different successional states in those communities. Studies were made at a number of sites along the elevational range with intensive analyses performed in aspen (*Populus tremuloides*) and spruce (*Picea engelmannii*) communities. More complete descriptions of climate, vegetation, soils, hydrology, and geology can be found in Gosz (1975, 1980a, b) and Graustein (1981).

Methods

Water samples

Precipitation and throughfall samples were collected for a water-year from 1 Nov. 1982 and to 31 Oct. 1983. Precipitation samples were collected adjacent to the spruce and aspen stands as well as at sites adjacent to a spruce-fir stand (at 3450 m) and to a pinon-juniper stand (at 2365 m). Throughfall samples were collected under the canopy of the spruce and aspen stands.

During winter, precipitation was collected in 120 l plastic garbage buckets that had been acid washed and lined with plastic bags. Throughfall was collected in 20 l (29 cm dia.), acid-washed plastic buckets. Fifteen buckets were placed beneath each of the spruce and aspen canopies at corner points of a 25 m grid surveyed on the stands. After a precipitation event precipitation and throughfall samples were weighed and subsamples from each collector transferred to 4 l plastic bottles for transportation to the laboratory. Recording precipitation gauges, located adjacent to all precipitation collectors, were used to determine precipitation volumes. During the summer (May–Oct.), precipitation and throughfall were collected in 26 cm diameter funnels draining into 4 l plastic bottles. Fifteen funnels were used to collect throughfall in each stand. Two funnels collected open precipitation at each site to obtain sufficient volumes for analysis. During summer, precipitation and throughfall were collected weekly if there had been sufficient precipitation during the previous week.

A volume-weighted sample was composited from the 15 throughfall samples for each week. At approximately two month intervals, volume-weighted composites were made for precipitation and throughfall for each two-month period. Samples were filtered through 0.45 μ membrane filters resulting in a soluble and particulate fraction for each sample. The filters were analysed for Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios.

Aerosol samples

Aerosol samples were collected using a series 280 Cyclade In-Stack Cascade Impactor in line with a Series 112 Constant Flow Air Sampler. Cyclone stages and a backup filter allowed the separation of aerosol size classes at 10, 5, 2, and 0.45 microns. Although the air sampler was allowed to run

continuously for up to 2 months, only small amounts of aerosols were collected in each stage.

Vegetation samples

Tissue samples from 30 trees were collected from the spruce and aspen stands. These included foliage, branches, bole wood, bark, roots and epiphytic lichens (at spruce only). At the spruce site, needle, branch and lichen samples were collected by climbing randomly selected trees and harvesting 3 branches from each of the lower, middle, and upper sections of the crown. Representative subsamples of needles, small branches, large branches, and lichens were obtained from the nine branches and composited for analysis. Bark samples were excised from the boles of the trees and bole wood samples were obtained using an increment corer. Root samples were obtained from 10 soil core samples and were carefully washed before analysis. Small sample mass required that roots from the 10 replicate cores be composited into 3 samples for Sr analyses. At the aspen site, aboveground tissue samples were collected from an adjacent watershed (AW-3) to avoid altering the canopy of the aspen study site (AW-1). Entire trees were harvested to obtain representative leaf and branch samples. Random leaf and branch samples were collected from each tree and a disk was removed from the bole for wood and bark samples. Root samples were removed from soil cores collected in the study watershed (AW-1) and composited as described above.

All tissue samples were analysed for ash content and Sr. The 30 composite tissue samples from each site were randomly subsampled into 3 composite subsamples for $^{87}\text{Sr}/^{86}\text{Sr}$ analyses.

Chemical analyses

Strontium isotopic ratio analysis was performed using a Nier-design, Nuclide 1290 mass spectrometer. All samples were dissolved in HF and HClO_4 acids and Sr and Rb were separated by ion exchange. Samples containing organic matter were ashed at 500 °C for two hours to remove all organic C prior to acid dissolution. All samples were normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. Analysis of the Eimer and Amend standard SrCO_3 gave a mean identical to that reported by other laboratories (0.7080). Total strontium was analysed by atomic absorption spectrophotometry.

Results and discussion

General

It is necessary to establish the ratios of the end members of the mixing equation (i.e. atmospheric inputs and bedrock on which the vegetation is growing) in order to calculate the relative contribution of atmospheric and weathering products to total Sr pools found in the vegetation. While measurement of Sr isotope ratios on individual samples is extremely precise, the establishment of spatial variability of bedrock ratios and the temporal variability of the atmospheric inputs requires extensive sampling. This paper will concentrate on the interpretation of atmospheric and vegetation Sr ratios.

Separation of all precipitation and throughfall samples into soluble and particulate fractions was done to facilitate analyses of Sr concentrations and isotope ratios. The soluble Sr presumably is the form readily incorporated into vegetation biomass. The biological availability of Sr in the particulate phase is not known. In virtually all cases, there was a considerable difference between isotope ratios of soluble and particulate material. However, during this particular year the contribution of particulate Sr to the total sample was very small and had little influence on the whole sample ratio. Samples collected during different years may have had different contributions of particulate material which would affect ratios for bulk precipitation (e.g. Gosz et al. 1983; Graustein & Armstrong 1983). Because of the uncertainty of the time frame in which particulate Sr becomes available for biological uptake, we feel only soluble Sr should be used in mixing ratio equations. This also may explain some of the difference between results of this study and those of previous studies (Gosz et al. 1983; Graustein & Armstrong 1983).

Precipitation

Table 1 lists bimonthly and annual Sr concentrations, fluxes, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$ for precipitation and throughfall in the soluble and the total (soluble + particulate) fractions. Precipitation input to the Tesuque watersheds for the 1982–83 water year was fairly typical for this area; however, seasonal distribution was atypical. Inputs came as snow from November through April with abnormally high snow inputs during March and early April due to an El Nino event (Quinn et al. 1987). This is especially significant because these spring months normally are associated with high winds and dust transport from lower elevations. May and June were quite

Table 1. Bimonthly and annual precipitation volumes, concentrations, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$ for soluble and soluble + particulate at four elevations.

Date	cm of Precip.	Soluble Sr				Soluble + Particulate Sr			
		μg/l	g/ha	⁸⁷ Sr/ ⁸⁶ Sr	Δ ⁸⁷ Sr	μg/l	g/ha	⁸⁷ Sr/ ⁸⁶ Sr	Δ ⁸⁷ Sr
Spruce (3600 m)									
Nov-Dec	20.79	0.20	0.42	0.7090	0.7	0.21	0.43	0.7091	1.1
Jan-Feb	16.6	0.08	0.13	0.7099	3.8	0.11	0.18	0.7100	4.1
Mar-Apr	38.9	1.11	4.32	0.7099	3.8	1.32	5.14	0.7108	7.0
May-Jun	3.7	9.08	3.36	0.7103	5.1	10.29	3.81	0.7108	6.8
Jul-Aug	18.5	1.51	2.79	0.7103	5.1	1.54	2.84	0.7104	5.6
Sep-Oct	16.4	0.91	1.49	0.7101	4.4	0.98	1.61	0.7119	10.5
Spruce-fir (3450 m)									
Nov-Dec	23.5	0.50	1.17	0.7096	2.7	0.54	1.27	0.7099	3.8
Jan-Feb	9.1	0.24	0.22	0.7105	5.8	0.28	0.26	0.7106	6.2
Mar-Apr	32.3	1.48	4.79	0.7106	6.2	1.56	5.05	0.7108	6.9
May-Jun	6.2	5.73	3.54	0.7098	3.4	7.07	4.37	0.7107	6.5
Jul-Aug	16.9	1.86	3.15	0.7101	4.4	1.89	3.20	0.7102	4.8
Sep-Oct	16.3	1.02	1.66	0.7098	3.4	1.04	1.69	0.7100	4.1
Aspen (3200 m)									
Nov-Dec	17.0	1.08	1.83	0.7130	14.3	1.15	1.95	0.7130	14.5
Jan-Feb	14.8	0.15	0.22	0.7131	14.7	0.17	0.25	0.7130	14.3
Mar-Apr	27.9	0.95	2.65	0.7111	7.9	1.05	2.93	0.7114	8.9
May-Jun	5.8	6.36	3.70	0.7105	5.8	6.99	4.07	0.7109	7.1
Jul-Aug	18.1	2.10	3.80	0.7107	6.5	2.27	4.11	0.7112	8.2
Sep-Oct	14.5	1.12	1.62	0.7115	9.2	1.16	1.68	0.7119	10.5
Pinon-juniper (2365 m)									
Nov-Dec	9.6	0.81	0.78	0.7095	2.4	0.87	0.84	0.7099	3.9
Jan-Feb	9.0	0.14	0.13	0.7104	5.5	0.24	0.22	0.7119	10.7
Mar-Apr	11.3	2.66	2.99	0.7104	5.5	2.83	3.18	0.7107	6.5
May-Jun	3.1	9.27	2.91	0.7113	8.5	11.84	3.72	0.7127	13.3
Jul-Aug	13.4	2.38	3.18	0.7105	5.8	2.63	3.52	0.7112	8.1
Sep-Oct	12.1	1.42	1.72	0.7107	6.5	1.46	1.77	0.7110	7.4
Total annual precipitation (1982-83)									
Sr concentrations and Sr ratios are volume-weighted means									
Date	cm of Precip.	Soluble Sr				Soluble + Particulate Sr			
		μg/l	g/ha	⁸⁷ Sr/ ⁸⁶ Sr	Δ ⁸⁷ Sr	μg/l	g/ha	⁸⁷ Sr/ ⁸⁶ Sr	Δ ⁸⁷ Sr
Spruce	114.9	1.09	12.5	0.7101	4.4	1.22	14.0	0.7108	6.8
Sp.-fir	104.3	1.39	14.5	0.7101	4.5	1.52	15.8	0.7105	5.8
Aspen	98.0	1.41	13.8	0.7112	8.1	1.53	15.0	0.7115	9.2
P.J.	58.5	2.00	11.7	0.7106	6.3	2.26	13.2	0.7114	8.8

dry and summer thundershowers began in July and continued through October.

Precipitation volumes increased with elevation due to orographic effects but Sr concentrations in precipitation decreased with elevation resulting in rather uniform annual inputs of Sr at all elevations (Table 1). Uniform inputs of nutrients over the elevational range of these watersheds has been reported for other elements (Gosz 1975, 1980b). Early snows (November–February) contributed considerable moisture but little Sr. Concentrations of Sr increased somewhat during the March–April collection period which, coupled with the large precipitation volumes, resulted in the greatest Sr flux during this time period. A more typical, drier spring could have resulted in Sr inputs many times those of 1983. May–June precipitation volumes were low but Sr concentrations were high. Generally, Sr isotope ratios were lowest during the clear-air months of late fall and mid-winter; however, the aspen site differed from this pattern (Table 1). The lowest ratios agree well with the valley precipitation value obtained by Graustein and Armstrong (1983) of 0.7088.

Aspen precipitation reflected a local contamination source of Sr. Elevated K concentrations (Gosz 1980b; Gosz et al. 1983) and Sr isotope ratios that were greater than those found in precipitation at both higher and lower sites (Table 1) implicate local vegetation as the probable source of contamination. The apparent contamination during both winter and summer months argues against any local soil or road dust. This contamination of samples was assumed to be the result of resuspension of soluble material (i.e. internal tissue solutes, Martell & Poet 1972) from the aspen canopy with a higher isotope ratio which was subsequently returned in precipitation resulting in the increase in the isotope ratio of bulk precipitation. Therefore, quantifying the true net atmospheric inputs was complicated by this mixing of locally and non-locally derived material. It is possible to use isotope ratios and a mixing equation to estimate the amount of this resuspended Sr returned to the aspen system by precipitation. Using the mean $\Delta^{87}\text{Sr}$ of aspen tissue for an end member, the resuspended (local contamination) Sr contribution was estimated at $1.1 \text{ g ha}^{-1} \text{ yr}^{-1}$, 9% of the total precipitation input. The sphere of influence of these “contaminants” may not be confined to the vicinity of the aspen stands. Some proportion of solutes resuspended in the aspen stand probably was carried downwind (upslope) to the spruce-fir areas where it may have been removed by precipitation or dry deposition. The spruce and fir vegetation also may have generated resuspended solutes which could

have been scavenged from the atmosphere by precipitation or dry deposition. This contamination of precipitation by material from a different geologic source may account for the difference between mountain rain and valley rain.

Aerosols

The chemistry and Sr isotope ratios of incident aerosols is the key to quantifying dry deposition in vegetation canopies of the study area. Unfortunately, clear air conditions in the area made it difficult to collect sufficient sample sizes to perform all of the desired analyses. Low concentrations of Sr in mineral material (about 100 $\mu\text{g/g}$), makes it necessary to collect large samples of aerosols which was feasible only during spring months when winds transported sufficient quantities of aerosols to the vicinity of the collector. Due to technical problems and unusually low dust conditions, only one aerosol sample was of sufficient size to allow analysis of most fractions (Table 2).

Strontium isotope ratios of the aerosols confirmed that large aerosols had

Table 2. Strontium concentrations and isotope ratios for a single aerosol collection and the particulate portions of precipitation and throughfall samples.

Aerosols (digested with HF-HClO ₄)				
Size class (microns)	Sample weight (mg)	Sr ppm	⁸⁷ Sr/ ⁸⁶ Sr	$\Delta^{87}\text{Sr}$
0.5-1.5	3.47	86	0.7102	4.8
1.5-15	8.08	198	0.7118	10.3
> 15	0.43	—	0.7124	12.3
Particulate portion of precipitation and throughfall				
Site	Sample	HF-HClO ₄ digestion		HCl-HNO ₃ digestion
		Sr ppm	⁸⁷ Sr/ ⁸⁶ Sr	Sr ppm
Spruce	Precipitation	152	0.7145	46
	Throughfall	122	0.7154	41
Spruce-fir	Precipitation	150	0.7146	81
Aspen	Precipitation	116	0.7146	48
	Throughfall	128	0.7155	41
Pinon-Juniper	Precipitation	66	0.7177	26

higher isotope ratios than did small aerosols; 0.7124 vs. 0.7102. Particulates ($> 0.45 \mu$) in precipitation also had isotope ratios higher than those of soluble fractions. However, during the January–February collection period, isotope ratios of particulates in precipitation were low and similar to those of the soluble fractions. Based on concentrations of nutrients in the winter precipitation (Gosz 1980b), this also was the period of cleanest air. Presumably, only smaller sized aerosols would have been transported by incident air masses during that time (Whitby 1977) resulting in a similarity between soluble and particulate ratios. The Sr concentrations and Sr isotope ratios of particulates do not necessarily reflect material available to the vegetation because the aerosol samples were digested with HF-HClO₄ for a total Sr analysis. The particulate portion also does not account for the contamination of precipitation mentioned previously. Table 2 demonstrates a typical 2–3 fold difference between an analysis of Sr that is relatively available (i.e. digested with HCl-HNO₃, Anderson 1974), versus a total digestion (HF-HClO₄) of particulates in precipitation, throughfall, and streamwater.

Bedrock Sr ratios

Determination of reliable $\Delta^{87}\text{Sr}$'s for bedrock underlying the spruce and aspen watersheds is vital to establishing the relative contributions of atmospheric and weathering inputs to vegetation Sr. Two factors make this determination difficult. The geology of the Embudo granite underlying the study sites is very diverse with minerals having $\Delta^{87}\text{Sr}$ ranging from 42 to > 1000 . The relative abundance and distribution of these mineral types will determine the mean $\Delta^{87}\text{Sr}$ of locally available Sr in the watershed. Even if sufficient samples could be collected to establish an accurate watershed average it still may be meaningless. The $\Delta^{87}\text{Sr}$ of Sr weathered from rock is likely to be different than that of the bedrock as a whole. Certain minerals are more readily weathered than others and it is the mean $\Delta^{87}\text{Sr}$ of this "weatherable" Sr that should be considered the end member in a mixing ratio.

Table 3 provides examples of weathering source $\Delta^{87}\text{Sr}$ for spruce and aspen sites based on the deepest soil horizon collected (25–35 cm) as well as from two chemical methods of deriving $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for bedrock and weathering products (Gosz & Moore, in prep.). For this paper, a mean of the three $\Delta^{87}\text{Sr}$'s in Table 3 will be used as the bedrock end member values for each site; 72 for spruce and 79 for aspen. These values are relatively close to the estimate of 88 by Graustein & Armstrong (1983) for this area. However, we feel that more extensive sampling and weathering experiments are needed to quantify watershed representative values.

Table 3. $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$ values for samples of C horizon, regolith, and weathering products for othe spruce and aspen sites.

Gosz (in prep.)	Spruce		Aspen	
	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$
C Horizon 25–35 cm	0.7305	73.4	0.7330	81.7
Regolith	0.7305	73.4	0.7320	78.4
Weathering products	0.7295	70.0	0.7322	79.1

Vegetation Sr ratios

Table 4 lists the Sr isotope ratios and concentrations for individual composite samples of spruce and aspen tissues. An indication of the sensitivity of the $^{87}\text{Sr}/^{86}\text{Sr}$ analysis can be seen from those results. $\Delta^{87}\text{Sr}$'s of all aspen tissue types within a composite were similar; however, at aspen there were significant differences ($p < 0.05$) among composites. For spruce, there were no significant differences between $\Delta^{87}\text{Sr}$'s of composites, but lichens were significantly different ($p < 0.05$) from all tree tissues. This difference is expected since the lichens obtain a majority of their nutrients (and Sr) from the atmosphere and foliage leachates. Presumably, a reduction in the leaching contribution and/or the leaching of lower isotope ratio material characteristic of composite 2 trees accounts for the lower ratio of composite 2 *Usnea*. The $\Delta^{87}\text{Sr}$ of roots from both the aspen and spruce stands did not follow the trend of the other tissues; not surprising since the composite root samples were not collected from the trees sampled for other tissues. For aspen, roots were not collected from the same watershed. Elimination of the roots and *Usnea* tissues from the ANOVA's and Duncan's tests, resulted in all aspen composites being significantly different ($p < 0.05$) and the second spruce composite significantly different from the other two (Table 4). Sr ratios for the small branches of the spruce, while not being significantly different than the other tissues, had the lowest $\Delta^{87}\text{Sr}$ values infering a larger atmospheric component. The rough surfaces of small spruce branches may be more efficient at capturing aerosols than needles or less easily washed of their collected aerosol material by precipitation. Despite the considerable within-site variation of trees, a significant ($p < 0.001$) difference between spruce and aspen $\Delta^{87}\text{Sr}$ values for vegetation tissues was demonstrated.

A relatively large difference occurred for isotope ratios between aspen tissues of the study watershed (AW-1) and the adjacent watershed (AW-3), where most tissues were collected. A composite sample of aspen leaf litter collected adjacent to each of the 15 throughfall collection funnels in AW-1

Table 4. $\Delta^{87}\text{Sr}$ values for tissue composites from the spruce and aspen stands. Means in a row or column, having the same letter, are not significantly different ($p < 0.05$).

Site	Tissue type	Composite			Mean	Std. error
		1	2	3		
Spruce	Roots	23.5	22.5	19.8	21.9 a	1.1
	Boles	22.9	19.5	22.5	21.6 ab	1.1
	Needles	22.2	18.4	22.2	20.9 ab	1.2
	Bark	21.2	19.8	21.8	20.9 ab	0.6
	Lg. Branch	21.5	18.4	22.2	20.7 ab	1.2
	Sm. Branch	19.8	14.7	19.8	18.1 b	1.7
	Usnea sp.	13.7	12.0	13.3	13.0 c	0.5
	Mean	20.7 a	17.9 a	20.2 a	19.6	
	Std. error	1.3	1.3	1.2		
	Excluding roots and Usnea sp.					
	Mean	21.5 a	18.2 b	21.7 a	20.5	
Site	Type	Composite			Mean	Std. error
		1	2	3		
Aspen	Roots	48.6	51.3	43.2	47.7 a	2.4
	Boles	46.9	53.6	49.6	50.0 a	2.0
	Leaves	44.2	54.0	50.6	49.6 a	2.9
	Bark	42.8	54.0	47.2	48.0 a	3.2
	Branches	44.5	53.3	47.2	48.3 a	2.6
	Mean	45.4 a	53.2 b	47.6 a	48.7	
	Std. error	1.0	0.5	1.3		
	Excluding roots					
	Mean	44.6 a	53.7 c	48.7 b	49.0	

had a $\Delta^{87}\text{Sr}$ of 35.4 compared with the mean of 49.6 for leaves from AW-3. Also, a composite sample of bark samples taken from the 15 trees in closest proximity to the 15 throughfall collectors had a mean $\Delta^{87}\text{Sr}$ of 34.7 in AW-1 compared with 47.9 for bark collected from trees in AW-3. This spatial variation may occur within a given watershed as well. The $\Delta^{87}\text{Sr}$ of dead and down tree bole samples (40.8) that were taken from throughout the entire AW-1 watershed suggests that the average $\Delta^{87}\text{Sr}$ for vegetation for the entire watershed approximated 40.8. Slight differences in aspect (different weathering processes) and mineralogical differences (Graustein 1981) would contribute to variation in isotope ratios. Values reported by Graustein & Armstrong (1983) were lower than those reported here which attests to the spatial variation that can occur in the same general area.

Table 5 lists biomass, Sr concentrations, calculated Sr pools and $\Delta^{87}\text{Sr}$'s for aspen and spruce vegetation. Summation of these pools indicated that, although the living biomass was virtually identical between aspen and spruce, the Sr pool in aspen vegetation was more than twice that in spruce. Using a mixing equation from page 3 to calculate the contribution of atmospheric and bedrock Sr to the pool of Sr in aspen and spruce vegetation showed that despite assumed greater dry deposition in spruce, aspen vegetation contained about 50% more atmospheric-derived Sr (3348 g ha^{-1}) as did spruce vegetation (2265 g ha^{-1}). Using the range of $\Delta^{87}\text{Sr}$ values in aspen tissue (34.7–50.0), the calculated range of atmospheric-derived Sr was 4826–3185 g ha^{-1} . A large uptake of bedrock-derived Sr by aspen resulted in a high $\Delta^{87}\text{Sr}$ and created the impression that aspen vegetation contained less atmospheric Sr than spruce.

Since annual inputs of atmospheric Sr to aspen are low, what is the source of this large pool of atmospheric-derived Sr? An example using soluble Sr in precipitation during the 1982–83 water year as a mean input and allowing for 100 years since the beginning of the aspen growth demonstrates that only

Table 5. Comparison of biomass, Sr concentrations and Sr pools in aspen and spruce stands. The proportion of biomass Sr from atmospheric and bedrock sources was estimated from isotope ratios and mixing equations.

Aspen					Spruce			
$\Delta^{87}\text{Sr}$	Sr g/ha	Sr $\mu\text{g/g}$	Biomass kg/ha	Tissue	Biomass kg/ha	Sr $\mu\text{g/g}$	Sr g/ha	$\Delta^{87}\text{Sr}$
49.6	183	67	2728	Leaves	18609	20	372	20.9
48.3	547	71	7706	Branch	48300	20	966	19.8
50.0	2357	18	130940	Bole	88385	6	530	21.5
48.0	2937	133	22081	Bark	11179	33	369	20.9
47.7	1878	85	22090	Root	27940	25	699	21.9
				Lichen	1375	16	22	13.0
40.8	104	8	13053	Dead stems	5058	6	30	21.0
48.7	70	56	1300	Understory			0	
48.5	8076	42	199898	All veg.	200846	15	2988	20.8
79.7				Bedrock				72.3
4.4				Atmospheric inputs				4.4
Aspen					Spruce			
Sr g/ha					Sr Source			
3348					Atmospheric Sr			
4728					Bedrock Sr			
					2265			
					724			

about 1400 g ha^{-1} out of the approximately 4000 g ha^{-1} of atmospheric-derived Sr in the AW-1 vegetation could have been obtained from atmospheric inputs since the fire that initiated the aspen stand. This means that the input of atmospheric material in 1982–83 was abnormally low (supported by our discussion about a wet spring) and/or the residual atmospheric Sr, collected by conifer forests before the 1886 burn, was not eroded away following the fire. Subsequent mineralization and uptake of that Sr by aspen could account for the difference.

Throughfall

Precipitation collected under the forest canopy often is termed gross throughfall. Table 6a lists bimonthly precipitation and gross throughfall volumes, Sr concentrations, fluxes, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$'s for aspen and spruce. The annual 1982–83 totals and means for the gross throughfall as well as totals and means for summer and winter periods also are listed.

Net throughfall, the difference between gross throughfall and incident precipitation, is a measure of the enrichment or depletion as precipitation passes through the canopy. Table 6b lists net throughfall Sr concentrations, fluxes, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$'s for the spruce and aspen sites on an annual and semi-annual basis. The form of precipitation, rain or snow, entering the canopy is critical to the chemistry of resultant throughfall. Winter precipitation volumes were almost twice as great as those of summer but enrichment of Sr by the canopy during the winter was slight.

Application of mixing equation (b) to throughfall $\Delta^{87}\text{Sr}$ is ideally suited for calculating the relative contribution of foliage leachates and dry deposition to net throughfall. Throughfall Sr is a mixture of Sr from atmospheric sources (precipitation and dry deposition) and vegetation leachates (Sr leached from within foliage tissue). The $\Delta^{87}\text{Sr}$ of vegetation leachates can be assumed to equal that of the bole wood (both reflect Sr taken up from the soil) and $\Delta^{87}\text{Sr}$ of atmospheric inputs are assumed to be that of soluble Sr in precipitation. Table 7 lists calculated contributions of precipitation, dry deposition, and foliage leaching to 1982–83 throughfall for spruce and aspen. The influence of seasonality on throughfall concentrations and fluxes is demonstrated in the comparison between winter and summer values.

For 1982–83, the canopy throughfall approximately doubled Sr inputs reaching the forest floor by precipitation alone (Table 6b). However, the origin of the contributed Sr was quite different beneath aspen and spruce. For 1982–83, dry deposition accounted for $\sim 40\%$ of the Sr in net throughfall beneath spruce while beneath aspen it contributed only about 10%

Table 6a. Bimonthly precipitation and throughfall volumes, concentrations, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, and $\Delta^{87}\text{Sr}$ for spruce and aspen stands.

Date	cm of Precip.	Precipitation Sr				cm	Throughfall Sr			
		$\mu\text{g/l}$	g/ha	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$		$\mu\text{g/l}$	g/ha	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$
Spruce (3600 m)										
Nov-Dec	20.79	0.20	0.42	0.7090	0.7	19.7	0.32	0.63	0.7102	4.8
Jan-Feb	16.6	0.08	0.13	0.7099	3.8	17.0	0.11	0.19	0.7111	7.9
Mar-Apr	38.9	1.11	4.32	0.7099	3.8	37.6	1.67	6.27	0.7104	5.5
May-Jun	3.7	9.08	3.36	0.7103	5.1	2.0	21.89	4.40	0.7116	9.6
Jul-Aug	18.5	1.51	2.79	0.7103	5.1	17.4	5.26	9.15	0.7116	9.6
Sep-Oct	16.4	0.91	1.49	0.7101	4.4	14.4	3.09	4.46	0.7121	11.3
Aspen (3200 m)										
Nov-Dec	17.0	1.08	1.83	0.7130	14.3	17.5	1.47	2.57	0.7178	30.7
Jan-Feb	14.8	0.15	0.22	0.7131	14.7	15.1	0.16	0.24	0.7162	25.2
Mar-Apr	27.9	0.95	2.65	0.7111	7.9	34.0	1.64	5.58	0.7116	9.6
May-Jun	5.2	4.67	2.45	0.7108	6.7	4.1	11.70	4.82	0.7139	17.4
Jul-Aug	18.1	1.89	3.42	0.7106	6.0	13.0	4.68	6.08	0.7150	21.2
Sep-Oct	14.2	1.19	1.69	0.7114	8.9	10.9	4.68	5.11	0.7160	24.6

Table 6b. Precipitation and throughfall Sr concentrations, fluxes, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and $\Delta^{87}\text{Sr}$'s on an annual and seasonal basis.

Site	cm	Precipitation Sr				cm	Throughfall Sr			
		$\mu\text{g/l}$	g/ha	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$		$\mu\text{g/l}$	g/ha	$^{87}\text{Sr}/^{86}\text{Sr}$	$\Delta^{87}\text{Sr}$
Annual										
Spruce	114.9	1.09	12.5	0.7101	4.4	108.2	2.32	25.1	0.7114	8.7
Aspen	97.1	1.26	12.3	0.7112	8.3	94.7	2.58	24.4	0.7145	19.5
Winter										
Spruce	76.3	0.64	4.87	0.7098	3.5	74.4	0.95	7.08	0.7104	5.5
Aspen	59.6	0.79	4.70	0.7119	10.7	66.7	1.26	8.40	0.7136	16.5
Summer										
Spruce	38.6	1.98	7.64	0.7103	5.00	33.9	5.32	18.01	0.7117	10.0
Aspen	37.5	2.01	7.55	0.7108	6.86	28.0	5.71	16.01	0.7150	21.1

(Table 7). Graustein and Armstrong reported 66% and 0% respectively for spruce and aspen during the June-September period of 1975. The summer values for 1983 were 38% and 3%, respectively. The differences between these studies are likely due to the drier nature of 1975 and the total digestion technique used by Graustein & Armstrong versus the soluble Sr analyses in this study. On a seasonal basis, foliage leachates demonstrated a greater contribution to net throughfall during the summer.

Table 7. Annual and seasonal net throughfall and contributions from aerosol and foliage leachates during 1982–83.

Site	Net Tfall Sr				g/ha		% of Total	
	μg/l	g/ha	⁸⁷ Sr/ ⁸⁶ Sr	Δ ⁸⁷ Sr	Aerosol	Foliage	Aerosol	Foliage
Annual								
Spruce	1.16	12.6	0.7126	13.0	5.19	7.40	41	59
Aspen	1.28	12.2	0.7178	30.8	1.41	10.75	12	88
Winter								
Spruce	0.30	2.2	0.7117	9.8	1.25	0.97	56	44
Aspen	0.55	3.7	0.7158	23.8	1.18	2.52	32	68
Summer								
Spruce	3.07	10.4	0.7128	13.7	3.94	6.44	38	62
Aspen	3.02	8.5	0.7187	33.8	0.23	8.23	3	97

On a g/ha basis, dry deposition was proportionately greater during the summer at spruce. This apparent difference in seasonality of dry deposition between spruce and aspen may be the result of a difference in the onset of “summer” at the two sites. May 1 was designated as the break between winter and summer for both sites. At spruce, precipitation was primarily snow at this time while precipitation at aspen often was rain. Therefore, aerosols collected by the aspen canopy during the winter may have been removed by rains prior to May 1. Material deposited on the conifers during the winter may not have been washed from the foliage until precipitation arrived as rain (late May or early June) and recorded as summer deposition. Also, dry deposition in aspen may have been overestimated because of exaggerated throughfall volumes caused by the open nature of the stand which resulted in collection of wind-transported snow in throughfall buckets. Using precipitation collector volumes to correct throughfall volumes gave a calculated dry deposition of 0.42 g/ha instead of 1.18 g/ha.

The strontium cycle

Principal hypotheses of this study have been that conifers should be better at filtering aerosols from the atmosphere than deciduous tree species and as a result, soil and plant tissue chemistry in conifer stands should display a more “atmospheric” signature.

Strontium isotopes appeared an ideal method of testing these hypotheses as the Δ⁸⁷Sr of the bedrock underlaying the coniferous and deciduous forests in the Tesuque Watershed is high (> 72) while inputs of precipitation and

dry deposition have low $\Delta^{87}\text{Sr}$ (~ 4). Preliminary analyses of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios performed on conifers (spruce-fir) and on deciduous trees (aspen) appeared to confirm these hypotheses (Graustein 1981; Graustein & Armstrong 1983; Gosz et al. 1983). The $\Delta^{87}\text{Sr}$'s for spruce and fir tissues were about 18 while those of aspen were about 38.

The more comprehensive isotope analyses of tissues performed in this study also supported the above hypotheses (Table 4). Lower $\Delta^{87}\text{Sr}$ for spruce suggested that conifers have a greater capacity of capturing atmospheric Sr than do deciduous tree species such as aspen and this capacity is reflected in their tissue isotope ratios. The greater collection of aerosols and moisture by conifers led to the development of an "atmospheric" characterization in their soils which was reflected in an "atmospheric" characterization in their tissues.

Comparison of Sr pools in vegetation biomass indicated that while conifers displayed a $\Delta^{87}\text{Sr}$ closer to that of atmospheric sources than did aspen, aspen biomass actually contained substantially greater quantities of atmospheric Sr. While this would argue for greater inputs of atmospheric Sr to the aspen system, the precipitation and throughfall data refute such a hypothesis. Instead, the data may indicate an additional pool of atmospheric-derived Sr as well as an ability of aspen systems to conserve Sr strongly. Biomass in the two systems is virtually identical but the spruce trees are at least twice as old as the aspen. Slower growth rates, coupled with lower incorporation of base cations in biomass increment, results in a smaller annual base cation demand by spruce. Mean annual Sr increment for spruce and aspen are estimated at 14 and 76 g/ha respectively. Mixing equations permit calculation of yearly Sr increment that must be derived from atmospheric Sr to maintain current vegetation ratios. At spruce this amounted to about $10 \text{ g ha}^{-1} \text{ yr}^{-1}$ of the total increment of $14 \text{ g ha}^{-1} \text{ yr}^{-1}$ (Fig. 1). Based on the 1982–83 water-year, this amount was easily supplied by precipitation. Almost twice this amount was supplied by the total of precipitation and dry deposition. However, aspen, with an annual Sr increment about 6 times that of spruce requires 32–45 g/ha of atmospheric Sr to maintain the current isotopic ratio (Fig. 2). Precipitation inputs of Sr in aspen were about the same as in spruce but aspen obtains virtually no contribution from dry deposition, so that total atmospheric inputs of Sr at aspen were only about 75% of those at spruce. Even assuming no loss of atmospheric-derived Sr in stream discharge, atmospheric inputs only accounted for about a third of the aspen annual increment of atmospheric-derived Sr. Therefore, aspen trees must obtain much of their atmospheric Sr from some source other than current atmospheric inputs. One obvious source of this Sr was the large pool left by the 1886 fire (i.e. combustion of

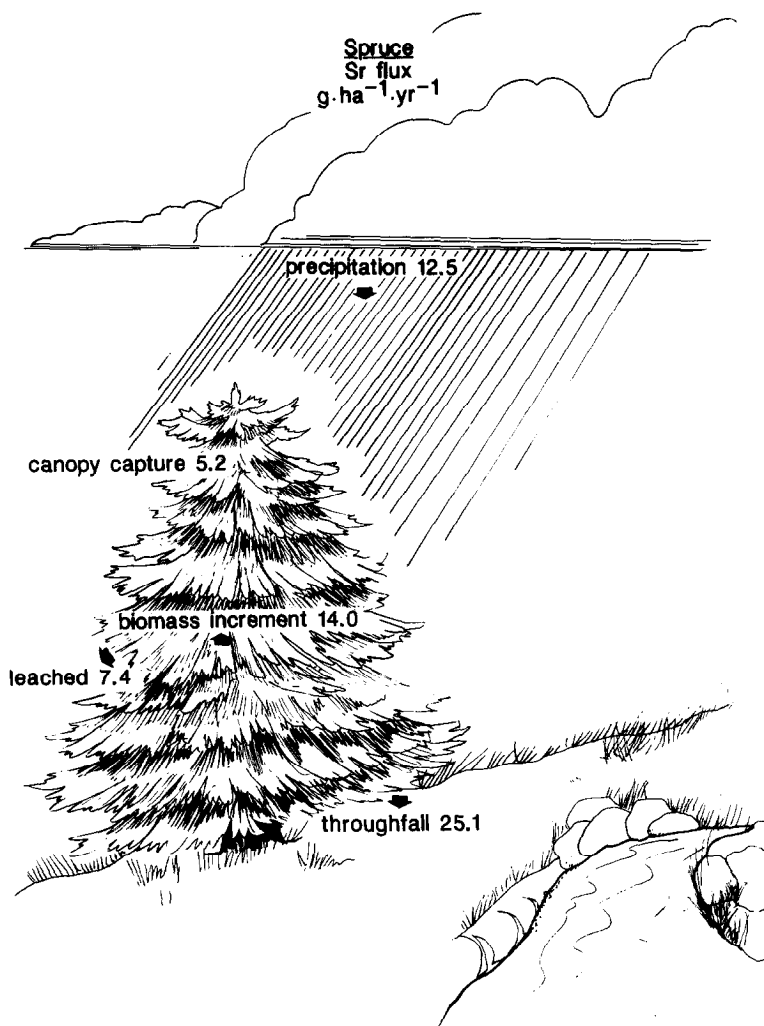


Fig. 1. Strontium fluxes for a spruce ecosystem in the Sangre de Cristo Mountains of New Mexico. Values are g per ha per year.

biomass and forest floor). An additional source of atmospheric Sr may be in soil organics formed by the conifer forest that preceded the aspen forest. Gosz et al. (1983) reported organic matter to be an important sink for Sr. Fire probably had a small effect on soil organics. Subsequent mineralization could have made that Sr available; however, and aspen may have "mined" the atmospheric-derived Sr from that reserve.

The pools and dynamics shown in Figs. 1 and 2 are essential for interpret-

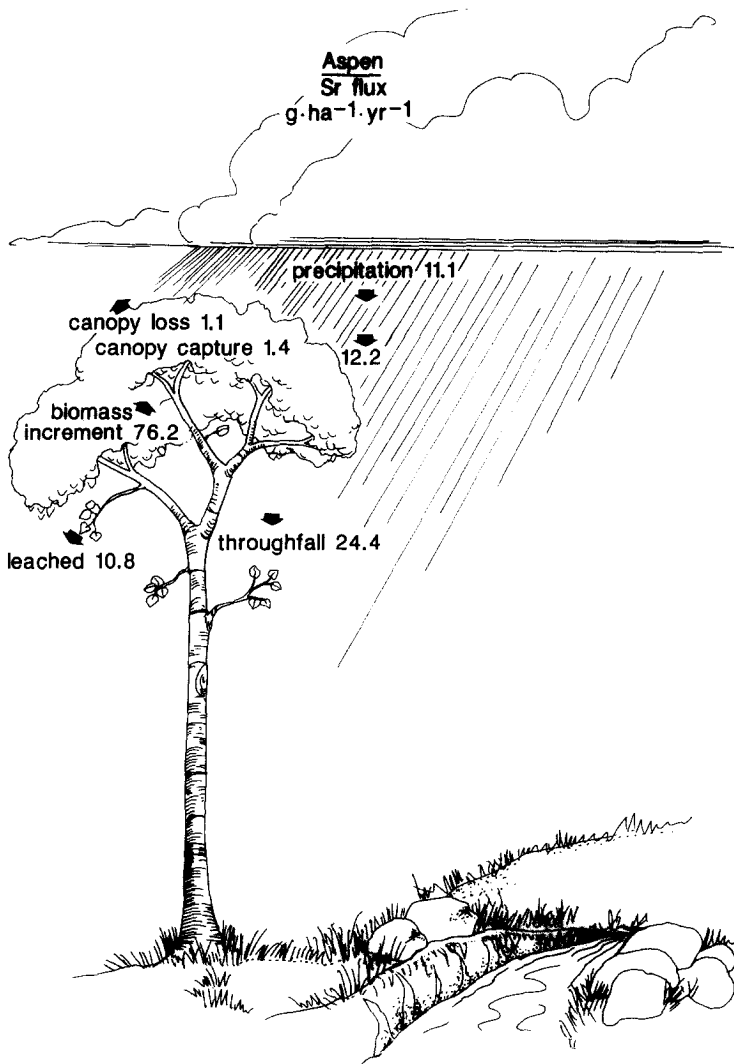


Fig. 2. Strontium fluxes for an aspen ecosystem in the Sangre de Cristo Mountains of New Mexico. Values are g per ha per year.

ing the dynamics of other nutrients in the ecosystems. Atmospheric inputs of other nutrients can be estimated by correlations with Sr inputs (Moore & Gosz, in prep.). This technique offers excellent possibilities for estimating the relative importance of atmospheric and weathering inputs to certain ecosystems that have the benefit of such contrasts in input geochemistry.

References

- Anderson J (1974) A study of the digestion of sediment by the $\text{HNO}_3\text{-H}_2\text{SO}_4$ and the $\text{HNO}_3\text{-HCl}$ procedures. Atomic Absorption Newsletter 13: 31-32
- Bender MM (1968) Mass spectrometric studies of carbon-13 variations in corn and other grasses. Radiocarbon 10: 468-472
- Gosz JR (1975) Nutrient budgets for undisturbed forests along an elevational gradient in New Mexico. In: Howell FG et al. (Eds) Mineral Cycling in Southeastern Ecosystems. (p 780-799) ERDA Symposium Series CONF-740513, Technical Information Center, Office of Public Affairs, ERDA, Washington, D.C.
- Gosz JR (1980a) Biomass distribution and production budget for a nonaggrading forest ecosystem. Ecology 61: 507-514
- Gosz JR (1980b) Nutrient budget studies for forests along an elevational gradient in New Mexico. Ecology 61: 515-521
- Gosz JR, Brookins DG & Moore DI (1983) Using strontium isotope ratios to estimate inputs to ecosystems. BioScience 33: 23-30
- Graustein WC (1981) The effects of forest vegetation on solute acquisition and chemical weathering: a study of the Tesuque Watersheds near Santa Fe, New Mexico. Ph.D. dissertation. Yale University, New Haven, CT
- Graustein WC & Armstrong RL (1983) The use of strontium-87/strontium-86 ratios to measure atmospheric transport into forested watersheds. Science 219: 289-292
- Martell EA & Poet SE (1972) Residence times of natural and pollutant aerosols in the troposphere. In: Brittin WE, West R & Williams R (Eds), Air and Water Pollution (pp 459-476) Colorado Associated University Press, Boulder, CO
- Quinn WH, Neal VT & Antunez de Mayolo SE (1987) El Nino occurrences over the past four and a half centuries. J Geophysical Res. 92: 14, 449-14, 461
- Whitby KT (1977) The physical characteristics of sulfur aerosols. Atmos. Environ. 12: 135-159
- Ziegler H, Osmond CB, Stickler W & Trimborn D (1976) Hydrogen isotope discrimination in higher plants: correlation with photosynthetic pathway and environment. Planta 128: 85-92