

Project Description

a) Introduction

The biota and soils of Black Rock Forest represent a balance between processes of formation and transformation that are continually changing. The rates of these processes can be very difficult to quantify but critical to the long-term health of the ecosystem. For many of the fundamental chemical components of the forest the delivery of new supplies via precipitation and "dry" fallout is the dominant income source. On the other side of the ledger, the primary transport of many elements out of the forest occurs in solution or suspended particles via the streams. Thus empirical data on the chemistry of precipitation and surface waters within the forest can provide fundamental information on many different processes of broad interest for future research at BRF.

One subject of global interest for which research at BRF could provide new insights concerns the role of temperate forest biota and soils in net long-term storage of fossil fuel (ff) combustion CO_2 . About half of this anthropogenic perturbation has remained in the atmosphere, while about one third has been dissolved in the world ocean (Broecker et al., 1979). The fate of the remainder is currently unresolved, but many scientists from the oceanographic community think that the soils and biota of terrestrial ecosystems, especially forests, are the most likely "sink" for the "missing" ff combustion CO_2 . Since the global nitrogen cycle has also been greatly altered by human influence, there is potential for enhanced N transports to have resulted in more storage of C in these latter ecosystems (Simpson et al., 1977). Compounding the problem of the "missing" ff carbon dioxide is the added effect of net destruction of tropical forests over the past half century. Most estimates of the magnitude of this flux suggest that it is comparable to or greater than the missing ff component. Thus the fate of the combined ff combustion and tropical forest destruction CO_2 that is currently unresolved has a magnitude similar to the two largest "known" fluxes in the global carbon budget: (1) net atmospheric increase and (2) uptake by the ocean. One of the greatest uncertainties remains the influence of terrestrial ecosystems of mid and high latitudes. Black Rock Forest is a logical site to pursue long-term research on this issue for several reasons. The forest lies in a zone for which the chemistry of precipitation and dry fallout has been greatly altered from natural conditions, providing much higher influxes of acidity and fixed nitrogen, both of which might plausibly alter

the net storage of C in the ecosystem. Secondly, BRF is in a region which has had substantial climatic warming and is likely to experience more of this trend in the next century. Thirdly, the soils in most of BRF are, on the average, quite thin due to glacial erosion and thus any change in net storage of C in this century would have been "diluted" to a lesser extent than for a site with more developed soils. Finally, the storage and exchange of surface waters with deeper ground waters, which can greatly complicate measurement of chemical budgets in surface waters, should be relatively minor at BRF due to the low permeability crystalline rocks which underly the entire site.

Justification of the need for a long-term monitoring program for precipitation chemistry at BRF is based, in part, on the history of important contributions to ecosystem research at other sites such as the Hubbard Brook Ecosystem Study (HBES) in the White Mountains of New Hampshire. These contributions include documenting the dominance of hydrogen ions, as the highest abundance cation, in precipitation chemistry of the region (Likens et al., 1977). These observations as part of the HBES remain one of the most comprehensive empirical data compilations of acid rain deposition in the entire world. The HBES precipitation data, combined with stream chemistry data, permit quantitative estimates of total dry fallout deposition of sulfate over forested drainage basins (Likens et al., 1990), and clear documentation of the increasing importance of nitric acid, as compared to sulfuric acid, in the control of hydrogen ions in precipitation (Bormann et al., 1977). The combination of long-term precipitation and stream chemistry data at HBES provided quantitative estimates of the role of aggrading forests in the net storage of hydrogen ions (Driscoll et al., 1982), the annual budgets of nitrogen and sulfur species (Nodvin et al., 1988), and the importance of temporal changes in base cations delivered by precipitation as a critical control on ecosystem acidity loading (Driscoll et al., 1989). Many of the long-term trends observed in the HBES precipitation data are quite surprising and still subject to further analysis before explanations of those trends achieve broad consensus (Hedin et al., 1990).

The nitrogen cycle in temperate forests is one of the most complex and central aspects of the processes which govern the dynamic balance of these ecosystems (Schlesinger, 1991). Although BRF does not receive discharges of sewage or agricultural wastes as occurs for many of the world's large river systems (Peierls et al., 1991), the extent of perturbation of the N cycle via precipitation is substantial and provides an important entry to investigation of net changes in the C cycle via N budget studies (Vitousek et al., 1988).

b) Changes in Sampling Strategy from Previous Program

We will not discuss sampling strategy or techniques here and refer instead to earlier proposals to access that material. The introduction section of the project description given above is taken from our proposal of last year with a few minor editing changes and is repeated here for convenience in reviewing.

During the past year, collection of samples of precipitation and surface runoff have continued, with some modifications, from the plan initiated during late summer of 1991. The basic concept remains to attempt to quantify amounts of a range of chemical species which enter the forest from the atmosphere, primarily dissolved in precipitation, and compare these with amounts of related chemical species which leave the forest dissolved in surface runoff. We consider these activities worthy of support for research in their own right, and also as supportive of long-term development of other research programs at BRF.

One significant change in sampling strategy has been to reduce the total number of precipitation samples collected at BRF, as part of our cooperative effort. From August, 1991 through September, 1993, plastic funnel collectors were emptied after each precipitation "event", to permit resolution of the range of chemical influxes as a function of the history of atmospheric transport represented by each episode of rain or snow. Such a sampling plan is preferred to help distinguish characteristics of each major storm, dependent especially upon the air mass trajectories generating the precipitation event. Since October, 1993 precipitation sampling has been altered to obtain weekly composites (initiated on Tuesday morning of each week), to permit direct comparison with results from samples collected at West Point as part of the National Acid Deposition Program (NADP) network (Sisterson et. al., 1991). Our current program at BRF is not redundant with that of the NADP station at West Point, which is usually described as based on "wet" deposition samples, where operation of the collector is initiated by opening of a cover at the beginning of each precipitation event, followed by closing of the collector at the end of an event. Between precipitation events, no collector surface is exposed to the atmosphere to be influenced by "dry" deposition. The precipitation collectors at BRF can be described as "bulk" samplers, that include all dry deposition to the collector surfaces between events, combined with that from discrete rain and snow episodes.

The above change was initiated to shift emphasis somewhat away from

measurements of precipitation chemistry towards stream chemistry. At this stage of our data collection at BRF, we have collectively decided that, with the limited personnel resources available, it is more important to outline the chemical characteristics of surface runoff. Although data from the NADP precipitation samples are generally not available for up to several years after collection, they are of very high quality and can provide quantitative estimates of many important chemical fluxes to BRF. As will be discussed below, there are several interesting observations which can already be made from combining results from the NADP data and those from BRF.

A second significant change in sampling strategy has been to appreciably expand collection of stream samples to provide information from several drainage basins at BRF. In addition to the weekly sample obtained by the Cornwall Water Department at the downstream end of the Aleck Meadow catchment, which was initiated in autumn, 1991, three new sampling sites have been established by Bill Schuster. These sites, which provide surface runoff from the catchments of Mineral Spring Brook, Canterbury Brook and Cascade Brook, should be representative of several different types of forest environments at BRF. Mineral Spring Brook is derived from Sutherland Pond and a large (>100 acre) wetland, and flows out of BRF to the southwest. Sutherland Pond was the site of a gas exchange rate tracer experiment (conducted by Lamont-Doherty Earth Observatory personnel and others) during the summer of 1993, and has been the focus of sediment sampling to reconstruct changes in vegetation history over the past 10,000 to 15,000 years from pollen measurements (NYU). As the largest "unmodified" catchment system which includes an appreciable surface water body in BRF, this drainage basin is likely to be the site of appreciable research efforts in the future. Sutherland Pond is a natural lake and is not part of the Cornwall Water Supply network. Canterbury Brook, which drains to the north, contains appreciable stands of old-growth forest, including hemlocks, and relatively deep glacial till soils. In contrast, Cascade Brook drains to the south, and the species of trees are appreciably different from those of Canterbury Brook. Since one possible direction for future research at BRF involves watershed studies that would require significant investments in stream gauging infrastructure and laboratory analyses of samples, it was judged to be more valuable to expand collection of surface waters during this pilot program and to reduce effort somewhat on collection and analyses of precipitation samples. The initial results could then help guide selection of the best watersheds for more intensive study.

Although the above changes involve appreciable change in emphasis, the general plan remains to collect samples of water (rain and snow) entering and leaving BRF (streams), and to use the chemical constituents in those samples to reveal information about processes in the forest that are difficult to quantify by other means.

c) Mean Annual Concentrations of Dissolved Species in Precipitation at the West Point NADP Collection Site

A precipitation chemistry collection site for the National Acid Deposition Program was initially established at West Point in the fall of 1979. A new site, at a nearby location, was chosen in the fall of 1983, and collection at the first location was ended about a year later at the end of the summer of 1984, providing an overlap at the two sites of about one year. We have compared data from both sites, and combined them into a single time-series of annual (calendar year) values, using the initial site (#51) through the end of calendar year 1993, and the second site (#99) subsequent to 1993, to generate a single set of "average" annual precipitation chemistry values for a period of 12 years (1980 through 1991). Mean annual values were obtained by multiplying individual concentration measurements by precipitation amount for each weekly period, and thus are a flux-averaged concentrations for an entire year.

These data were originally reported in terms of mass per unit volume units (mg/l), and are listed in the upper part of Table 1. Mean pH (unweighted by amount variation between years) for this 12 year period was 4.27, and there does not appear to be a significant net positive or negative trend with time in the acidity of rain and snow in the BRF region. The mass data were converted to molar charge units (equivalents) to permit simple comparisons of the various anion and cation concentrations (Table 1, lower half). Of the total measured cations (82 microequivalents), hydrogen accounted for an average of 54 ueq/l, or about 66% of the total measured cations. Clearly average precipitation at BRF can be described as "acidic". If the mean annual values of hydrogen ion concentrations are plotted as a function of year, a linear fit to the data has a slope which does not differ from zero (Figure 1). The period of 1982-84, and especially 1983 fall appreciably below (less acidic) the general trend. One interesting question to explore would be to examine, from weekly precipitation chemistry data, what systematic differences that group of years may have had from the remainder of the 12-year period. It is not likely that those years had a higher proportion of storm tracks that entered from the Atlantic, as opposed to more polluted

continental regions to the west, since there was not an unusually high mean concentration of chloride for those years.

Throughout the period of 12 years, sulfate equivalents were about twice that for nitrate (Figure 2). There may have been a slight decrease in sulfate concentrations since 1980, but such a trend does not appear to be statistically significant. With the exception of 1981, mean annual concentrations of sodium and chloride in precipitation at BRF have been reasonably constant (Figure 3), indicating that the influx of marine-derived chemical species does not appear to vary dramatically from one year to the next. Thus we should be able to use the 12-year average values of precipitation chloride concentrations as an initial estimate of influx of this ion to BRF by wet deposition in any given year.

The temporal trends in mean annual precipitation concentration at West Point for the three major anions illustrate some interesting systematic variations (Figure 4). The changes from one year to the next in sulfate and nitrate have been very similar. Since these ions provide the primary charge balance for hydrogen ions in precipitation at BRF, this suggests that sources of acidity from nitrogen and sulfur emissions are quite well integrated before advection to our region. The increases and decreases in sulfate and nitrate are not reflected in an opposite trend in chloride, as might be expected for a two end-member system (polluted continental precipitation and "clean" marine precipitation). As an initial model for discussion, one could consider three general types of precipitation: (1) high acidity continental; (2) clean continental; and (3) clean marine. A similar conclusion can be drawn from plots of monthly precipitation concentrations of nitrate or sulfate vs chloride (Figure 5).

One issue of considerable interest is the relative contribution of anthropogenic sulfate and nitrate acidity at BRF, since the former is likely to decrease in the future as sulfur emissions controls at electricity generation facilities evolve, while the latter is likely to increase because technical solutions to reduce nitrate emissions are so much more difficult to employ. If mean annual sulfate and nitrate concentrations at West Point are examined, the total equivalents of nitrate is about 0.51 times that for sulfate [Table 1: 25.8/50.1 (microequivalents per liter)]. However this "bulk" ratio could be somewhat misleading because it neglects consideration of the natural levels of these two ions in precipitation. Another approach to this question can be made by plotting mean annual nitrate vs sulfate equivalents (Figure 6), which yields a slope of about 0.46. Assuming that the primary cause of variations in mean

annual concentrations of these ions at West Point are from interannual differences in the proportion of precipitation derived from polluted continental air masses vs the other two "end-members" mentioned above, this also indicates that sulfate sources of acidity are about twice as important as nitrate sources at BRF. If monthly mean concentrations of nitrate and sulfate are compared for the period 1979-1992, the slope nitrate relative to sulfate is somewhat higher (Figure 7, 0.61 in microequivalents units) than for the annual mean data.

Clearly the suggested conclusions given above should be considered as only very preliminary, but they do indicate that the large quantity of data obtained for the NADP collection site at West Point represents a major source of information for ecosystem studies at BRF. These data have now been obtained for well over a decade and can provide a valuable resource for many purposes.

d) Budget for Chloride Ion in Aleck Meadow Drainage Basin

If we assume that the 12-year mean annual concentration of chloride in precipitation at West Point (Table 1, 1980-1991) is representative of the average delivery of this ion to BRF by wet deposition, then we can compare observed surface water concentrations with data for precipitation to obtain some insights about the integrated impact of at least two other processes of major importance: (1) dry deposition of chloride ion to the forest canopy and (2) increases in dissolved ion concentrations in surface waters above that delivered in precipitation due to evaporation and transpiration losses of water vapor to the atmosphere. The second of these processes is possible to derive from careful measurements of water budget parameters by stream and precipitation gauging, but it likely to take a number of years before that is feasible for all watersheds at BRF. The first process, however, is probably only possible to derive from chemical budgets for "unreactive" ions such as chloride. At this stage, we can only consider the integrated effects of both of these processes.

Considering the large range of monthly mean chloride concentrations in precipitation, as measured at West Point (Figure 5), the variations in chloride for the surface water leaving the Alec Meadow catchment were very small for a five month period in 1991-92 (Figure 8). There appears to have been a slight increase in chloride from the beginning of this period in the autumn (40 microequivalents/liter) to the following spring (50

microequivalents/liter), but the changes from one week to the next were quite small. As an initial approximation, we can take the mean of these data as indicative of average annual surface water runoff chloride concentrations at BRF. This value (45.2 uequiv/l) is much higher than for West Point precipitation (11.8 uequiv/l).

From the known geological history of BRF, it is unlikely that there are significant source of dissolved chloride from local chemical weathering. Given the very low tendency for dissolved chloride to bind to soil minerals, it is also not likely that appreciable fractions of incoming chloride are being retained "permanently" in the basin. If we assume that chloride can be considered as a "conservative" tracer in BRF, then the combined effects of dry deposition and evapotranspiration are almost a factor of 4 (3.83). Although this assumption needs further examination and testing, it is not likely to be greatly in error. Since the loss of water vapor to the atmosphere from BRF is unlikely to be more than half of the precipitation influx, this suggests that dry deposition of chloride could be approximately equal to wet deposition, requiring that total ion influx estimates from the atmosphere based on extrapolation of the West Point NADP site data be increased appreciably, by perhaps a factor of two.

For the northeastern USA region in general, it is very difficult to use chloride as a tracer for dry deposition influxes in many areas because of the massive amount of road salt that are added to large surface water catchments. In Rockland County, the increase of chloride in streams feeding the reservoir system of the Spring Valley Water Company has been more than an order of magnitude over the past half a century. From the geography of the BRF catchment feeding Aleck Meadow Reservoir, there are no plausible influxes of this type to influence the chloride budget.

Although dry deposition contributions to chloride influxes may not be extrapolated directly to species that have more chemical reactivity with vegetation, such as nitrogen and sulfur gases in the atmosphere, the general indication that dry deposition to BRF is likely to be a major influx should be accurate. We need a much longer time-series of surface water measurements of chloride at BRF, and to examine seasonal variability in precipitation chloride for the West Point data, but the approach outlined here looks quite promising to pursue to quantify important ecosystem processes that have no other plausible method to measure directly.

e) Budget for Dissolved Silicate in Aleck Meadow Drainage Basin

In contrast to chloride, the budget for which should be dominated by delivery from the atmosphere, mixing within the soils and surface waters in BRF and export in solution with no significant influxes from chemical weathering, dissolved silicate should be derived almost exclusively from this latter process. Precipitation contains almost no dissolved silicate. Considering the very small week to week variability of dissolved chloride in the Aleck Meadow system discharge, dissolved silicate had substantial variations (Figure 8). Although it is possible that these are analytical errors in the data, the variations do appear to be systematic, with large excursions on the time-scale of about one month. The mean molar concentration of silicate over a period of five months was similar to that for chloride, but weekly values varied over a range of more than a factor of four. If such variations are observed for a more extensive time-series of data, it may be feasible to examine other important drainage basin properties. One possible mechanism that could lower dissolved silicate would be uptake by planktonic diatom communities in the ponds and swamps, as has been observed in many areas for lakes. Silicate should be an interesting tracer of biological processes at BRF because its removal should be primarily by aquatic communities, while nitrogen is more likely to be dominated by the terrestrial plants.

If the large excursions of dissolved silicate in stream waters from the Aleck Meadow catchment are real, they indicate that if changes in relative contributions of ponded surface water and soil water discharges are involved in silicate removal budgets, these do not generate significant changes in dissolved chloride fluxes out of the system.

f) Other Trends in Concentrations of Dissolved Species in Precipitation at the West Point NADP Collection Site

We mention here a few other issues of relevance to BRF studies that are based on the West Point wet deposition data. If we plot a time series of mean annual concentrations for all the major cations (Figure 9), the dominance of hydrogen ion is very apparent. The two cations in highest abundance after hydrogen are sodium and ammonia, each of which is about a factor of five lower. The year with highest sodium (1981) was also highest in magnesium, consistent with a higher proportion of marine sources of precipitation in that year.

If the two dominant atmospheric nitrogen species, ammonia and nitrate are plotted against each other, a linear fit to the data yields a slope of

about 0.37 (Figure 10). Thus for each increment of nitrate delivered in wet deposition, an additional fixed nitrogen supply of about one third arrives at BRF. The slope from these data has a relatively low correlation ($r^2 = 0.42$), suggesting that it would be worth further examination of the covariation of ammonia and nitrate in monthly precipitation data at West Point. In contrast, the linear correlation coefficient ($r^2 = 0.61$) for mean annual nitrate and sulfate concentration was appreciably higher (Figure 6).

g) Proposed Work During 4/1/94 - 3/31/95

As indicated in the report given above on changes in sampling strategy, our plan is to put more emphasis on analyses of stream samples in the coming year. Analytical equipment to measure chloride and silicate are available at LDEO currently, and our intention would be to work initially on those species, along with electrical conductivity, a very simple lab measurement. These data would permit some important issues about dry deposition flux estimates and hydrologic budgets at BRF to be addressed in the short term.

As discussed in our proposal from last year, we are having difficulties locating ion chromatography equipment that is accessible enough to permit students at Columbia and Barnard to make much progress on anion measurements, especially nitrate. This remains a high priority for the coming year, and we have hope that one of several options currently under investigation will prove practical.

A third priority will be to make cation measurements on a number of stream samples by atomic absorption spectrometry. This equipment is also currently available at LDEO, and should be feasible to use for students during the coming summer months.

h) Literature Cited

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West Point NAPD Station 1980-1991 Annual Means of Precipitation Chemistry

Stat Code	Year	Ca (mg/l)	Mg (mg/l)	K (mg/l)	Na (mg/l)	NH4 (mg/l)	NO3 (mg/l)	Cl (mg/l)	SO4 (mg/l)	pH	Conduct (uS/cm)	Ppt (cm)	
51	1980	0.12	0.039	0.020	0.274	0.18	1.67	0.38	2.81	4.21	28.3	108.40	
51	1981	0.15	0.088	0.030	0.468	0.22	1.72	0.88	2.86	4.21	35.1	117.68	
51	1982	0.11	0.037	0.016	0.135	0.16	1.41	0.25	2.29	4.32	27.0	115.67	
51	1983	0.08	0.047	0.023	0.271	0.12	1.16	0.48	1.83	4.40	22.3	184.90	
99	1984	0.10	0.047	0.034	0.215	0.17	1.51	0.42	2.11	4.32	26.1	137.59	
99	1985	0.10	0.044	0.017	0.145	0.22	1.89	0.34	2.55	4.22	31.5	108.09	
99	1986	0.09	0.036	0.016	0.142	0.20	2.04	0.36	2.74	4.22	31.7	123.10	
99	1987	0.07	0.033	0.018	0.170	0.15	1.51	0.38	2.28	4.27	27.5	128.30	
99	1988	0.10	0.030	0.012	0.147	0.13	1.53	0.34	2.38	4.26	26.8	108.59	
99	1989	0.06	0.029	0.022	0.160	0.23	1.69	0.41	2.25	4.27	28.1	144.70	
99	1990	0.07	0.036	0.016	0.226	0.21	1.45	0.49	2.23	4.28	27.4	147.99	
99	1991	0.11	0.025	0.042	0.128	0.22	1.59	0.31	2.51	4.24	29.5	121.11	
51+99	'80-'91	0.10	0.041	0.022	0.207	0.18	1.60	0.42	2.40	4.27	28.4	128.84	
		(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	(ueq/l)	sum pos	sum neg	net +/-
51	1980	6.00	3.20	0.51	11.91	10.00	26.94	10.70	58.54	61.65	93.27	96.18	-2.91
51	1981	7.50	7.21	0.77	20.35	12.22	27.74	24.79	59.58	61.65	109.70	112.11	-2.41
51	1982	5.50	3.03	0.41	5.87	8.89	22.74	7.04	47.71	47.86	71.56	77.49	-5.93
51	1983	4.00	3.85	0.59	11.78	6.67	18.71	13.52	38.13	39.81	66.70	70.36	-3.66
99	1984	5.00	3.85	0.87	9.35	9.44	24.35	11.83	43.96	47.86	76.37	80.14	-3.77
99	1985	5.00	3.61	0.43	6.30	12.22	30.48	9.58	53.13	60.26	87.83	93.19	-5.36
99	1986	4.50	2.95	0.41	6.17	11.11	32.90	10.14	57.08	60.26	85.41	100.13	-14.72
99	1987	3.50	2.70	0.46	7.39	8.33	24.35	10.70	47.50	53.70	76.09	82.56	-6.47
99	1988	5.00	2.46	0.31	6.39	7.22	24.68	9.58	49.58	54.95	76.33	83.84	-7.51
99	1989	3.00	2.38	0.56	6.96	12.78	27.26	11.55	46.88	53.70	79.37	85.68	-6.31
99	1990	3.50	2.95	0.41	9.83	11.67	23.39	13.80	46.46	52.48	80.83	83.65	-2.82
99	1991	5.50	2.05	1.07	5.57	12.22	25.65	8.73	52.29	57.54	83.95	86.67	-2.72
51+99	'80-'91	4.83	3.35	0.57	8.99	10.23	25.77	11.83	50.07	53.70	81.67	87.67	-5.99

21

Fig 1

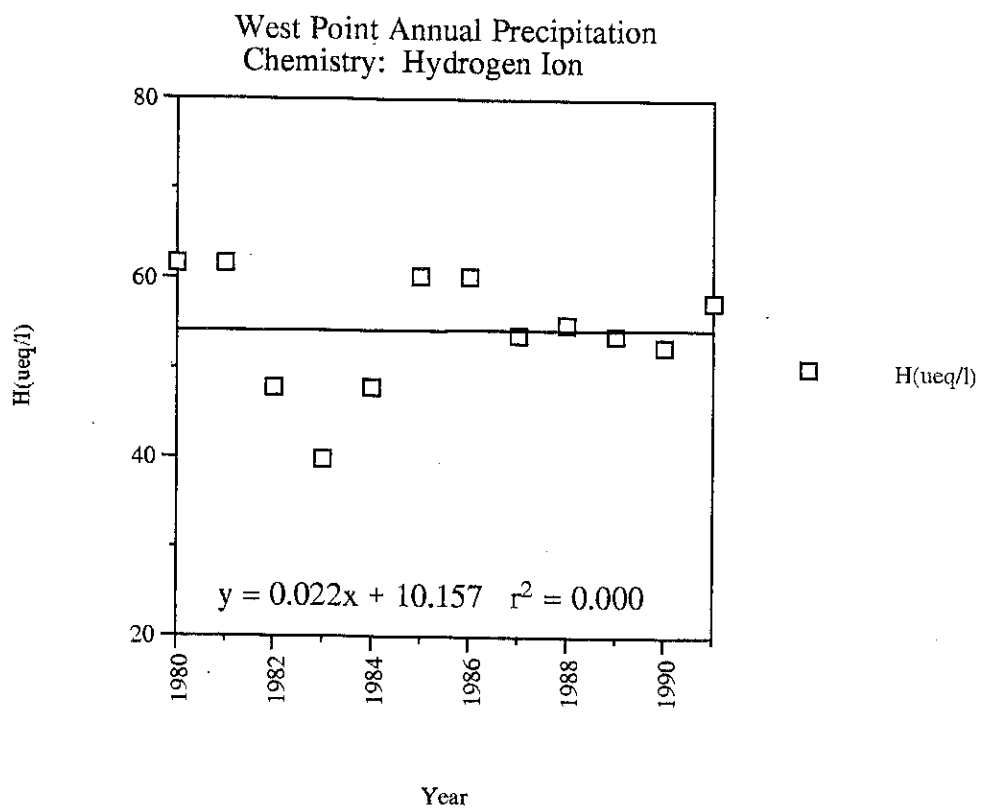
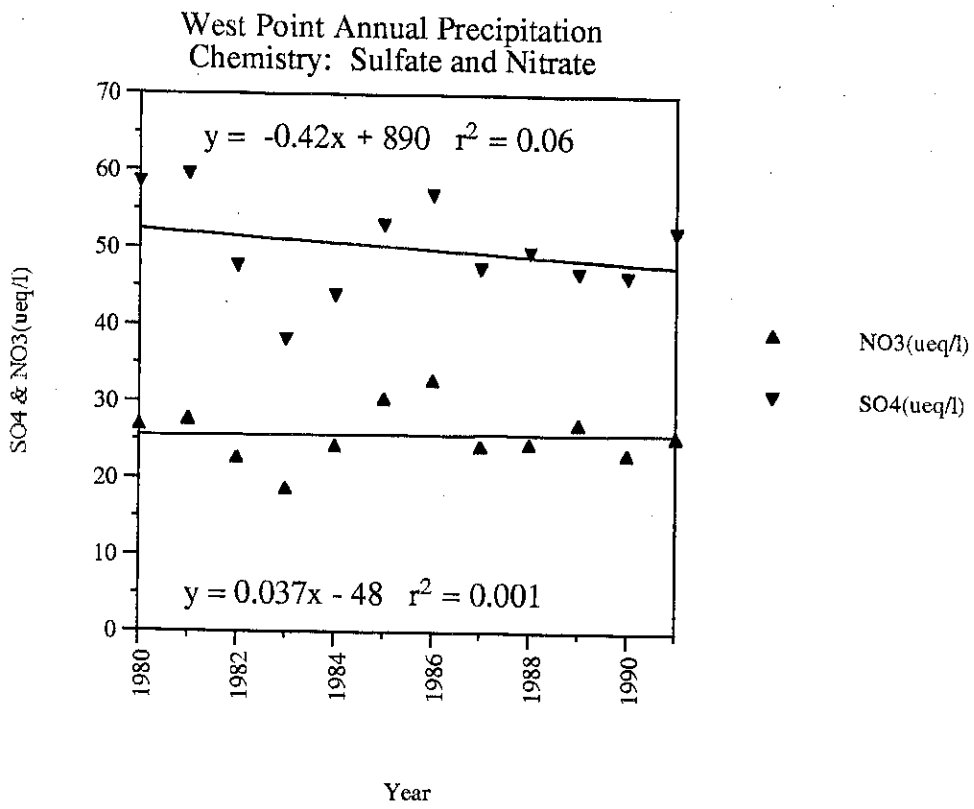
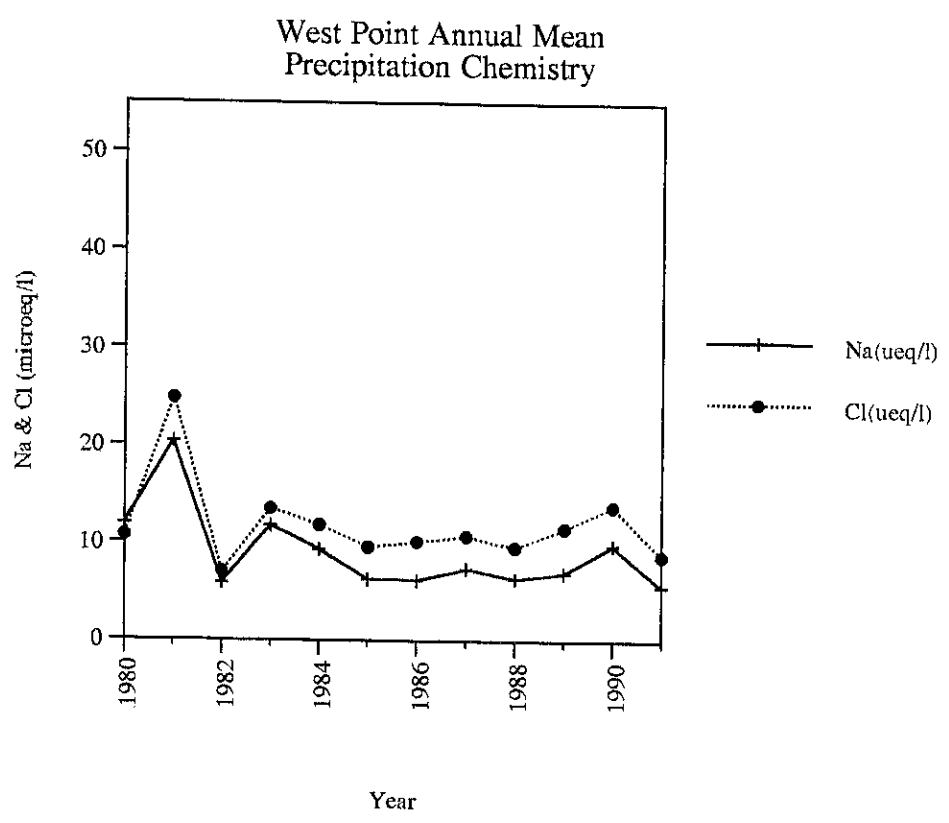
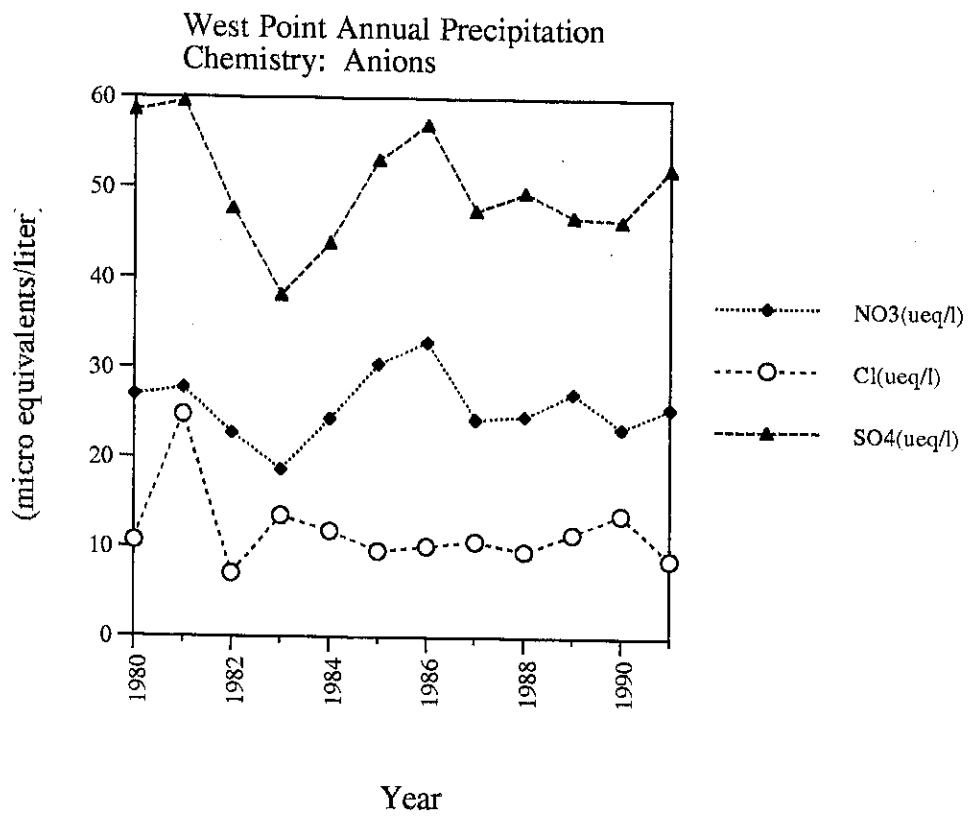


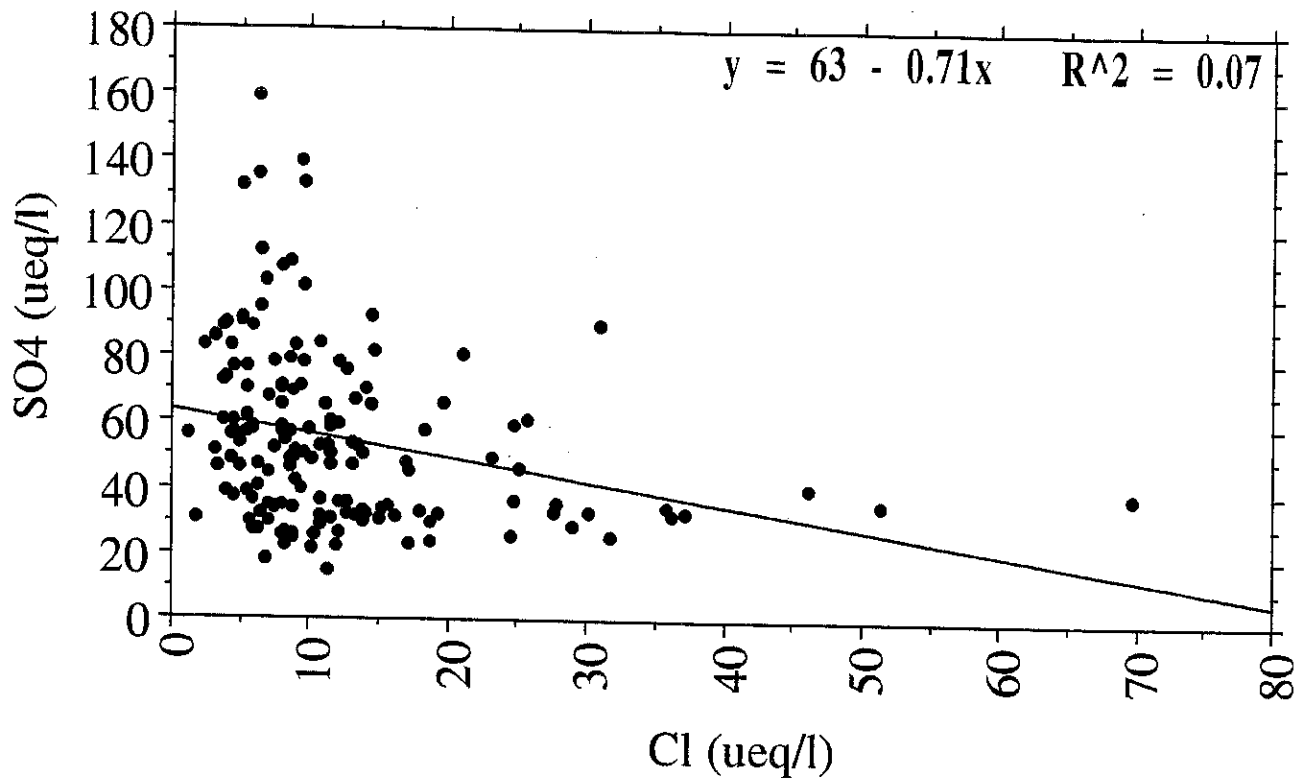
Fig 2







NADP West Point Monthly Precip Anions
(1979-92)



NADP West Point Monthly Precip Anions
(1979-92)

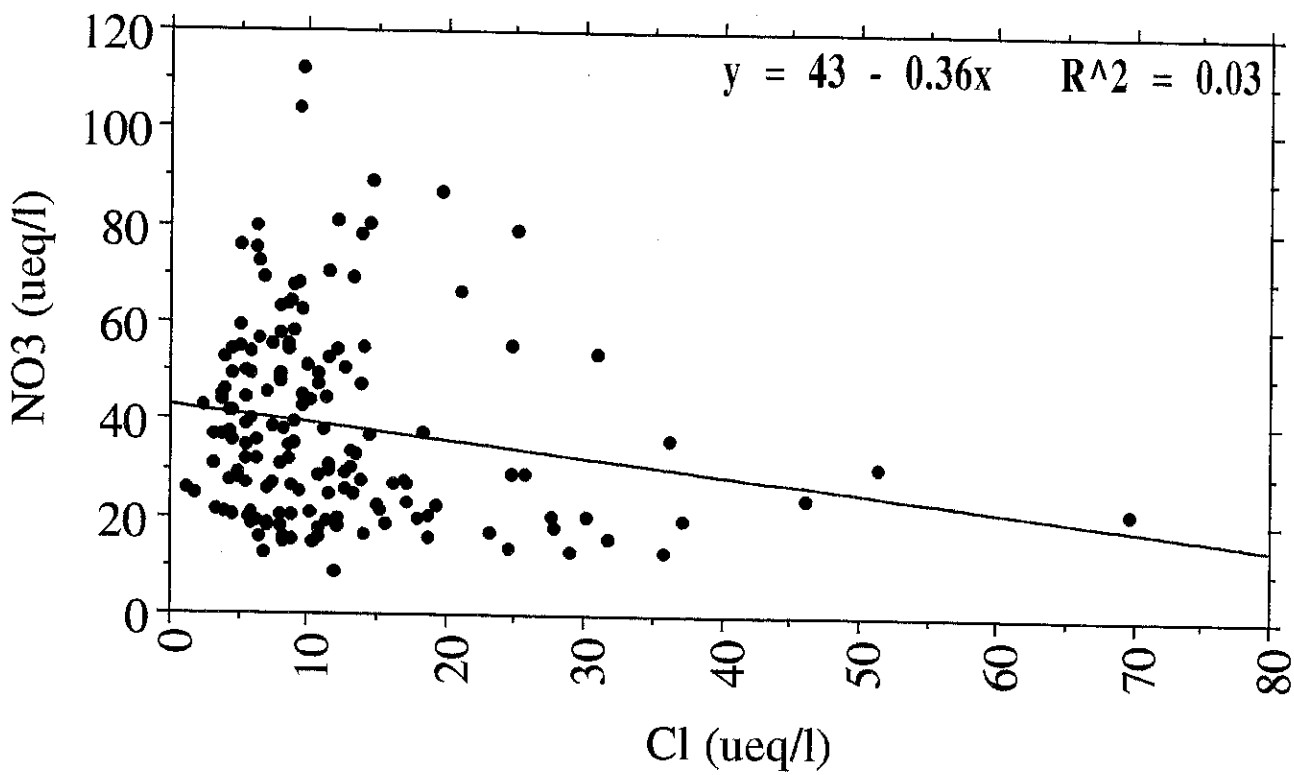


Fig 6

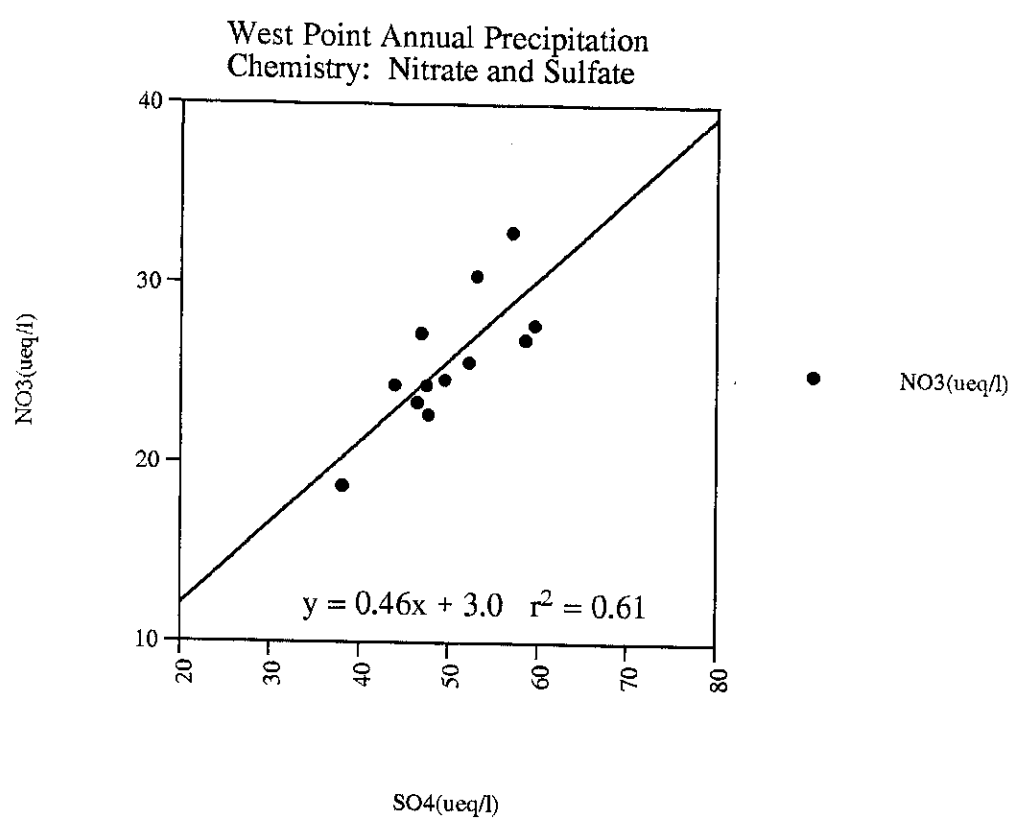


Fig 7

NADP West Point Monthly Precip Anions (1979-92)

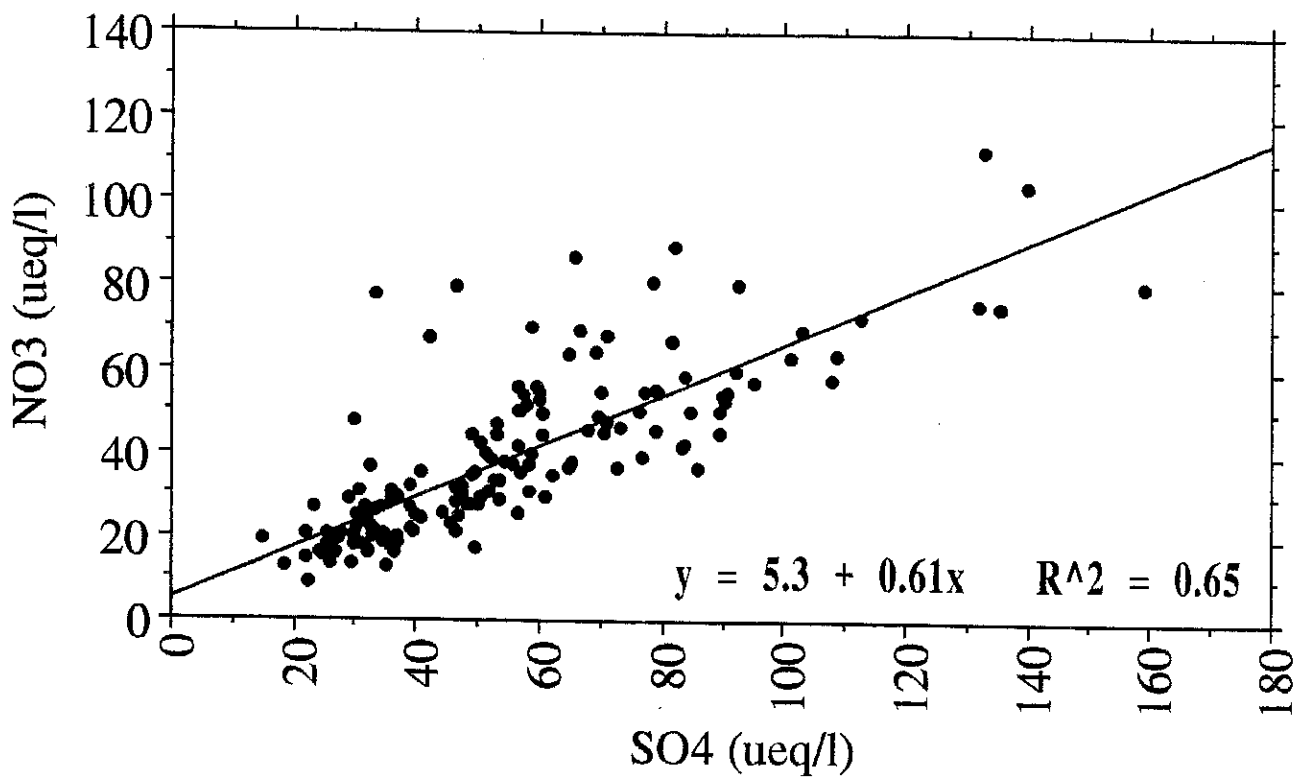
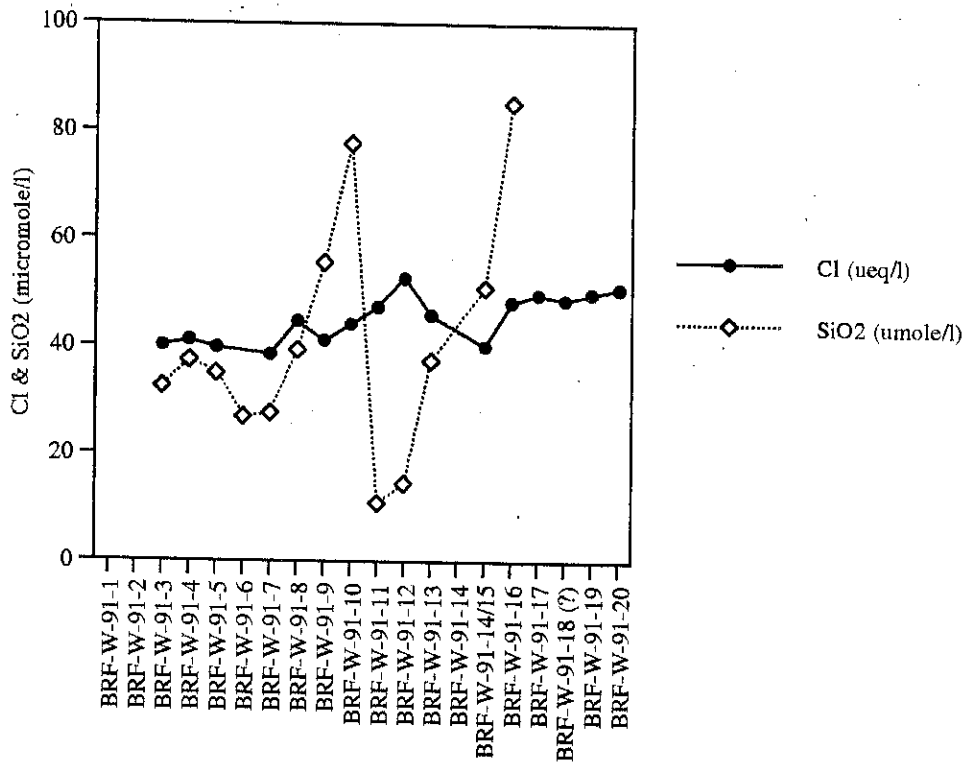
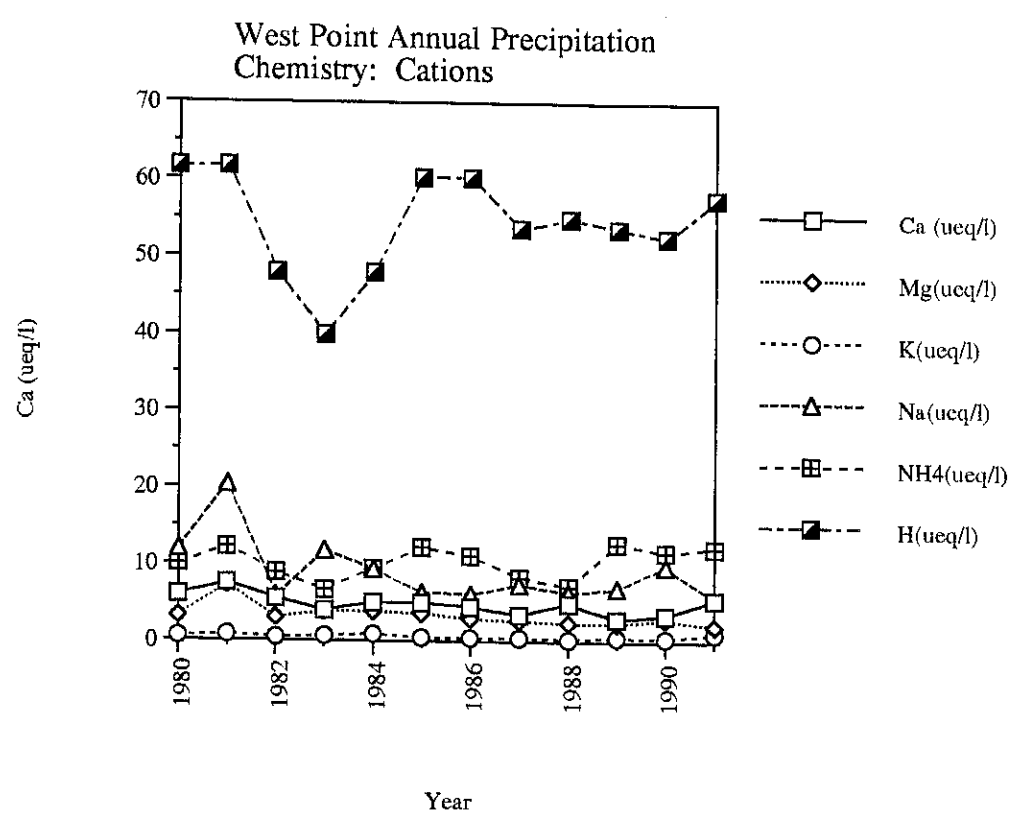


Fig 6
BRF Stream Chemistry





West Point Annual Precipitation
Chemistry: Nitrate and Ammonia

