### ACID RAIN IN PUERTO RICO

by Elba D. Osborne Engineering Research Center University of Puerto Rico Mayaguez, Puerto Rico 00708

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### Introduction

The problem of acid precipitation has become a major environmental concern in the world. Many long range projects in the United States and Europe are devoted to the analysis of the problems associated with acid pollution deposition (2,5,10,13,18).

The importance of the contributions of ions in rainfall to the quality of surface water has been widely discussed (9,12). In their study of acid precipitation and drinking water supplies Reed and Henningson (15) selected three problem areas in the evaluation of the effect of acid precipitation, the increased levels of heavy metals in raw water, biological imbalances in water supplies, and increased corrosivity of delivered water.

Other works in the field of acid rain are concerned mainly with the development of theoretical models of wet deposition of contaminants (1,8). These works emphasize mostly the importance of factor and meteorological analyses.

So far, in the Caribbean area, reports by Jordan and Fisher, (7) for St. Thomas, Virgin Islands, Dalal, (4) for Trinidad and Quinones (14) in Puerto Rico point out to the presence of nitrate  $(NO_{\overline{3}})$ , sulfate  $(SO_{\overline{4}}^{2-})$ , chloride  $(Cl^{-})$ , magnesium (Mg), calcium (Ca), sodium (Na), and potassium (K), in the chemical composition of rain samples. Quiñones, also reports pH values ranging from 4.8 to 7.6 measured in rain samples corresponding to the four locations in his study.

More recently, research conducted by McDowell (12) revealed that the pH of bulk precipitation for one location in eastern Puerto Rico ranged from 4.1 to 6.5. These findings suggest that

acid rain may be a phenomenon of occurrence at some selected locations in Puerto Rico.

Considering that all surface waters in Puerto Rico are polluted by both organic and inorganic contaminants, and, since no systematic study of rain events throughout the different precipitation areas of the island had been undertaken, our study was aimed at determining the possible prevalence of acid rain on an islandwide basis.

To this purpose, we established a network of rain sampling stations which included fourteen precipitation areas in Puerto Rico. Rainfall samples were collected for about nine months which included the dry and most of the wet season. Chemical analyses were performed to determine pH, specific conductivity and ionic concentrations of nitrate (NO $\frac{3}{3}$ ), sulfate (SO $\frac{2}{1}$ ) and chloride (Cl $^{-}$ ) on the collected samples.

### Experimental

### Area Description

### Location

Puerto Rico is the most eastern and smallest of the Greater Antilles. It lies between 18° 31' N and 17° 55'N latitude and 65° 37'W and 67° 17'W longitude. It is about 100 miles from East to West and 40 miles North to South. The overall area is slightly over 3,400 square miles.

### Precipitation

Rainfall in Puerto Rico varies markedly from place to place. Measurements of rainfall are made at approximately 100 climatological stations.

The majority of all Puerto Rico's rainfall is orographic in nature. Moisture laden air from the ocean is carried by the trade winds inland and forced to ascend over the mountains where it gets cooled thus causing condensation in the form of rainfall. Easterly waves and cold fronts are the two rainfall producing mechanisms. Easterly waves prevail from May through November. During November to April the trailing edge of continental cold fronts may have definite effects upon the island rainfall.

### Wind

One of the most outstanding features of the wind in Puerto Rico is the steadiness of the Trade Winds. They blow between North-Northeast and South-Southeast. Being sorrounded by water,

the land and sea breeze effect is important in most coastal areas.

### Methodology

A network of fourteen sampling sites was established around the island following an analysis of geographical factors, variation in population density, industrial point sources of pollution and meteorological parameters. Precipitation records were maintained at each site.

Bulk rainwater samples were collected by means of a manual collector system consisting of a two liter polyethylene bottle equipped with a screw cap of the same material through which a plastic funnel and an overflow tube were attached. After collecting precipitation for seven days the bottles were immediately closed with a self sealing screw cap to prevent evaporation and or contamination.

At the laboratory, the samples were weighed and within 2-3 minutes after they were opened the pH measurement was made with a standard pH meter. The samples were then filtered through Whatman 40 paper and stored in stoppered polyethylene bottles until further analyses.

Nitrate ion concentration was determined by the brucine spectrophotometric assay (16), the sulfate ion concentration was measured by a turbidometric spectrophotometric method using BaCl as precipitating agent (17). An Orion chloride ion electrode, model 93-17, was used to determine the chloride ion concentration. Specific conductivity measurements of the samples were done with an YSI conductivity bridge.

### Results and Discussion

### A. Rainfall data

Three hundred seventy three bulk rain samples were collected at fourteen sampling locations around Puerto Rico, (Fig. 1) from October 1984 to June 1985. Each sample was assumed to be representative of the rainfall occurring during a seven day period. Eleven sampling stations reported 0.05 or less inches of rain during mid January and in these cases there was either no sample or the volume collected was not enough for analyses. This was the case at Lajas, in which no sample was collected in nine different seven days periods for lack of rainfall.

Table I presents the mean values obtained from the analyses of pH, specific conductivity, ion concentrations and rainfall data for each location. All three ions analyzed,  $NO_3$ ,  $SO_4$ , and  $Cl^-$  were present in the samples obtained at the precipitation areas under study. There was a wide fluctuation in the concentration of all the ions with maximums appearing during the day season, mainly January, and minimums at mid May after a big rainstorm.

The presence of chloride, the highest constituent of the three ions analyzed in the rainfall of Puerto Rico, with values ranging from 0.9 to 58 mg/l, can be attributed to the spread of oceanic salts by the action of the wind. We obtained an average combined data maximum concentration of chloride of 38 mg/l which compares with the value found by Jordan (7) in St. Thomas V.I. but, it is somewhat higher than the ones reported by Quiñones in Puerto Rico (14).

There is no data related to the contribution of chloride to the environment from man made emissions in Puerto Rico. In other areas studied, specially coastal areas, anthropogenic emissions of Cl are considered negligible compared to natural ones.

The nitrate ion was present in almost all of the rain samples analyzed with values in the range of 0.01 to 3.08 mg/l. The mean value of 0.20 mg/l obtained from the data shown on Table I is within the range of values reported by Jordan in St. Thomas (7) but only half of what was found by Quiñones (14) at four locations in Puerto Rico.

Nitrate in precipitation has it origin in the nitrogen oxides which are emitted into the atmosphere. The action of bacteria in the ground and lighthing are the main natural producers of nitrogen oxides, while the combustion of fossil fuels is the most important source of man made emissions. Nitrogen oxides produce nitrates and for each mole of nitrate in precipitation one mole of the hydrogen ion is available.

We also found that sulfate is an ionic component of precipitation at all the locations sampled (Table I). The values obtained ranged from 0.21 mg/l to 32 mg/l with maximums obtained usually after a dry period. The mean value for all stations of 2.36 mg/l shows a slight increase over those reported by Jordan (7) and Quiñones (14) for some locations in the Caribbean.

Sulfate in precipitation comes mainly from three sources: natural biological action, ocean spray and man made emissions. The sulfate from the ocean is considered neutral sulfate and will not contribute to the formation of hydrogen ions. Calculations

of rain samples' sulfate to chloride ratio show that in seven locations in Puerto Rico the ratio value exceeded 0.14 which is the sulfate to chloride ratio in sea spray (Table III).

This excess sulfate can be assumed to come from sulfur dioxide and hydrogen sulfide emissions to the atmosphere. For each mole of sulfate in rain two moles of hydrogen ions are produced, thus increasing the acidity of rain.

A very important step in the evaluation of the chemical composition of rain in the determination of its pH. Table (I) includes the mean values of this parameter obtained for the locations under study. Values ranged from a minimum of 4.2 in rain samples from Adjuntas to a maximum of 6.1 on those from San Sebastián.

### B. Data Analyses

Tables (IV-XVII) present Correlation Coefficients (r) among ions and parameters of rain samples collected at the sampling stations. There was great variation in the correlation coefficients obtained among the fourteen stations. At Gurabo, for example, significant correlations (P<0.05) were obtained in nineteen out of twenty one correlations considered, while in Ponce only two of twenty one were significant.

Table II shows the significant correlation coefficients among major ions obtained in their correlations to pH and conductivity for all locations.

In general, more frequent significant correlations were found in the pH vs  $SO_4^2$ -correlation than in the pH vs  $NO_3^-$ . Based

on these data analyses, we can say that in our study, in which three hundred seventy three rain samples were analyzed, the was a better indicator of rain acidity than the  $NO_3$ .

However, when the mean values of pH and  $SO_4^{2-}$  are examined, Table (I) a high concentration of  $SO_4^{2-}$  does not necessarily mean a low pH because of the many other factors that control the acidity of rain (3,6,11) and also because much of the  $SO_4^{2-}$  present might come from ocean spray and would not contribute to the production of hydrogen ions.

The Cl $^-$  was much higher and more frequently correlated to the specific conductivity than  $SO_4^-$  and  $NO_3^-$  as shown on Tables (IV-XVII). This was expected in view of the higher concentration values obtained for Cl $^-$ .

### C. Meteorological aspects

The main precursors of acid rain, sulfate and nitrate, may stay up in the atmosphere after they are emitted until they settle down as dry deposition or are washed down by precipitation. Their movement depends on the wind direction and speed.

Fig. 1 presents the location of the sampling stations and the prevailing direction of the wind in relation to the geography of Puerto Rico. Surface winds, which would be the ones responsible for the diffusion of pollutants, are characterized in the Northern part of the island by their NE and ENE components. In the South, the wind of the SE quadrant tends to be stronger than the other quadrants.

Fig. 2 shows the pH contour lines obtained by data extrapolation of the pH values from each location. The pH increases to a 5.4 maximum followed by a decrease to a 4.3 minimum in the Northern part of the island and in an East-West direction. In the same direction there is another maximum of 6.1 decreasing to 4.4 in the West coast.

In the Southern portion there is a 5.5 maximum starting from the coast and decreasing to a 4.2 in the mountains.

A possible explanation to this pH behavior is in terms of the wind patterns and the probable sources of emissions. The 5.4 maximum lies within the Luquillo rain forest, while the 4.3 minimum in Corozal is Southwest of the densely populated metropolitan area of San Juan. The next maximum pH 6.1 is in the mainly agricultural area of San Sebastian.

In the Southern portion of the island we encounter the same situation with respect to the wind direction, the minimum pH of 4.2 lying Northwest of the populated and industrial area of Ponce.

### Conclusions

Acid rain, by definition, is rain having a pH value of 5.6 or below. But, also, in the absence of basic materials such as NH and  $CaCO_3$ , average pH values of "pure" rain can be expected to be in the vecinity of 5 (3).

Based upon the findings of this study we can conclude that acid rain, although still not a very serious problem, is a phenomenom present in the environment of the island of Puerto Rico.

This is more noticeable at the areas of Corozal and Adjuntas. The dry season, running from January to April, manifested a higher acidity in rain than the periods of October to December and the month of May sampled in our work. The  $SO_4^{2-}$  was found to be a better indicator of acidity than  $NO_3^-$ .

Our study indicates that prevailing wind patterns play a very important role in the geographical distribution of pollutants that cause the acidity of rain in Puerto Rico.

Considering the damage that acid rain causes to the environment, particularly aquatic ecosystems, we suggest that a monitor system should be implemented in those geographical areas where this phenomenom seems to be developing into a serious problem.

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Table I. Mean values from analyses of rainfall samples in Puerto Rico.

	рН	Cond umohs	$\frac{NO_{\overline{3}}}{mg/L}$	SO∏ mg/L	Cl - mg/L	Rainfall inches
Adjuntas	4.2	19.32	0.24	2.07	9.36	2.00
Arecibo	4.8	36.64	0.23	4.40	18.16	0.89
Bayamón	4.9	40.79	0.18	1.93	15.31	1.24
Corozal	4.3	27.93	0.23	1.67	14.36	1.55
Fajardo	5.3	30.88	0.33	2.45	17.51	1.22
Gurabo	5.4	31.27	0.29	1.90	17.01	0.79
Isabela	5.0	32.67	0.29	2.36	16.17	0.91
Lajas	4.9	31.74	0.22	2.94	12.83	0.80
Mayaguez	4.4	42.68	0.12	3.19	8.96	5.52
Ponce	5.5	28.31	0.04	2.16	11.32	0.87
Rio Piedras	4.9	30.06	0.23	1.64	13.58	1.15
San Sebasti <b>á</b> n	6.1	58.66	0.11	1.44	17.60	1.75
Utuado	4.5	32.46	0.18	2.53	19.23	1.32
Yabucoa	5.3	28.53	80.0	2.40	15.76	1.70

Table II

Correlation coefficients (r) among major ions and parameters of rain samples collected at fourteen sites in Puerto Rico (P<0.05)

	pH vs so2- 4	pH vs NO - 3	Cond vs so2- 4	Cond vs NO <sub>3</sub> -	so vs NO <sub>3</sub> -
Adjuntas	<b></b>				
Arecibo	0.80	0.67	0.44		0.81
Bayamón			0.85	0.66	0.51
Corozal	*** -**			0.40	0.37
Fajardo	0.53	0.44			
Gurabo	0.43	0.48	0.49	0.68	0.62
Isabela			0.68		0.54
Lajas	0.42				
Mayaguez					
Ponce					
R <b>i</b> o Piedras				<del></del>	
San Sebasti <b>á</b> n	0.49	<del></del>	0.45		
Utuado	0.55		0.70		
Yabucoa	0.44		0.73		

Table III

Sulfate to Chloride Ratio Obtained from Mean Values of Rainfall Samples in Puerto Rico

Location	so4/c1
Adjuntas	0.22
Arecibo	0.24
Bayamon	0.13
Corozal	0.12
Fajardo	0.14
Gurabo	0.11
Isabela	0.15
Lajas	0.23
Mayaguez	0.36
Ponce	0.19
Rio Piedras	0.12
San Sebastian	0.08
Utuado	0.13
Yabucoa	0.15

Table IV. Matrix of correlation coefficients (r) among ions and parameters of rain samples

## ADJUNTAS

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нd	0.0678 ( 25 ) P = .374					
Conductivity	-0.5889 ( 24 ) P = .001	-0.3579 (24) P = .043				
NO 3	-0.2317 ( 24 ) P = .138	0.2422 (24) P = .127	0.1137 (23) P = .303			
SO 7	-0.3851 (23) P = .035	0.0998 (23 ) P = .325	0.2611 (23) P = .114	0.2017 ( 22 ) $P = .184$		
c1 -	-0.1398 (17) P = .298	-0.5058 (17) P = 019	0.3933 (17) P = .059	-0.1677 ( 16 ) P = .267	-0.0608 (17) P = .408	
Rainfall	0.3495 (29) P = .032	0.2271 (25) P=.137	-0.4943 (24) P = .007	-0.2081 ( 24 ) P = .165	-0.2639 ( 23 ) P = .112	-0.2467 (17) P = .170
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The number in paretheses represent the number of samples included in the computation.

Table V. Matrix of correlation coefficients (r) among ions and parameters of rain samples

## ARECIBO

************	Sample Volume Volume	- 41	PH Conductivity NO 3 SO 4 C1-	NO 3	SO4	C1-
ЬН	-0.4852 ( 26 ) P = .006					
Conductivity	-0.2667 (24) P = .104	0.1851 ( 24 ) P = .193				
NO3	-0.2272 ( 26 ) P = .132	0.6655 (26 ) P = 0.000	0.0970 (24) P = .326			
so <u>₹</u>	-0.3443 (24) P = 0.050	0.8032 (24) P = 0.000	0.4388 (22) P = 0.021	0.8123 ( 24 ) P = 0.000	·	
c1 <sup>-</sup>	-0.2850 ( 20 ) $P = .112$	0.0881 (20 ) P = .356	0.9434 (20) P = 0.000	0.1775 ( 20 ) P = .227	0.4499 ( 18 ) P = .030	
Rainfall	0.8029 ( 26 ) P = 0.000	-0.4551 ( 26 ) P = .010	-0.3306 ( 24 ) P = .057	-0.1595 ( 26 ) P = .218	-0.1636 (24) P = 222	-0.3767 ( 20 )

The number in parentheses represent the number of samples included in the computation.

Table VI. Matrix of correlation coefficients (r) among ions and parameters of rain samples

## BAYAMON

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	Sample	нd	Conductivity	NO 3	So₫	C1_
*************************************	*****		****	******	********	<b>计算的数据数据数据数据数据数据数据数据数据数据数据数据数据数据数据数据数据数据数据</b>
рн	-0.2727 (20) P = .122					
Conductivity	-0.3591 (20 ) P = .060	-0.1849 ( 20 ) P = .218				
NO 3	-0.2852 (19) P = .118	-0.2218 (19) P = .181	0.6556 ( 19 ) P = .001			
SO 4	-0.5384 (10) P = .011	-0.0379 (18) P = .441	0.8534 (18) P = 0.000	0.5064 (18) P = .016		•
c1 ~	-0.1893 (18) P = .226	0.1442 (18) P = .284	0.8090 (18) P=0.000	0.5971 ( 18 ) P = .004	0.1710 ( 18 ) P = .249	
Rainfall	0.5400 (25) P = .003	-0.0058 (20) P = .490	-0.2749 (20) P = .120	-0.1076 ( 20 ) P = .326	-0.2684 (19) P = .133	-0.2706 (19) P = .131
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The number in parentheses represent the number of samples included in the computation.

Table VII. Matrix of correlation coefficients (r) among ions and parameters of rain samples

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Sample Volume	Sample Volume	PH	Conductivity ************************************	NO 3	SO 4	pH Conductivity NO3 SO4 C1 C1 C1
hď	-0.1196 ( 26 ) P = .280					
Conductivity	-0.3596 ( 25 ) P = .039	-0.0859 (25) P = .341				
NO3	-0.0635 ( 26 ) P = .379	-0.2871 (26) P = .078	0.3978 (25) P = .024			
SO4	-0.3200 (25) P = .059	0.0252 (25) P = .452	0.3359 ( 25 ) P = .050	0.3682 (25) P = .035		
C1~	-0.3243 (23) P = .066	-0.4476 (23) P = .016	0.9155 (22) P = 0.000	0.5769 (23) P = .002	0.3844 ( 22 ) $P = .039$	
Rainfall	0.7174 (29) P=0.000	0.0322 (26) P = .438	-0.5009 ( 25 ) P = .005	-0.2981 ( 26 ) $P = .070$	-0.2693 (25) P = .097	-0.4648 (23) P = .013
<b>机状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状状</b>	*****************	***************	********	**********	***	***********

The number in parentheses represent the number of samples included in the computation.

Table VIII. Matrix of correlation coefficients (r) among ions and parameters of rain samples

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	Sample Volume	##************************************	pH Conductivity NO $_{\overline{3}}$ SO $_{\overline{4}}$ Cl-	NO 3	SO4	C1-
Нq	-0.3562 (23) P = .048					
Conductivity	-0.3852 (20) P = .047	0.1256 ( 20 ) P = .299				
NO 3	-0.4576 (23) P = .014	0.4366 ( 23 ) P = .019	-0.0868 ( 20 ) P = .358			
SO = 1	-0.4745 (21) P = .015	0.5288 ( 21 ) P = .007	0.1994 ( 20 ) P = .200	-0.0085 ( 21 ) P = .485		
c1-	-0.6462 { 19 } P = .001	0.3138 ( 19 ) P = .095	0.8104 (19) P = 0.000	0.4813 ( 19 ) P = .018	0.3672 ( 19 ) P = .061	
Rainfall	0.4629 (23) P = .013	-0.2009 (23) P = .179	-0.4602 (20) P = .021	-0.2111 ( 23 ) P = .167	-0.2157 (21) P = .174	-0.5406 (19) P = .008

The number in parentheses represent the number of samples included in the computation.

Table IX. Matrix of correlation coefficients (r) among ions and parameters of rain samples

**************************************	**************************************	HQ	GURABO ************************************	*****	**************************************	GURABO ************************************
**************************************	-0.6147 (24) P = .001	7. 教授 法转移 ** ** ** ** ** ** ** ** ** ** ** ** **	·····································	· · · · · · · · · · · · · · · · · · ·	***	化物项化物化物化物物物物物物物物物物物物物物物物物物物物物物物物物物物物物物
Conductivity	-0.6376 (21) P = .001	0.4400 (21) P = .023				
NO3	-0.4920 ( 24 ) P = .007	0.4834 (24) P = .008	0.6822 (21) P = 0.000			
SO.4	-0.4028 (22 ) P = .032	0.4288 (22) P = .023	0.4926 (21) P = .012	0.6186 (22) P = .001		,
c1 <sup>-</sup>	-0.4421 ( 18 ) $P = -0.33$	0.0666 (18) P=.396	0.8703 (18) P = 0.000	0.7860 ( 18 ) P = 0.000	0.5812 ( 18 ) P = .006	
Rainfall	0.9232 (27) P = 0.000	-0.5255 ( 24 ) P = .004	-0.6199 (21) P = .001	-0.3490 ( 24 ) $P = .047$	-0.3610 (22) P = .049	-0.3701 ( 18 ) P = .065

The number in parentheses represent the number of samples included in the computation.

Table X. Matrix of correlation coefficients (r) among ions and parameters of rain samples

ISABELA

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Нď	-0.5979 ( 26 ) P = .001					
Conductivity	-0.4224 ( 24 ) P = .020	0.1157 (24) P = .295				
NO 3	-0.4465 ( 26 ) P = .011	0.1674 ( 26 ) P = .207	0.1432 (24 ) P = .252			
SO 24	-0.5292 (23) P = .005	0.2176 (23) P = .159	0.6780 (23) P = 0.000	0.5350 ( 24 ) P = .004		
	-0.4308 (22) P = .023	-0.0229 (22) P = .460	0.9116 (22) P = 0.000	0.3134 (23 ) P = .073	0.5558 ( 22 ) P = .004	
Rainfall	0.8072 (27 ) P = 0.000	-0.5445 ( 26 ) P = .002	-0.3855 (24) P = .031	-0.4561 ( 27 ) P = .008	-0.3613 ( 24 ) P = .041	-0.3884 (23) P = .034
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The number in parentheses represent the number of samples included in the computation.

(r) among	
coefficients	rain samples
Matrix of correlation	ions and parameters of rain
Table XI.	

LAJAS

*******************************	***************	*****	*************	***********	**********	<b>的现在分词 阿拉克斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯斯</b>
	Sample Volume		Conductivity	NO 3	SO⊈	c1_
***************************************	*******		****	****	****	***************************************
Нd	-0.5143 (22) P = .007					
Conductivity	0.1674 ( 16 ) P = .268	0.1231 (16) P = .325				
NO3	-0.1011 (22) P = .327	-0.3425 (22) P = .059	-0.1605 (16) P = .276			
SO 2	-0.5419 (21) P = .006	0.4201 (21) P = .029	0.1930 (15) P = .245	0.3490 ( 21 ) P = .060		
c1 ~	-0.5727 (12) P = .026	0.0591 (12) P = .428	-0.2549 (12) P = .212	0.3363 (12) P = .143	0.4341 (12) P = .079	
Rainfall	0.8955 (29) P = 0.000	-0.2905 (22) P = .095	0.2130 ( 16 ) P = .214	-0.2215 ( 22 ) P = .161	-0.4597 ( 21 ) P = .018	-0.6775 (12) P = .014
在古代生物的人名英格兰人姓氏克拉特的变体 医克克特氏征 医克克特氏氏征 医克克特氏氏征 医克克特氏氏征 化二氯甲基苯甲基甲基苯甲基甲基苯甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基	****************	*****	<b>水水水水水水水水水水水水水水水水水水</b>	*****	*********	我有处于我们的有效的 计多数分类 医克拉氏性 化异性 的复数 化二氯甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基

The number in parentheses represent the number of samples included in the computation.

Table XII. Matrix of correlation coefficients (r) among ions and parameters of rain samples

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Sample Volume	Sample Volume	HQ ####################################	Conductivity ********	E ON *****	######################################	pH Conductivity NO3 SO4 C1 + + + + + + + + + + + + + + + + + +
рн	-0.2591 ( 22 ) P = .122					
Conductivity	-0.2509 (17) P = .166	0.0973 (17) P = .355				
NO3	0.0149 (18) P = .477	-0.1643 (19) P = .251	-0.1710 (15) P = .271			
\$08 \$	-0.6616 { 18 } P = .001	-0.3021 (19) P = .104	0.1241 ( 16 ) P = .324	0.0448 (17) P = .432		
c1-	-0.3731 (15) $P = .085$	-0.1751 (15) P = .266	-0.1551 ( 14 ) P = .298	0.5985 (13) P = .015	0.4865 ( 14 ) P = .039	
Rainfall	-0.1077 (25) P = .304	-0.1684 (23) P = .221	-0.4613 (17) P = .031	-0.2405 (19) P = .161	0.2585 ( 19 ) P = .143	-0.2222 (15) P = .213
***************************************	**************	*****************	************	*****	**********	经存储分割的 医克拉克氏征 医克拉克氏性 医克拉克氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 医克拉氏征 计记录器 计记录器 计记录器 计记录器 计记录器 计记录器 计记录器 计记录器

The number in parentheses represent the number of samples included in the computation.

Table XIII. Matrix of correlation coefficients (r) among ions and parameters of rain samples

***************************************	***************************************		PONCE	*****	***************************************	PONCE
************	Sample Volume		Conductivity	NO3	SO 4	pH Conductivity NO3 SO 4 Cl-
	-0.2561 (21) P = .131					
Conductivity	-0.0897 (18) P = .362	0.6370 { 18 } P = .002				
NO3	-0.2873 ( 20 ) P = .110	0.0405 ( 20 ) P = .433	-0.0191 (18) P = .470			
50≩	-0.2374 (14) P = .207	-0.0133 ( 14 ) P = .482	0.0462 (13) P = .440	-0.1729 ( 14 ) P = .277		
c1-	-0.4555 (12) P = .068	0.4669 (12) P = .063	0.2311 (12) P = .235	0.0405 (12) P = .450	-0.1377 ( 9 ) P = .362	
Rainfall	0.6507 ( 29 ) $P = 0.000$	-0.1308 (21) P = .286	0.0015 (18) P = .498	-0.1021 ( 20 ) P = .334	-0.3420 ( 14 ) P = .116	-0.4894 (12) P = .053
****	***************************************	***********	*****	******	****	*************************************

The number in parentheses represent the number of samples included in the computation.

Table XIV. Matrix of correlation coefficients (r) among ions and parameters of rain samples

## RIO PIEDRAS

Sample Volume	Sample Volume	HCI	pH Conductivity NO3 SO4 Cl-************************************	NO3	SO4	C1-
рн	0.0164 (27) P = .468					
Conductivity	-0.2018 (27) P = .156	-0.0171 (27) P = .466				
NO 31	-0.4801 (24) P = .009	0.0673 (24) P = .377	0.1005 ( 24 ) P = .320			
SO SIA	-0.5906 (22) P = .002	-0.2620 (.22.) P = .119	0.3146 (22 ) P = .077	0.1229 ( 19 ) P = .308		
cı_	-0.2521 (24) P = .117	0.0894 (24) P = .339	0.6250 (24 ) P = .001	0.4191 ( 41 ) P = .029	0.5609 ( 19 ) P = .006	
Rainfall	0.3887 (31) P = .015	-0.0091 (27) P = .482	-0.4317 ( 27 ) P = .012	0.1831 ( 24 ) P = .196	-0.0464 ( 22 ) P = .419	-0.2704 ( 24 ) P = .101
*************************************	******		据水板的预算机场或有效的现在分词 的现在分词 有关的 医多种性 医克勒特氏试验检尿道 医多种性 医多种性 医多种性 医多种性 医多种性 医多种性 医多种性 医多种性	****	*****	*****

The number in parentheses represent the number of samples included in the computation.

Table XV. Matrix of correlation coefficients (r) among ions and parameters of rain samples

# SAN SEBASTIAN

Sample	Sample Volume	HO	Conductivity	*************	**************************************	nnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnnn
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Нď	-0.4736 ( 19 ) P = .020					
Conductivity	-0.3849 (19) P = .052	0.6251 (19) P = .002				
NO3	-0.4716 (17) P = .028	0.3233 (17) P = .103	0.2699 ( 17 ) P = .147			
so <u>₹</u>	-0.3137 (19) P = .095	0.4949 (19) P = .016	0.4451 ( 19 ) P = .028	0.1613 (17) P = .268		
Cl-	-0.6110 (18) $P = .004$	0.4683 (18) P = .025	0.3076 ( 18 ) P = .107	0.6524 ( $16$ ) P = $.003$	0.3268 ( 18 ) P = .093	
Rainfall	0.8146 (25) $P = 0.000$	-0.4824 (19) P = .018	-0.4331 (19) P = .032	-0.3046 (17) P = .117	-0.3327 (19) P = .082	-0.6331 (18) P = .002
法有法律的法律的法律的法律的法律的法律的法律的法律的法律法律的法律法律的法律法	*************	***************	*********	******	**********	<b>经经验证据证据证据证据证据证据证据证据证证证证证证证证证证证证证证证证证证证证</b>

The number in parentheses represent the number of samples included in the computation.

Table XVI, Matrix of correlation coefficients (r) among ions and parameters of rain samples

## YABUCOA

************************************	***************	***************************************	***************	***********	*********	化分子处理器 化异丙基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲基甲
Sample Volume Volume	Sample Volume	Hd	Conductivity	NO3	§os	pH Conductivity NO $\bar{3}$ SO $\bar{4}$ Cl $^-$
ЬН	-0.2151 ( 20 ) P = .181					
Conductivity	-0.5480 (19) P = .008	0.5427 (19) P = .008				
NO 3	-0.1144 (20) P = .315	0.0074 (20 ) P = .488	-0.0257 ( 19 ) P = .458			
SO ₹	-0.1984 (20) P = .201	0.4393 (20) P = .026	0.7299 (19) P = 0.000	-0.1610 ( 20 ) P = .249		
C1 <sup>-</sup>	-0.4896 ( 16 ) $P = .027$	0.6023 (16) P = .007	0.8922 (16) P = 0.000	0.0354 (16) P = .448	0.4828 ( 16 ) P = .029	
Rainfall	0.3631 (20) P = .058	-0.3345 (20) P = .075	-0.4308 (19) P = .033	-0.1949 ( 20 ) P = .205	-0.2114 ( 20 ) P = .185	0.3568 (16) P = .087
***************************************	*****		*************	*****	*****	<b>物学学家教育技术的介绍中央教育技术的教育教育的教育的教育教育教育教育教育教育教育教育教育教育教育教育教育教育教育</b>

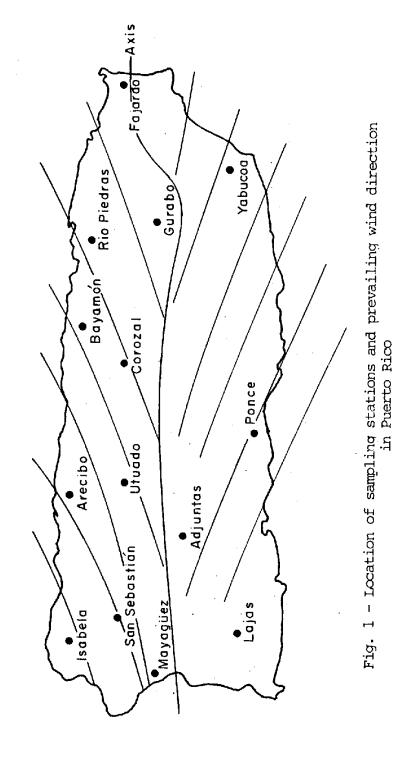
The number in parentheses represent the number of samples included in the computation,

Table XVII. Matrix of correlation coefficients (r) among ions and parameters of rain samples

UTUADO

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Sample Volume	Sample		Conductivity	NO 3	80₫	c1 <sup>-</sup>
***************************************	***************************************			***************************************	***************************************	经存款分词 化苯丙二苯甲甲苯甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲甲
рн	0.2078 (27) P = .149					
Conductivity	0.2513 (25) P = .113	0.7431 ( 25 ) P = $0.000$				
NO3	-0.1872 (25) P = .185	0.0119 (25) P = .478	-0.0814 (23) P = .356			
SO 4	-0.0546 (25) P.398	0.5479 ( 25 ) P = .002	0.6983 ( 24 ) P = 0.000	-0.0260 (23) P = .453		
	0.1340 (23) P = .271	0.5803 (23) P = .002	0.7104 (23) P = 0.000	0.5116 ( 21 ) P = .009	0.5674 ( 23 ) P = .002	
Rainfall	0.5967 ( 28 ) P = 0.000	-0.0352 (27) P = .431	-0.0792 (25) P = .353	-0.2201 (25) P = .145	-0.1781 ( 25 ) $P = .197$	-0.0968 (23) P = .330
化化物 化苯基苯酚 医克克克氏试验检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检	*****		**********	***	****	的现在分词 医克拉克氏试验检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检检

The number in parentheses represent the number of samples included in the computation.



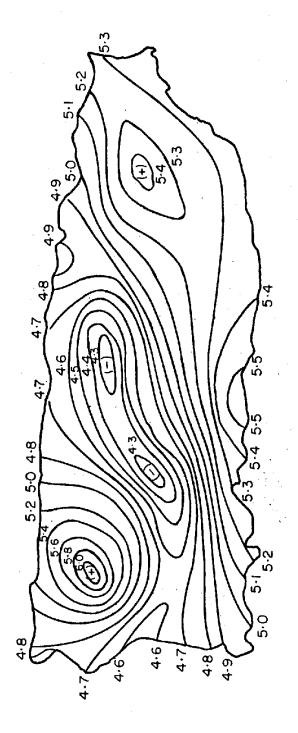


Fig. 2 - Rainfall pH in Puerto Rico