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ACID RAIN MONITORING IN EAST-CENTRAL FLORIDA FROM 1977 TO THE PRESENT

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ABSTRACT

Rainfall has been collected on the University of Central Florida campus and at the Kennedy Space Center over a twelve year period. The chemical composition has been determined and summarized by month, annual periods and for the entire twelve year record at both locations. The weighted average pH at each site is 4.58; however, annual weighted average pH has been equal to or above the twelve year average during six of the past eight years. Nitrate concentrations have increased slightly during recent years while excess sulfate concentrations have remained below the twelve year weighted average during six of the past seven years. Stepwise regression suggests that sulfate, nitrate, ammonium ion and calcium play major roles in the description of rainwater acidity. Annual acid deposition and annual rainfall have varied from 20 to 50 meq m^{-2} yr^{-1} and 100 to 180 cm yr^{-1}, respectively. Sea salt comprises at least 25% of the total ionic composition.

INTRODUCTION

During the past two decades an increasing interest and concern about acid rain has prompted extensive research and monitoring activities which have been designed to provide a better understanding of cause and effect processes and to measure current levels of acid rain. During the mid 1970's the National Aeronautics and Space Administration (NASA) funded an extensive environmental monitoring program (Madsen, 1979) which included a substantial acid rain monitoring component. That program provides the initial basis for findings summarized in this study. The University of Central Florida (UCF) provided technical assistance for the program and was responsible for monitoring activities. A multi-site network was established at and near the Kennedy Space Center (KSC) and one additional site was established on the UCF campus located near Orlando, FL. These activities continued until October, 1981 when KSC monitoring responsibilities were absorbed by other NASA contractors and the number of operational sites was reduced. The site at UCF was retained to be operated by UCF personnel. In late 1983, the primary KSC site was converted to be operated as a National Atmospheric Deposition Program (NADP) site. Several contract reports (Madsen, 1981a; Madsen, 1985) and two NASA Technical Memoranda (Madsen et al., 1986; Madsen et al., 1989) have been written which address various aspects of the monitoring effort. The UCF site has now been operated in continuous fashion for more than twelve years. One KSC site, whose location has been changed only slightly, has also been operated over the same time period. Unfortunately monitoring data from the KSC site for the period November, 1981 to late 1983 have been determined to be of lesser quality than that of other data (Madsen et al., 1986), and therefore, cause a discontinuity in the KSC monitoring record.

METHODS AND PROCEDURES

Sampling Locations

Site locations and periods of operation as they pertain to data and results described in this report are presented in Figure 1. The UCF site has been located on the roof of the chemistry building during most of the study period. During the initial phase of the study, the site was located 0.9 km south of the current site in a grassy field near the UCF Physical Plant Headquarters and was relocated to the roof of the chemistry building in June 1979. During September and October, 1982 and again in July and August, 1989 the collector was located on the roof of the UCF biology building located 70 m east of the chemistry building. Rainfall measurements have been made for several decades at the National Weather Service (NWS) office which is located 21 km south-southwest of the UCF campus at the Orlando International Airport. The USC campus is located at 28°35'59" N latitude and 81°12'00" W longitude. The KSC site 13, which operated during 1977-1981, was located at 28°31'48" N latitude and 80°39'44" W longitude. The KSC NADP site, which has operated from late 1983 to the present, is located 2.3 km northeast of site 13 at 28°32'33" N and 80°38'39" W.

Sample Collector and Collection Interval

Aerochem Metrics wet-dry collectors have been used exclusively for collection of rainwater samples. Collection intervals have typically been 24 hr. periods ending on Tuesdays through Friday and 72 hr. periods ending on Mondays for samples collected at UCF. Similar collection intervals were used at KSC prior to establishment of the NADP site. The NADP site sampling interval length is seven days ending on Tuesday mornings. The KSC site has been operated with this seven day interval since late 1983.

Sample Handling and Chemical Analysis

The protocol for working with collected samples has been described (Madsen, 1981b). Methods for the measurement of the major ion chemical composition of rainwater samples as part of the UCF/KSC program and the NADP program have been described (Madsen, 1981c; NADP, 1984). Methods employed are summarized in Table 1. Data quality has been evaluated by examining a number of diagnostic ratios (e.g. anion/cation ratio, measured/calculated conductivity, Cl/Na and Na/Mg) on an individual sample basis (Galloway and Likens, 1978).

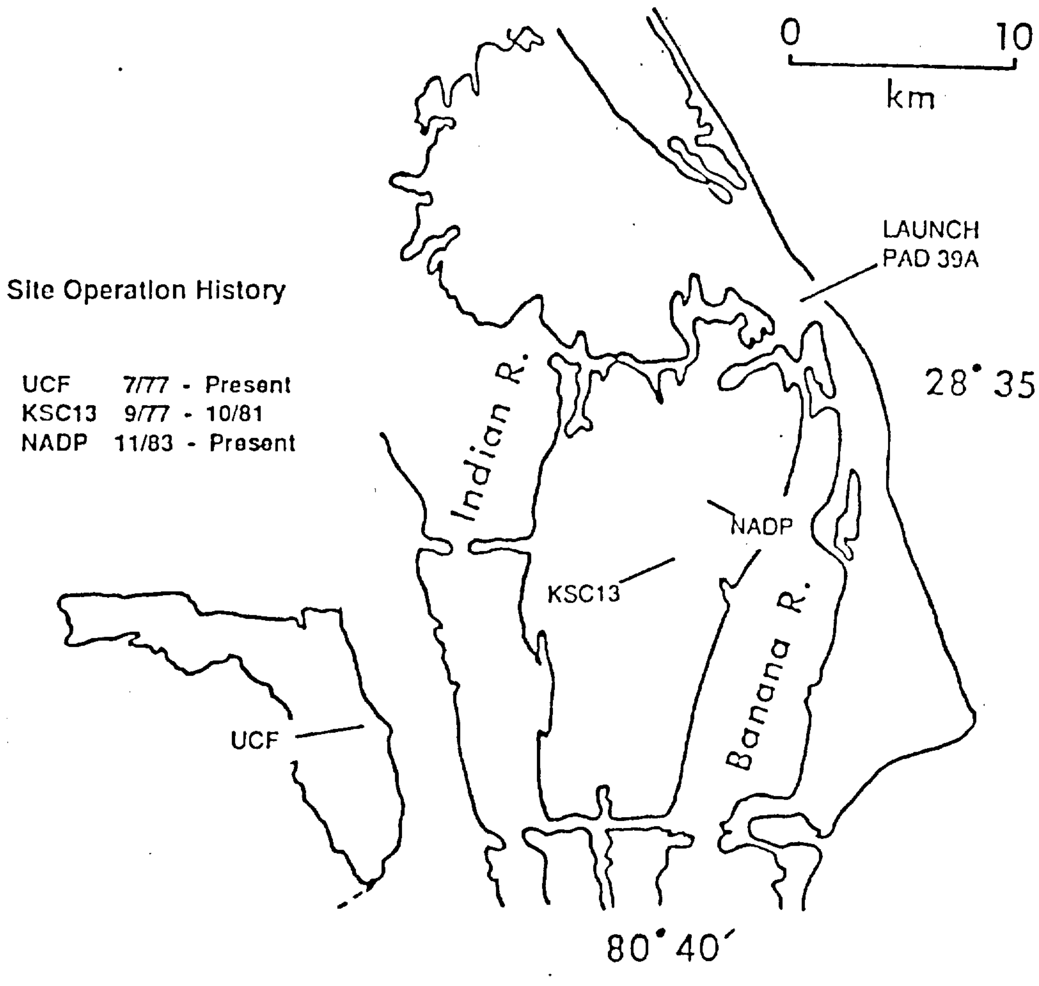


Figure 1. Rainfall Collection Sites in East Central Florida.

Table 1. Methods of Chemical Analysis.

Chemical Species	UCF/KSC Method	NADP Method
pH	glass electrode	glass electrode
Conductivity	electrode	electrode
Na	IC[1], FE[2]	AA
K	IC[1], FE[2]	AA
Ca	AA	AA
Mg	AA	AA
NH_4	IC[1], FIA[3]	TC
CL	IC[1], FIA[3]	IC
NO_3	IC[1], FIA[3]	IC
SO_4	IC[1], FIA[3]	IC

[1]IC Ion chromatography 11/77-10/81
[2]FE Flame emission spectroscopy 11/81-present
[3]FIA Flow injection analysis-Colorimetry 11/81-present
AA Atomic Absorption Spectrophotometry
TC Technicon Autoanalyzer Colorimetry

RESULTS AND DISCUSSION

Sample Collection History

Collection of rainfall on the UCF campus began in July 1977 and has continued in uninterrupted fashion. Only sample amount, pH and conductivity were measured until November 1977. Major cation and major anion concentrations as well as pH and conductivity have been measured since November 1977. Sample collection began at site 13 of the UCF/KSC network in late summer 1977. Measurement of all major chemical species, pH and conductivity started in November 1977 and were completed on samples collected at that site through the end of October 1981. Sampling for the NADP site commenced in the Fall of 1983. Results obtained for samples collected at these KSC sites during 1978-1981 and 1984-1989 are included here.

General Rainfall Composition

The general chemical composition of rainfall collected at UCF is summarized for the period 1978-1989 in Table 2. A similar summary for the KSC/NADP site for the periods 1978-1981 and 1984-1989 is also presented in Table 2. Several figures are used to display the annual volume weighted average composition and annual total deposition for several species determined in samples collected at UCF and KSC. Monthly volume weighted average composition and annual total deposition based on 12 years of continuous data for the UCF site and for the 10 years of data from the KSC/NADP site are also presented graphically. The presentation of monitoring data as annual or multi-year averages as included in Table 2 does not allow one to portray the variability in rainfall chemical composition that occurs on an individual sample or individual storm basis. It has been observed that concentrations of each of the major cations and anions present in rain collected at UCF and at KSC can vary over two to three orders of magnitude. The measured pH of more than 800 samples collected at UCF is summarized in Figure 2. The measured pH of more than 280 samples collected at KSC site 13 and more than 180 samples collected at the KSC NADP site are also presented in Figure 2. The median pH at both sites is 4.52 and the distribution of measured pH is quite similar at both sites. It should be emphasized that sampling at the NADP site occurs at weekly intervals while sampling at UCF and KSC site 13 typically occurred at one or three day intervals. The variability in major cation and anion concentrations in the UCF samples are also presented in Figure 2. All ions have similar distribution patterns. Marine salts, specifically sodium ion and chloride ion, show the greatest variability particularly at high concentrations. The most abundant anion is excess sulfate and the most abundant cation is hydrogen ion. Excess sulfate is measured total sulfate after correction for the presence of marine derived sulfate (Granat, 1972).

Measured sample acidity and conductivity are influenced to some degree by all ionic components present in each rain sample. Stepwise multiple regression has been used to evaluate the dependence of individual sample acidity upon the presence and concentration of major anions and cations. Independent variables considered in this treatment include sodium, potassium, calcium, magnesium, ammonium ion, chloride, nitrate and excess sulfate. Independent variables are presented in the order selected by the stepwise regression procedure. The model equations suggest that individual sample acidity is accounted for by the selected independent variables. The stepwise regression equations obtained from the 1978 to 1987 UCF and 1978 to 1981 plus 1984 to 1987 KSC data sets are presented below.

$$\begin{aligned} H(UCF) = {} & (0.86 \pm 0.03)xsSO_4 - (-0.84 \pm 0.04)Ca \\ & + (0.75 \pm 0.04)NO_3 - (0.41 \pm 0.04)NH_4 + 8.5, \\ & r^2 = 0.88 \end{aligned}$$

$$\begin{aligned} H(KSC) = {} & (0.89 \pm 0.01)xsSO_4 - (0.57 \pm 0.04)NH_4 \\ & + (0.55 \pm 0.02)NO_3 - (0.52 \pm 0.02)Ca \\ & + (0.13 \pm 0.02)Mg + 5.8,\ r^2 = 0.94 \end{aligned}$$

Similar stepwise regression equations are obtained for both sites. The influence upon acidity by

Table 2. Rainfall Composition at UCF and KSC.

	7/77-12/89[1] UCF[2]	10/77-10/81[1] KSC13[2]	1/84-12/89[1] NADP[2]
Number of Samples	1139	303	269
cm Rain/12 Months	139.0	115.0	121.3
Field Lab pH	4.58	4.53	4.71
Lab pH	—	—	4.83
Field Conductivity[3]	16.3	21.6	15.7
Lab Conductivity[3]	—	—	16.0
		TOTAL DEPOSITION (meq/m^2-yr)	
H	36.9	33.8	26.7
Lab H	---	—	20.1
Na	26.3	45.9	57.9
NH_4	12.6	13.2	7.0
NO_3	21.0	13.8	14.6
Excess SO_4	33.8	29.8	27.2
		IONIC CONCENTRATIONS ($\mu eq/l$)	
Field H	26.6	29.4	19.7
Lab H	—	—	14.8
Na	19.1	39.9	42.6
K	0.8	1.8	1.4
Ca	9.4	9.4	7.1
Mg	5.1	9.8	10.5
NH_4	9.2	1.4	5.1
Cl	22.7	41.5	51.0
NO_3	14.5	12.0	10.7
SO_4	26.8	33.9	25.1
Excess SO_4	24.6	29.6	20.0

[1] Time Period; [2] Site; [3] $\mu S/cm^2$

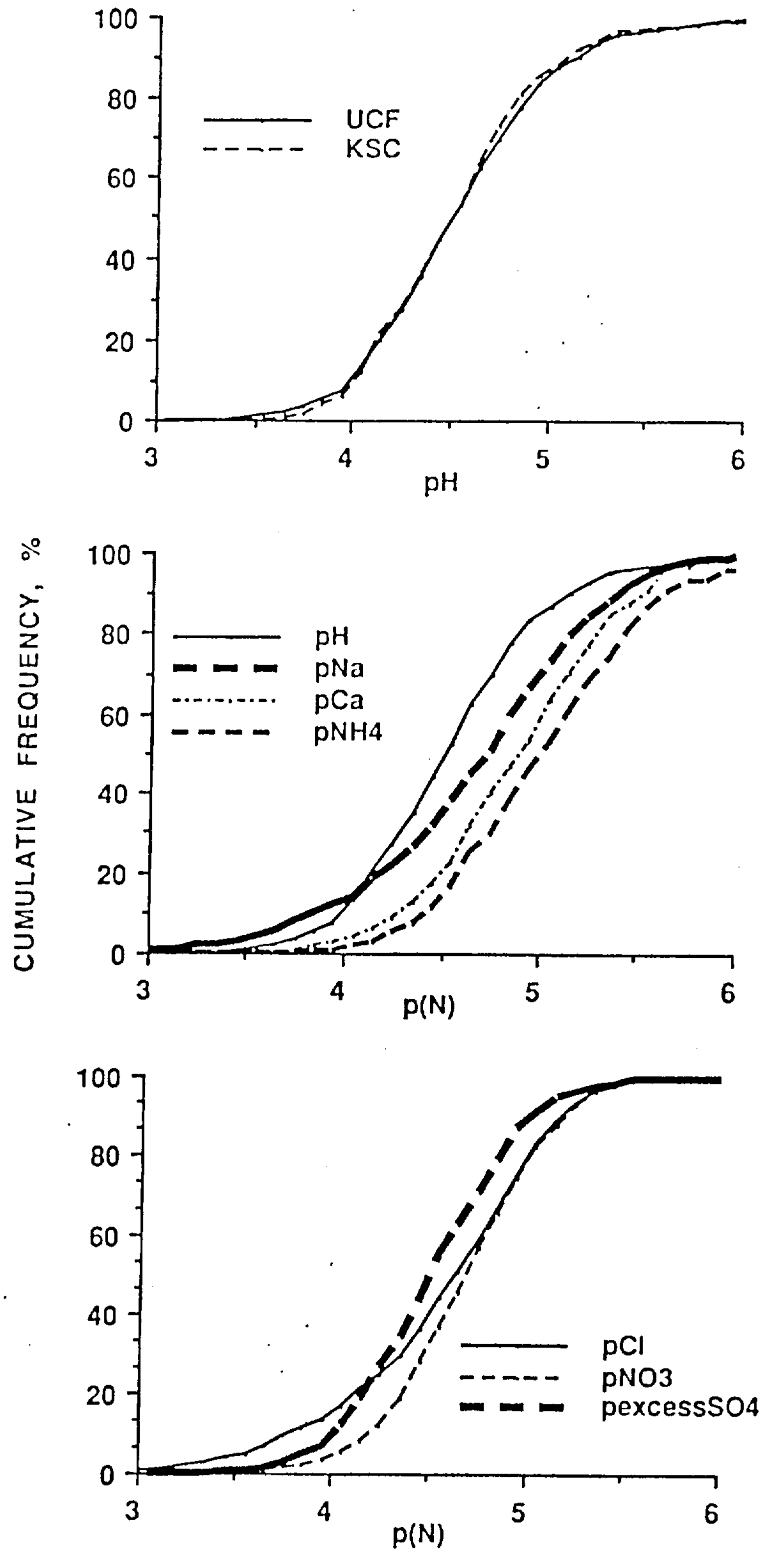


Figure 2. Individual Rain Sample Concentration Distributions.

excess sulfate and nitrate and the neutralizing cations calcium and ammonium ion is clearly illustrated. Graphical presentation of the relationship between measured individual sample acidity for samples collected at UCF during the period 1978 to 1987 and calculated acidity based on the obtained stepwise regression equation is shown in Figure 3. The amount of rainfall which falls within Florida is characterized by extreme variability. This variability occurs statewide and on a small regional basis. As an example consider the following measurements. Total rainfall for the month of August 1985 was recorded as 39.41 cm at UCF, 16.74 cm at the KSC/NADP site, and 29.54 cm at the Orlando NWS. Documentation of this variability is illustrated in Figure 4 where annual amounts of rainfall at UCF, at KSC and at the NWS office from 1978 to 1989 are compared. The 30 year NWS annual average of 121.6 cm is also shown. During 1978 to 1989 both the UCF and NWS sites received approximately 15% greater rainfall than the 30 year average while the KSC site received an amount equal to the 30 year average. In fact, the UCF site received less than the 30 year average only during 1980 and 1981. Total rainfall amounts can therefore have considerable influence on acidic deposition variability over small geographical distances. Seasonal variability in rainfall amounts is also quite high. Monthly average rainfall amount for the period 1978 to 1989 is shown in Figure 5. The summer months July through September provide considerably greater amounts of rain. In general, the amount of rain received at UCF has been greater than that received at KSC during the summer months while the opposite behavior has been observed during the cool weather months.

Rainfall Acidity

The volume weighted acidity of rain at both UCF and KSC since 1977 is 26 μN which represents a pH of 4.58. Annual weighted average hydrogen ion concentration and annual hydrogen ion deposition are presented in Figures 6 and 4 respectively. The acidity of rain was greatest early in the twelve year period but equal to or less than the twelve year average during six of the most recent eight years. The highest acidity was during 1978 at both sites and the lowest acidity occurred during 1984 at both sites. Annual deposition of acid has averaged 37 and 31 meq m^{-2} at UCF and KSC respectively during the 12 year and 10 year study periods. In general, deposition amount has been greatest early and late in the 12 year period with lesser amounts being observed during middle years. The year 1982 was an exception. Annual rainfall amounts have varied by a factor of nearly two as illustrated in Figure 4 and this variability exerts a substantial influence on deposition quantities. Deposition of acid during 1985 at UCF was about average in spite of the fact that pH was 0.08 units higher than average. Total acid deposition has been greater at UCF than at KSC in each year with the exception of 1987. The UCF site has received greater rainfall amounts in all years except 1987 which accounts for the differences in total acid deposition. Measured acidity at these sites is approximately one-half of that observed in the midwest and northeastern sections of the U. S. while deposition is somewhat greater than one-half the quantity that is received in the Northeast. This is attributed to the somewhat greater amounts of rain received in Florida. Acidity results, used as the basis for the discussion presented in the previous paragraph, are based on field measurements (Keene and Galloway, 1984). NADP laboratory measurements of acidity for samples collected at KSC during 1984 to 1987 are about 25% lower than the corresponding field acidity measurements (Madsen et al., 1989). Differences between field measured acidity and laboratory measured acidity are typically observed as part of all acid deposition measurements associated with the NADP program (Keene and Galloway, 1984) and the discrepancy has not been totally resolved.

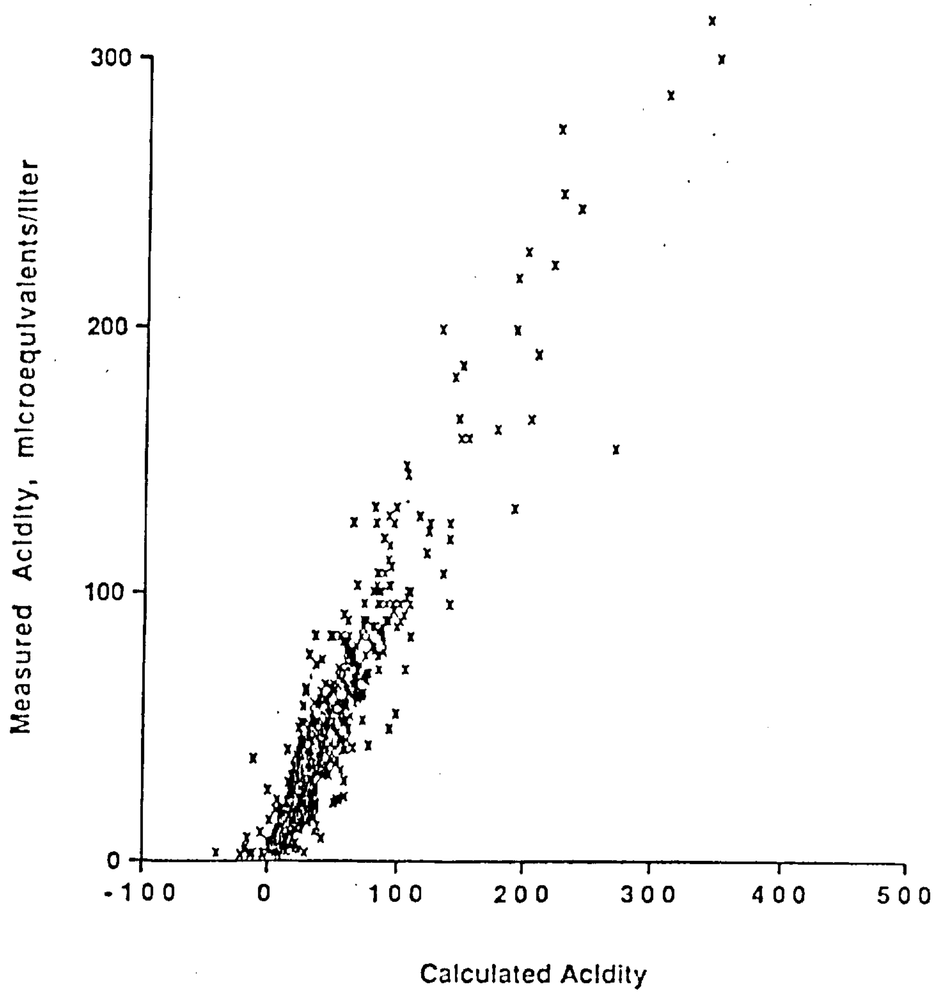


Figure 3. Relationship Between Measured Acidity and Calculated Acidity of Rain at UCF from Stepwise Regression.

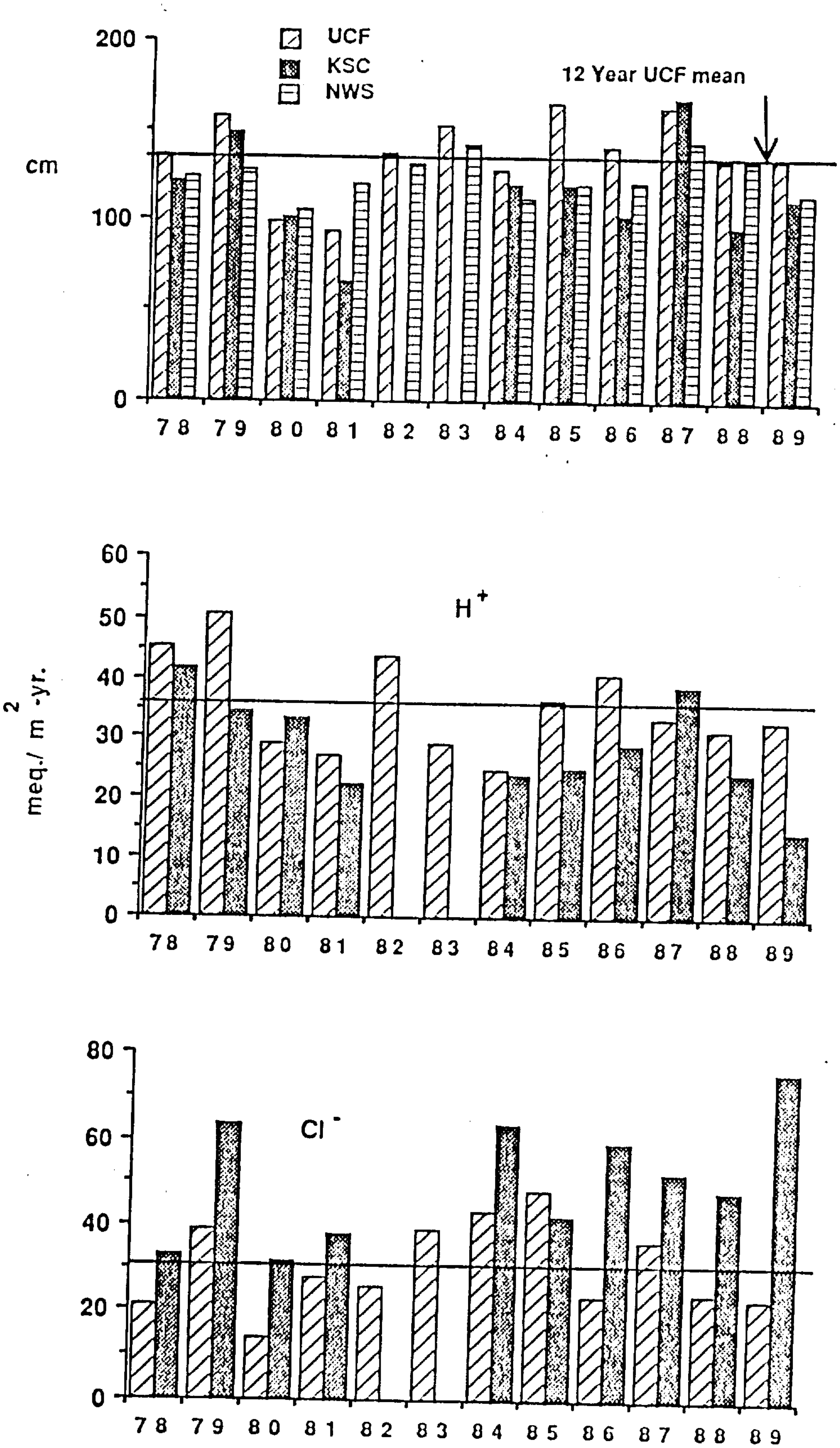


Figure 4. Annual Rainfall at UCF, KSC and NWS and Deposition at UCF and KSC.

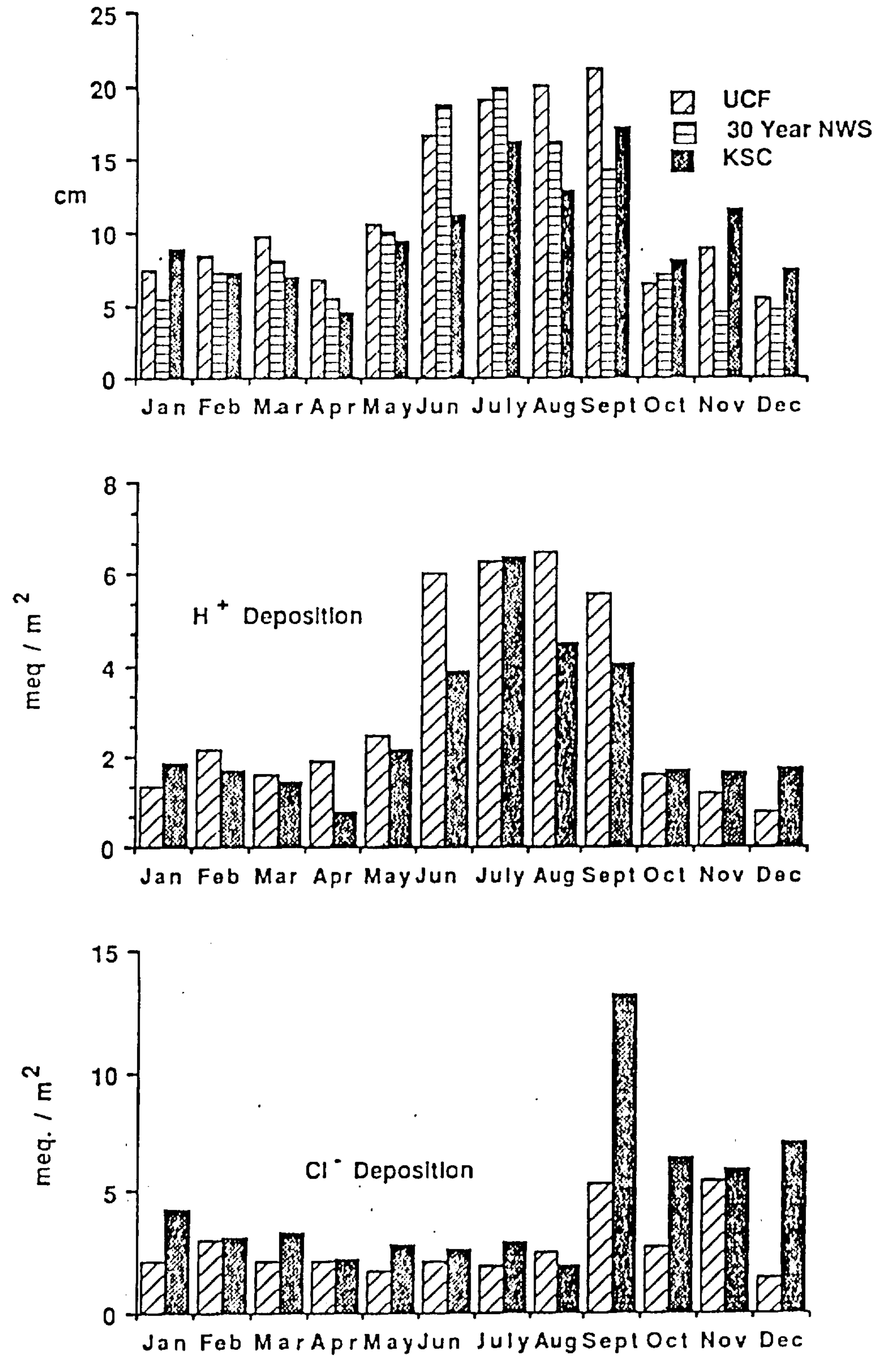


Figure 5. Monthly Rainfall and Deposition at UCF and KSC.

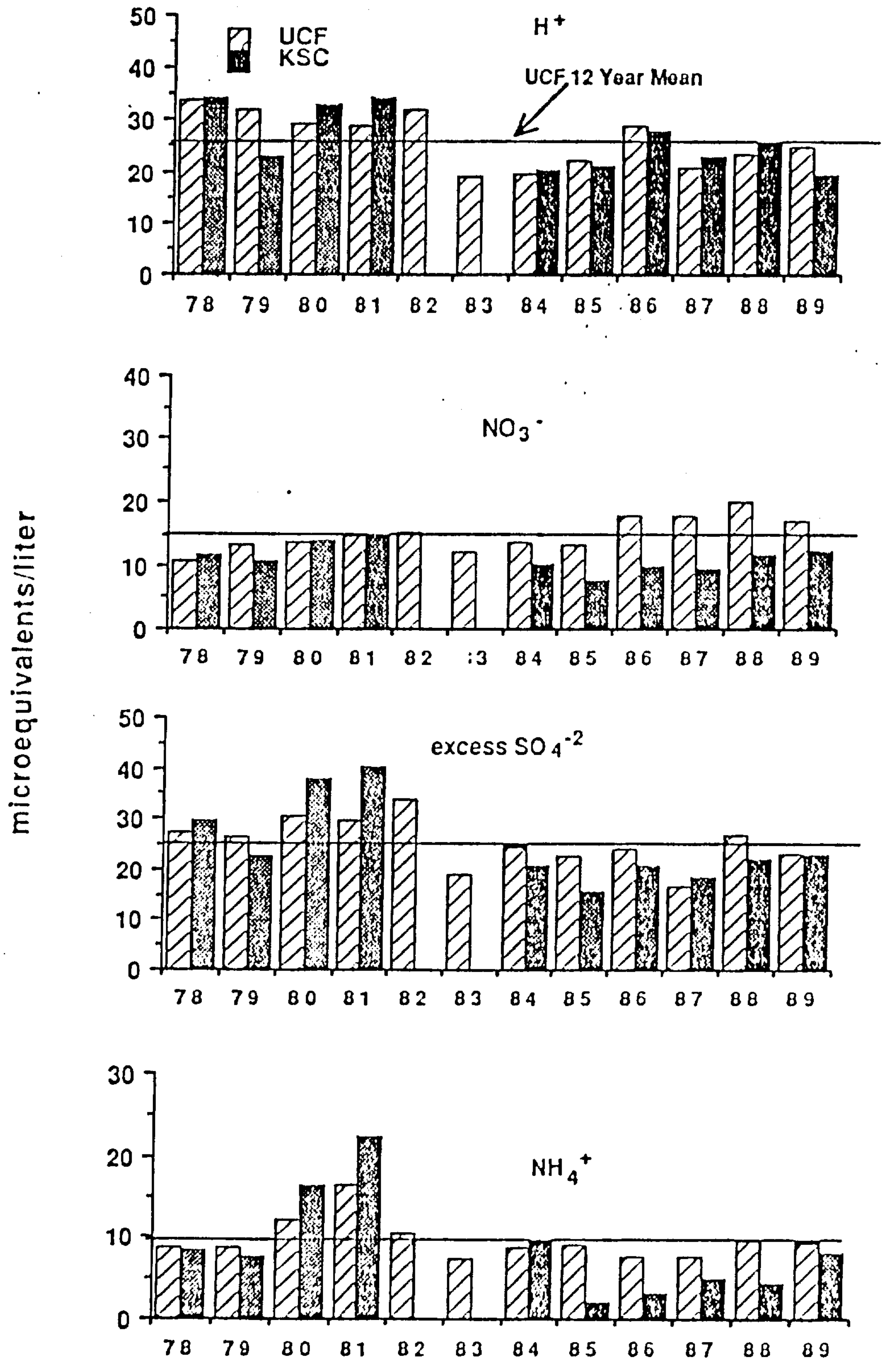


Figure 6. Annual Volume Weighted Mean Ion Concentrations in Rain at UCF and KSC.

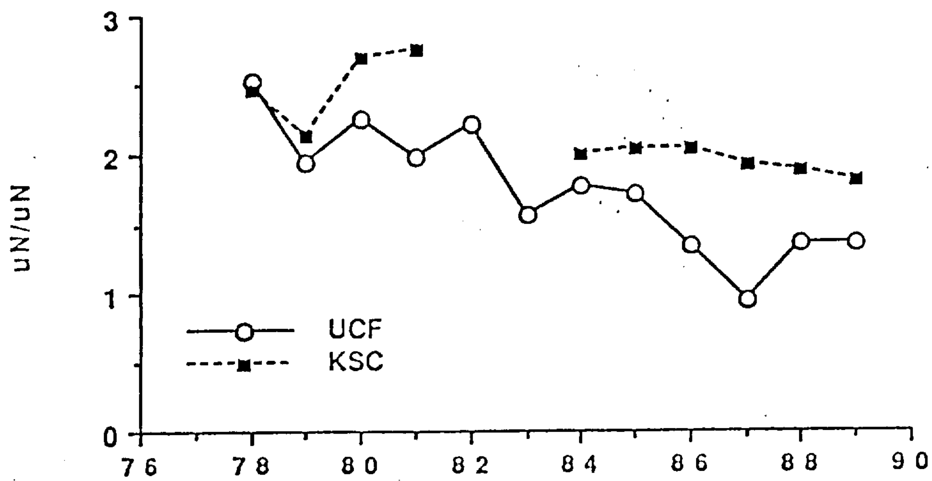
If appropriate quality control procedures are in place, then it is likely that the field acidity measurements will more correctly represent the acidity of rain because minimal time lag is associated with field measurements compared to lab measurements.

Anion Concentrations in Rain

The stepwise regression analysis that has been discussed previously suggests that nitrate and sulfate concentrations in rain are important predictors of sample acidity. These anions are present in rain in amounts that are more than sufficient to account for measured acidity. Excess sulfate is the most plentiful anion in rain collected at UCF and nitrate concentrations are somewhat smaller than chloride. It has been shown previously that the chloride concentrations typically observed in rain in East-Central Florida can be accounted for by marine influences associated with site proximity to the ocean (Madsen, 1984). Comparisons by year for excess sulfate and nitrate are shown in Figure 4. Chloride is the dominant anion observed in rain collected at KSC and nitrate ion is the least plentiful anion. The sulfate to nitrate ratio approaches a value of two in rainfall collected in the eastern U.S. (Dana and Easter, 1987). This ratio for rainfall collected at UCF and at KSC are summarized by year and by month in Figure 7. The ratio has shown a general decline over the period 1978 to 1989 and the decline is most apparent in samples collected at UCF. During 1986 to 1989 the ratio approached a value of one. When compared with acidity and nitrate levels over the same time period, it appears that the observed decrease in acidity is accounted for by the decrease in observed excess sulfate concentrations. When the excess sulfate to nitrate ratio is considered by month, it is observed that the KSC site yields a consistently greater ratio than UCF site. Large extremes from month to month are not observed; however, lowest values are typically observed from May to October. Annual weighted average nitrate and excess sulfate concentrations for the 12 year period are approximately 30% and 4% greater respectively at UCF than at KSC. Figure 6 illustrates the 12 year pattern. Nitrate concentrations have remained relatively constant from year to year, however there seems to be a substantial increase in concentration at UCF during 1986 to 1989. Excess sulfate concentrations have decreased substantially from concentrations measured prior to 1983 at both KSC and UCF. A seasonal pattern is quite apparent when nitrate and excess sulfate concentrations for the 12 year period are summarized by month (Figure 8). Concentrations for both anions exhibit summertime highs which are double the concentrations that are measured during October through January. This observation is consistent with those made by others (Dana and Easter, 1987) who find that relatively high summertime acidity is accompanied by higher concentrations of nitrate and sulfate.

Ammonium Ion Concentrations in Rain

The monthly variations that are observed for acidity, nitrate and sulfate concentrations are characterized by summertime maxima. Monthly variations in ammonium ion concentration which is presented in Figure 8, are similar to those observed for the other chemical species. However, the maximum monthly concentrations appear two or three months earlier during April and May than do the maxima for nitrate and excess sulfate. The extreme ammonium ion concentration observed at UCF during April is similar to the April maximum extreme observed for excess sulfate. The presence of ammonium ion suggests the presence of strong acid (e.g. sulfuric acid) which has been neutralized by ammonia to form the ammonium ion. Annual



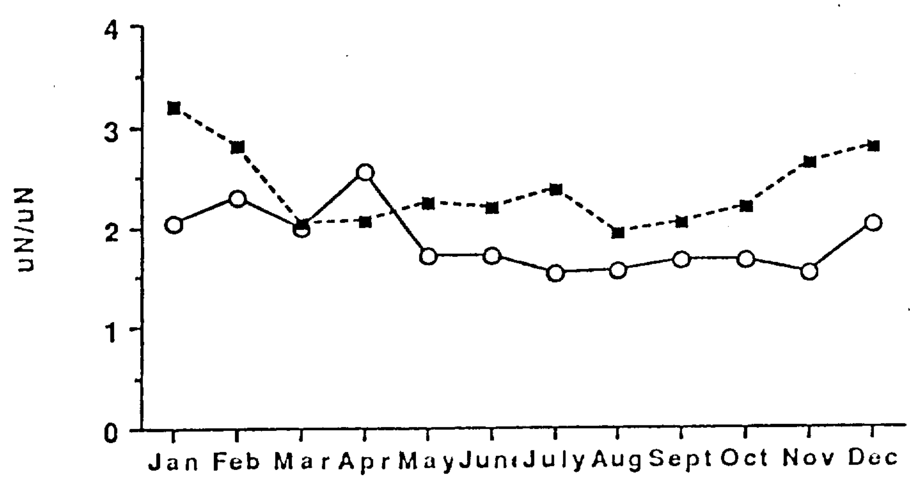


Figure 7. Annual and Monthly Excess Sulfate/Nitrate Concentration Ratios in Rain.

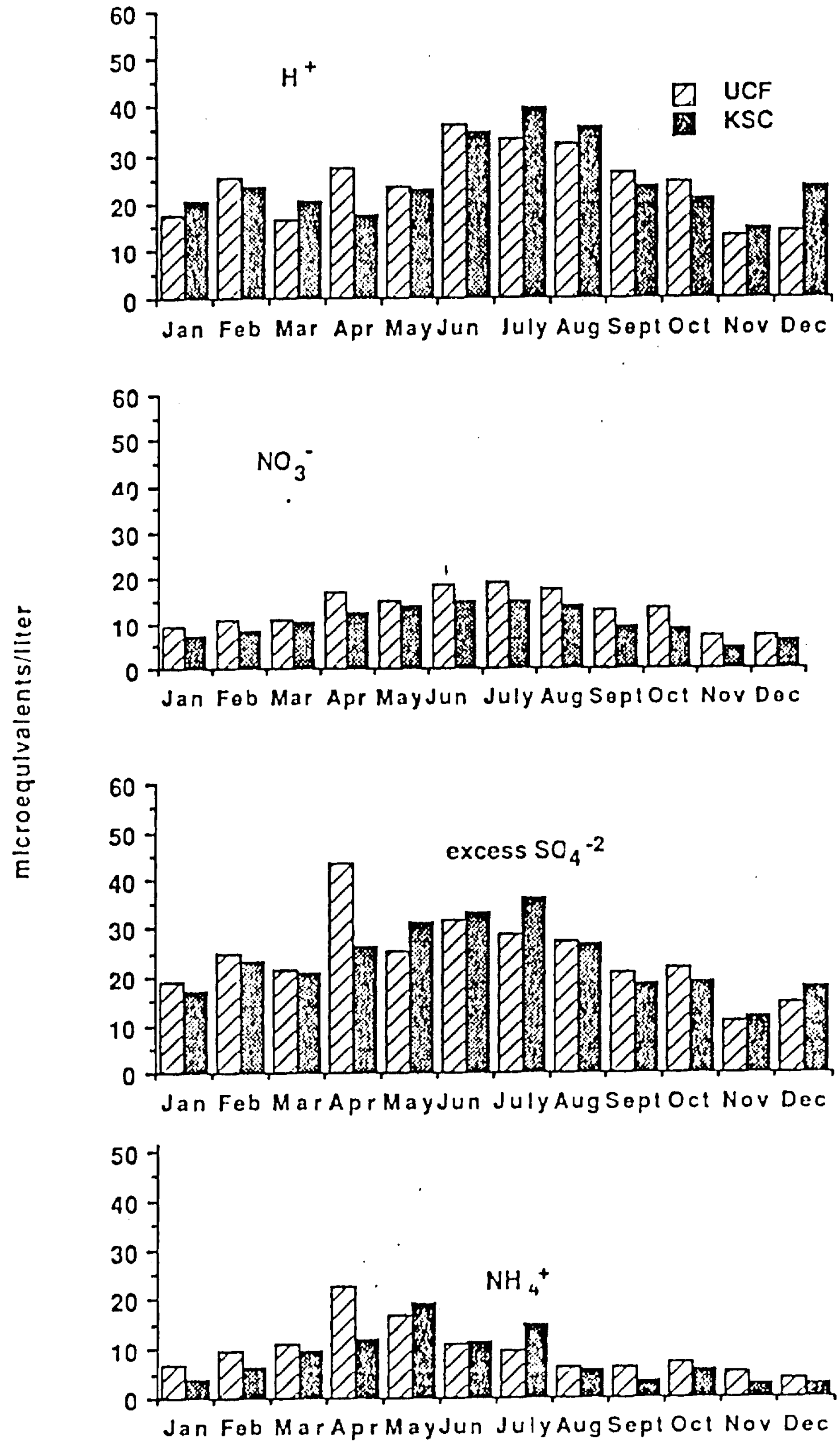


Figure 8. Monthly Volume Weighted Average Ion Concentrations at UCF and KSC.

ammonium ion concentrations as presented in Figure 6 have varied from 22 μN during 1981 to 2 μN during 1985 at KSC. During 1980 and 1981 when annual ammonium ion concentrations were highest, the annual excess sulfate concentrations were also observed to be the highest of the 12 year period.

Marine Influence on the Ionic Composition of Rain

The influence of site proximity to the ocean on ionic composition has been noted earlier in this report. Sea salts contribute the following ions to rainwater in order of decreasing importance: chloride, sodium, magnesium, calcium, sulfate and potassium. The Cl/Na ratio consistently approaches the ratio of 1.16 (eq./eq.) present in seawater which suggests that the concentrations of chloride and sodium measured in rain are a consequence of the marine influence. For this reason, chloride concentration and deposition can be used as a general measure of the marine influence. Monthly chloride concentrations exhibit a pronounced cool weather maximum and warm weather minimum. Prevailing winds and reinforcing weather patterns tend to occur from the north and east during the fall and winter, and therefore, can project the marine influence from the Florida east-coast inland. The monthly deposition of chloride is compared with acid deposition and total rainfall amounts in Figure 4. This comparison clearly shows the contrasting behavior that has been observed for the marine influence and acid related components of rainwater ion composition.

Trend Evaluation

An important objective of a long term monitoring program is to establish a data base that can be used to determine if a trend in observations can be established. Many approaches can be and have been used by others in attempts to answer this question using site specific or regional data from the NADP program. Individual sample, monthly or annual volume weighted average concentration or deposition data can be considered. Nonlinear regression has been used (Dana and Easter, 1987) to address both periodic tendencies and trend evaluation. Seasonal variability associated with monthly data from UCF and KSC have been observed as shown in Figures 5 and 8. Therefore, the nonlinear regression approach is based upon the following equation

$$-\log_{10}C = a + b*t + c*\sin(2\pi t + p)$$

where C is the concentration, t is time in years and a, b, c, and p are regression parameters. Monthly volume weighted concentration data from UCF for the 12 year period November, 1977 through October, 1989 have been evaluated. A significant periodicity was observed for the major ionic concentrations evaluated (e.g. acidity, nitrate, excess sulfate, chloride and ammonium ion); however, only pH and pNO_3 exhibited a significant time trend. The coefficients a, b, c and p were considered to be significant only if the value lies outside the range established by zero $\pm$2(s.e.) where s.e. represents the regression asymptotic standard error for each of the coefficients. The values for a, b, c and p in the above equation and associated standard errors are summarized in Table 3. The observed monthly UCF pH data and the results of the nonlinear regression evaluation are shown in Figure 9. Annual periodicity with pH minimum in July and the upward trend in pH as suggested by the slope of 0.018 pH units per year are apparent.

Table 3. Results of Nonlinear Regression Analysis Based Upon Monthly Weighted Average Concentrations.

SPECIES	a	b	c	p	month*
pH	4.549 (0.038)**	0.018 (0.005)	-0.167 (0.027)	3.344 (0.160)	July
pNH_4	5.064 (0.043)	-0.001*** (0.006)	-0.236 (0.030)	23.467 (0.127)	April
pNO_3	4.929 (0.033)	-0.012 (0.005)	0.155 (0.023)	6.867 (0.149)	June
pexcess SO_4	4.571 (0.037)	0.010*** (0.005)	0.155 (0.026)	13.613 (0.167)	May
pCl	4.673 (0.052)	-0.002*** (0.007)	0.200 (0.036)	-2.955 (0.182)	Dec-Jan

*Month where maximum concentration [minimum p (C)] is observed.
**Values in parenthesis are asymptotic standard errors.
***Nonsignificant coefficients.

SUMMARY

The results of 12 years of monitoring the chemical composition of rain in East-Central Florida have shown that the rain is moderately acid. Although the measured acidity is less than that observed in other regions of the U. S., it does suggest that the environmental impact of acid rain may eventually reach central Florida if it has not yet begun. The annual chemical composition of rain at UCF and at KSC has shown moderate variability. Extreme daily and monthly variations have been observed. However, these variations have not been addressed specifically in this report. Trends toward increased or decreased acidity or total ionic composition have been evaluated in preliminary fashion and it is suggested that pH has shown a very modest increase over the twelve year period. Total acid deposition has not shown a comparable decrease because rainfall amounts have increased in recent years compared to early years in the study period. The total ionic composition of rain collected at KSC is greater than that for rain collected at UCF; however, this can be accounted for by site proximity to the ocean with the accompanying marine influence.

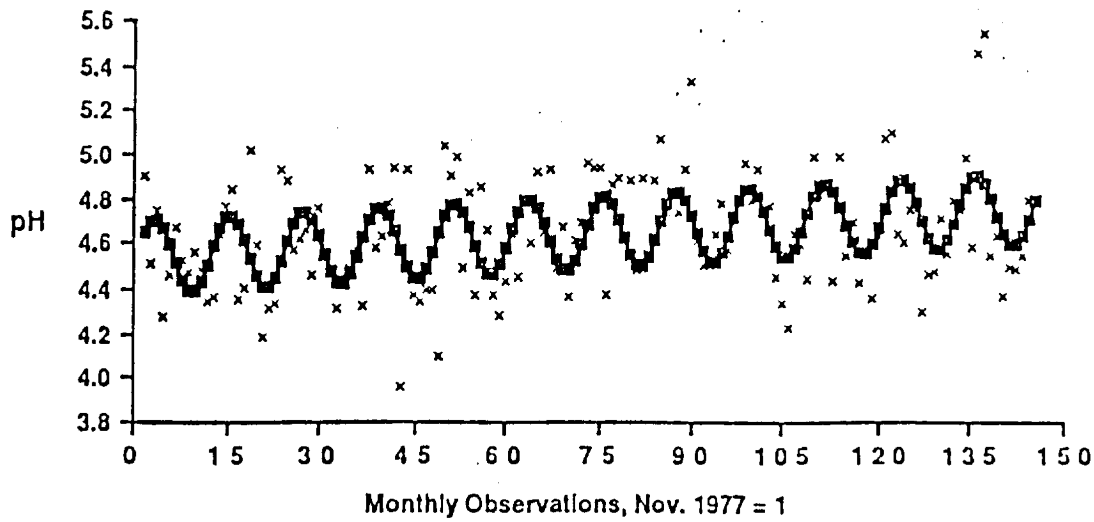


Figure 9. Observed Periodicity and 12 Year pH Trend at UCF.

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13. ABSTRACT *(Maximum 200 words)*

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