

# **Rainwater Chemistry Across the United States**

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## INTRODUCTION

Our study aims to understand the chemical composition of rainwater collected from 48 sites in 31 states throughout the United States, including Alaska and Hawaii (*see Fig. 1*). Using an ion chromatograph, we analyzed our samples for seven anions: chloride, nitrate, nitrite, sulfate, phosphate, fluoride, and bromide. We also sent some of our samples to the University of Utah to be analyzed by Scott Hynes for  $\delta^{18}O$ . Our hypothesis is that rainfall on the coast will contain higher levels of  $\delta^{18}O$  than rainfall further inland. This is based on the assumption that  $\delta^{18}O$  isotopes, which have a high molecular weight, are generally fractionated, and therefore are prevented from moving further inland by mountains. In order to better understand current knowledge and research regarding rainwater composition, we have reviewed several journal articles.

## OVERVIEW OF RESOURCES

*Diurnal Variations in major rainwater components at a coastal site in North Carolina* was conducted by G. Brooks Avery Jr., Joan D. Willey, and Robert J. Keiber on the campus of the University of North Carolina at Wilmington. The purpose of this study was to investigate diurnal variations in concentration of rainwater components having primarily aerosol or gas phase sources (Avery, Willey, and Keiber, 2001).

Anthropogenic, caused by humans, emissions such as nitrate or acetic acid were most pronounced in the afternoon and during the night. These rainwater components were removed by two possible processes; formation of dew or the advection of sea air that is relatively uncontaminated by anthropogenic emissions. This study highlights the importance of sinks (any process that removes components from rainwater) when

considering the variations in diurnal concentrations of the atmospheric components of rainwater. We can study the diurnal variation of the components of rainwater of our Carleton samples but we have insufficient data to accurately study these variations in the rest of our samples.

*A Long Term Study on Chemical Composition of Rainwater at Dayalbagh, a Suburban Site of Semiarid Region* was conducted by Ranjit Kumar, Abha Rani, S.P. Singh, K. Maharj Umari and S.S. Srivastava. This study's purpose was to see in the long-run the severity of the consequences expanding industry had on the pH of rainwater and to discover what ions were the most dominant players in determining the pH of local rainwater. The Indian government commissioned this study to determine the severity of the threat posed to the Taj Mahal by the air pollution caused by expanding industries. A mean pH of  $7.01 \pm 1.03$  was observed during their test period 1988, 1991-1996 which is well above the observed pH of 5.6 for rainwater. A strong presence of such pH neutralizing cations such as  $\text{Ca}^{+2}$ ,  $\text{Mg}^{+2}$ , and  $\text{NH}^{+4}$  was the cause for such high pH values. This study gives us a base on which we can make possible hypothesis to why the pH of some samples may be well above 5.6. This information will be most useful when applied to our samples taken from semiarid regions such as the Nevada or Arizona.

*Spatial distribution of  $\delta^{18}\text{O}$  in meteoric precipitation* was published in 2002. Data was taken from 583 stations established by The International Atomic Energy Agency-World Meteorological Organization Global Network for Isotopes in Precipitation. Their results can be applied for a number of purposes: to study the effect of local topography of the oxygen 18 isotope, to determine the relationship between the oxygen 18 isotope and latitude, altitude, vapor sources, storm tracks and other geographic variables.

*Chemical Composition of Rainwater around an industrial region in Mumbai* was conducted from 1994-1995 in Kaylan, Alibag and Cloaba India investigating the effects of a large industrial belt. Rainwater was studied in order to draw conclusion on how the industry affects the surrounding atmosphere. To determine the acidity of the rainwater, tests were conducted measuring the levels of sulphur dioxide and nitrogen dioxide in the water. They found that the pH of the rain is generally higher downwind of the industrial sites, and that the concentration of acid rain decreased when natural gas was burned instead of coal. It is possible that discrepancies in our data could be the result of pollutants in the air.

The study *Chemical composition of rainwater anthropogenic influence in the Piracicaba River Basin, Southeast Brazil* performed by Lara P. Artaxo, L.A. Martinelli, R.L. Victoria, P.B. Camargo, A. Krusche, G.P. Ayers, E.S.B. Ferraz, M.V, Ballester is a valuable resource because it investigated anthropogenic effects on rain water chemistry. From August 1997 to July 1998, 272 water samples were collected and tested for numerous minerals, pH and conductivity. The research indicated 3 main sources of human influence on the rain water for the area: sugarcane burning, soil dust and industrial emission, and the effect they had on the environment. While our methods of collection are less in-depth and more comprehensive, the results reveal potential explanation of the results of our project.

Chemical analysis of rainwater form 1999 to 2000 as stated in *Rainwater composition in northeast Uruguay* by M. Zunckel, C. Saizar and J. Zarauz discovered 4 contributing factors to rain composition in the area to be terrigenous (rock, soil, dust), agricultural and the result of marine air influx and biomass burning. The collected

samples were tested for a number of elements and qualities and then statically accessed. The results of the research reveal that the 7.0-4.6 pH level is caused by the terrigenous and agricultural sources, while a discovered correlation between chloride, sodium and magnesium; nitrate and ammonium; and fluoride and organic acids, suggests contributions from an influx of marine air from the Atlantic Ocean, livestock and fertilizers from near by farms and biomass burning from fuel wood, agro-industries and crop fertilizer. Though the effects are specific to the area, they might help to illuminate cases behind our results.

*Characterization of Groundwater from Rice County, Minnesota, using CFC dating, Hydrogen and Oxygen Isotope Analysis, and Ion Concentrations* was conducted by Carleton College Geology students in Rice County, Minnesota in January and February 2004. The students were trying to determine the age of and chemical composition of groundwater from the Rice County Area. They analyzed their samples for hydrogen and oxygen isotopes as well as anion and cation concentrations, and they attempted to determine the recharge dates of the samples using chlorofluorocarbon dating. They found high levels of the anion nitrate in several samples, which indicates anthropogenic contamination from fertilizer or improperly working septic systems. They also found amounts of Fluoride, Chloride, Sulfate, Sodium, Potassium, Magnesium, and Calcium in all of their samples. Nitrate and Nitrate-N were present in 11 of the 16 samples. They found the levels of the oxygen-18 isotope ranging from -8.67 to -9.80, which implies that the groundwater in Rice County is regionally or locally recharged. This data will be useful when compared to the data that we received from our oxygen-18 isotope testing of rainwater samples from Northfield and around the country. Since the

groundwater of Rice County is at least partially recharged from rainwater, the levels of anions and cations present in this study can also be compared to the test results from rainwater from Northfield. However, the chemical composition of the water can be altered significantly once it reaches the water table.

*Study of chemical elements in atmospheric precipitation in South Brazil* was conducted in Southern Brazil in the Candiota region in 2001. The goal was to investigate ion concentrations and pH levels in rainwater in order to determine the environmental effects of nearby coal and limestone mining industries. The study concluded that rainwater in the area examined was slightly acidic, probably as a result of emissions from nearby coal-powered power plants. They also attributed specific ions to various anthropogenic sources, including agricultural factors, and organic sources such as the soil and sea water. This study will help us determine possible sources for high concentrations of specific ions in our rainwater samples. However, because our sampling area is so broad, it will be difficult to determine the sources of anthropogenic pollutants.

## **CLAIM**

No study of this magnitude has been done at Carleton College. We have gathered 51 samples of rainwater from 31 states, including Alaska and Hawaii. It is also possible that this is one of the most comprehensive and wide-spread studies of rainwater across the entire United States. Our extensive results will allow us to draw firm conclusions about the nature of rainwater in the U.S..

## **AGENDA**

Initially we will explain how our samples were taken and where they originated from. Explanations of the tests conducted on the samples (Ion Chromatography and Oxygen Isotopes) will follow. Using tables and charts we will explain the significance of our results, and draw conclusions on the nature of rainwater.

## **METHODS**

We sent vials to relatives of our classmates and to Carleton College Geology Alumni, asking them to collect rainwater without contaminating the samples. The packages sent included a 150 milliliter collection vial and a letter. In the letter we asked participants to promptly return the samples, along with data about when they collected their samples, how much it rained when they collected their samples, and any other useful information. We also collected samples from Carleton's campus on four separate dates, in order to analyze the composition of rainwater in one place over time. We collected rainwater in large clean buckets that were prerinsed with deionized water and then transferred it into sterile vials. The vials were stored in a refrigerator in order to prevent evaporation.

We sent 26 of our samples to Scott Hynek at the University of Utah for  $\delta^{18}\text{O}$  testing. In our own lab we analyzed all 51 samples for anions using the Ion Chromatograph. All samples were filtered through a 0.25  $\mu\text{m}$  filter before being injected into the column. The filters were pre-rinsed with at least 30 ml of deionized water before filtering the sample to remove soluble salts on the filter. Our water samples were injected into a stream of carbonate-bicarbonate eluant and passed through a series of ion

exchangers. Based on their relative affinity for the capacity of a strongly basic ion exchanger, the anions are separated. The anions with a high affinity move slowly through the column, whereas the anions with a low affinity move much more quickly, thus the anions are separated based on their different retention times within the column. Once the anions are separated, they are passed through a strongly acidic cation exchanger in the form of a micromembrane suppressor, which is continuously bathed in a strong acid solution (regenerate). In the suppressor the separated anions are converted to their highly conductive acid forms and the carbonate-bicarbonate eluant is converted to weakly conductive carbonic acid. A specific conductance detector then measures the separated ions in their acidic forms. Anions species are determined by retention time and concentration is determined by peak area compared to known standards.

## **RESULTS**

### Anion Concentrations

We analyzed our samples for seven anions using the Ion Chromatograph (*For a sample readout from the Ion Chromatograph, see Figure 3 in Appendix*). Of the seven anions we analyzed for, fluoride and bromide were not present in any of the samples (*See Figure 4*).

#### *Chloride*

We found that all 51 samples of rainwater contained chloride, ranging from .12 mg/L in Bloomington, IN to 4.21 mg/L in Kurtistown, HI. Samples taken along the East and West Coast tended to have higher concentrations of chloride than samples from the interior of the country (*See Figure 5*).

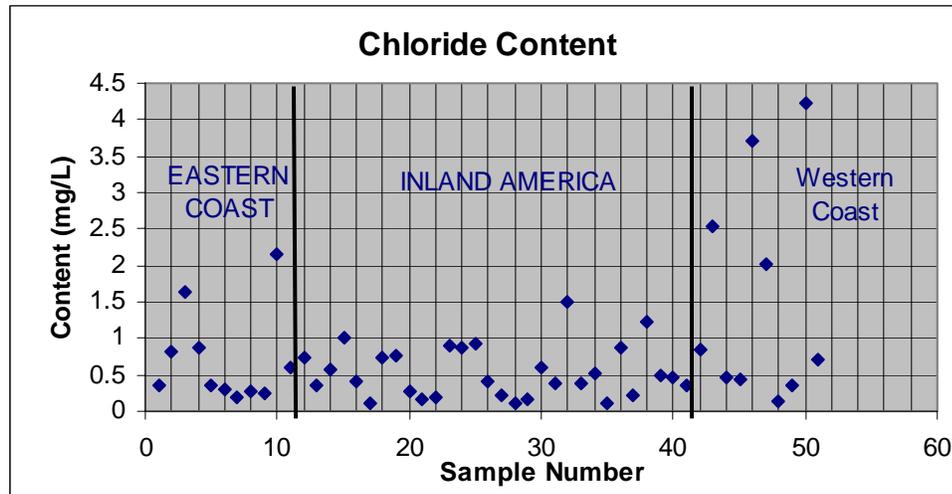


Figure 5. Inland America is defined as West of Pennsylvania and East of Nevada.

*Sulfate (see Figure 6 in Appendix)*

Sulfate was found in 49 of the 51 samples. The Lake Norden, SD and Kenai, AK samples were the only two samples without any sulfate present. The highest concentration found was 5.88 mg/L in Huntingdon, PA.

*Nitrate (See Figure 7 in Appendix)*

Nitrate was found in all but 8 samples. Our largest concentration was found in Northport, Michigan with 8.16 mg/L. The smallest concentration identified was .13 mg/L in Lake Norden, SD

*Phosphate (See Figure 8 in Appendix)*

Phosphate was found in 7 samples. Las Vegas, NV had the highest concentration of 1.14 mg/L.

*Nitrite (See Figure 9 in Appendix)*

Nitrite was only found in 5 of our samples. The largest concentration was .154 mg/L from Fair Banks, CA,

Anions within Northfield, MN

During four different rainfalls we gathered samples on Carleton College's campus for a comparative analysis. In each of the four samples only three anions were found: chloride, nitrate and sulfate (See Figure 10). The concentrations in all four samples varied and there was a wide spectrum of results. Chloride concentration varied from .11 mg/L to .60 mg/L, nitrate concentration varied from .42 mg/L to 1.02 mg/L, and sulfate concentration varied from .56 mg/L to 1.44 mg/L.

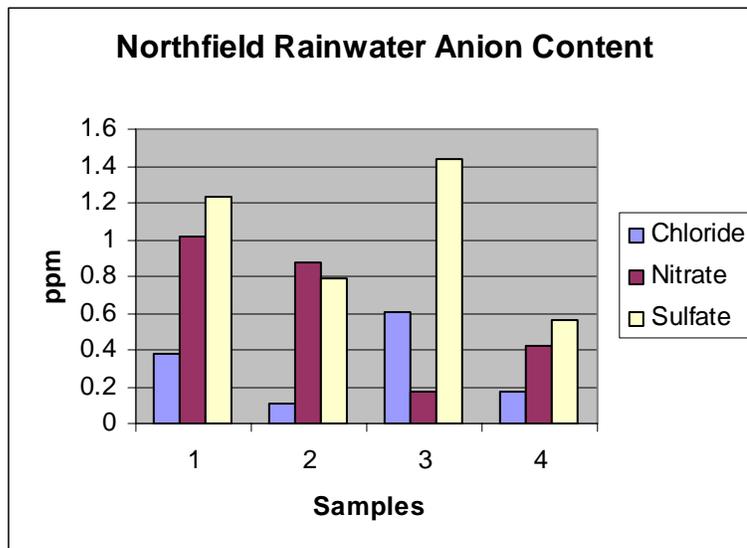


Fig. 10 Rainwater collected in Northfield, MN from 10/1/04 to 10/11/04.

### Oxygen Isotopic Concentrations

The 26 samples tested for oxygen isotope 18 ranged from  $-1.92$  found in Kurtistown, HI to  $-18.66$  found in Great Falls, MT (*See Figure 11*). The standard that these numbers are based on is ocean water which is rich in the oxygen 18 isotope, with an oxygen isotopic 18 value of 0. The results are reported in negative numbers because they are showing how much less of the oxygen 18 isotope is present compared to the ocean water. The lower negative numbers indicate heavier water with a higher concentration of oxygen 18 isotope, whereas high negative numbers signify lighter water where more of the oxygen 18 isotope has been precipitated out. The heaviest water was found along the West Coast.

In the western half of the United States, we observed values of  $-2.62$  in Seattle, WA and  $-2.44$  in Fair Oaks, CA. Moving inland, Great Falls, MT had a dramatically different value of  $-18.66$ . In Minnesota, values averaged around  $-10$ . We observed similar patterns in the eastern half of the United States, with values of  $-2.86$  in Miami, FL and  $-3.18$  in Allentown, PA. Moving inland, we found values of  $-8.49$  in Chicago, IL and  $-8.51$  in Milwaukee, WI.

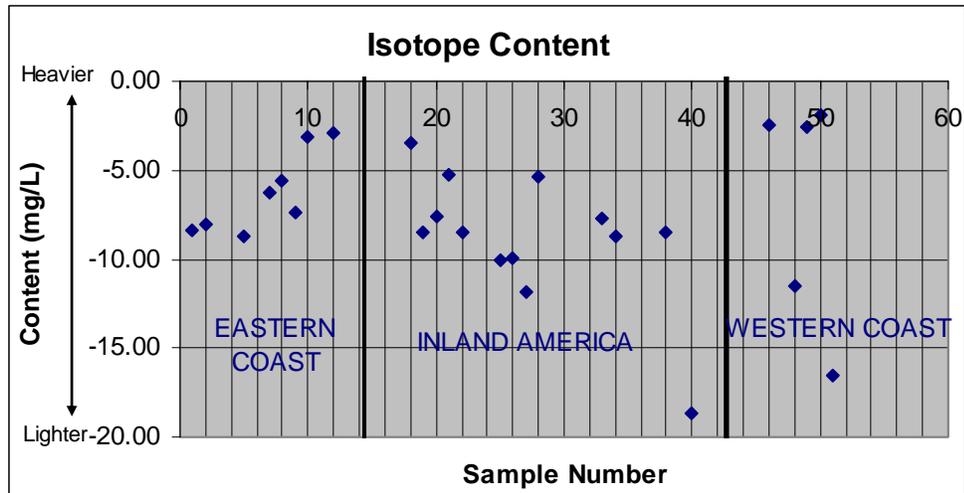


Fig. 11. Inland America is defined as West of Pennsylvania and East of Nevada.

## DISCUSSION

### Anion Concentrations

#### *Chloride (Refer back to Figure 5)*

There is a clear pattern in respect to where we found large concentrations of chloride. We saw higher concentrations of chloride around the East and West Coasts. Kurtistown, HI and Kenai, AK also had high concentrations of chloride. This is because rainwater acquires its chloride content from large bodies of salt water. When oceans evaporate, some anions travel with the water vapor (Avery et al., 2001). Rain generated from the evaporation of the Great Lakes will not have large quantities of chloride because they are fresh water bodies. This explains the lower concentrations of chloride in the Midwest in comparison to water obtained from the coasts.

### *Sulfate, Nitrate, Phosphate, and Nitrite*

Our results for sulfate, nitrate, phosphate and nitrite varied immensely and no firm conclusions could be drawn from them. Geography is apparently not a determining factor for these anions.

### Anions within Northfield, MN

The comparative analysis of the four samples from Northfield gave us conflicting information on how valuable our sampling was. On one hand, the same three anions were present in each of the samples. But the amount of variation within those anions was significant. This led us to believe that our sampling across the country has limited value because we obtained only one sample from each location. It is clear that anion concentrations in one location are affected by a number of variables, including atmospheric conditions. To get an accurate representation of anion concentrations in rainwater for an area, one must take multiple samples over a prolonged period of time.

### Oxygen Isotopic Concentrations

Oxygen isotope 18 is the heaviest variant of isotope present in all rainwater. The rare oxygen isotope 17 is slightly lighter, while oxygen isotope 16 is far lighter. When clouds first form over the ocean from evaporating water vapor, all types of oxygen isotopes are present. As they travel over land, these clouds lose energy and the ability to retain its composition. Due to their weight,  $\delta^{18}\text{O}$  fall to the ground first, resulting in their high concentration in coastal areas and lower concentration in areas further inland. The progressive decrease in  $\delta^{18}\text{O}$  levels in rain as storm systems move across the continent

cause rainstorms to become increasingly lighter (Kendall et al., 2004). This phenomenon is known as the continental effect. We noticed this effect in our data from both the Eastern and Western halves of the United States.

Other factors that affect the distribution of isotopic composition are altitude, latitude and the amount of rain in clouds. Clouds become lighter as they move upward and farther inland (See Figure 12).

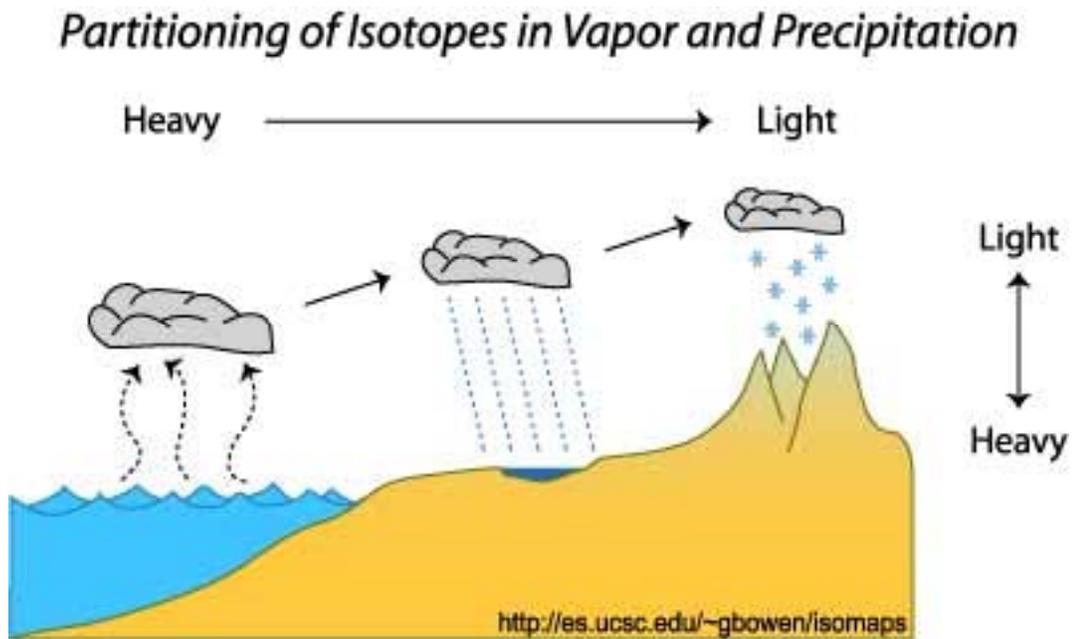


Fig. 12. Altitude and Continental Effect for  $\delta^{18}\text{O}$ . Obtained from Bowen, 2004.

The amount of rainfall also helps determine the distribution of oxygen isotopes across a continent. The greater the amount of rainfall, the lower the  $\delta^{18}\text{O}$  values of the rainfall overall, meaning heavier isotopes will be seen farther inland than usual. Snow is not affected by this process (Kendall et al., 2004). This particular effect explains some of the unusually low oxygen isotope 18 data we received from the East Coast on the 28<sup>th</sup> and 29<sup>th</sup> of September (See locations with asterisks in Figure 2). From mid-August to mid-October a series of 3 hurricanes, Hurricane Charley, Hurricane Ivan and Hurricane

Jeanne, swept up from Florida across the entire eastern Coastline of the United States. Several of our samples were collected from storm systems related to Hurricane Jeanne. Accordingly, they show unusually high amounts of  $\delta^{18}\text{O}$  compared to other samples from the East Coast.

## **CONCLUSIONS**

While it is impossible to make many definitive conclusions on the nature of rainwater throughout the United States, we discovered that rainwater usually contains chloride, nitrate, and sulfate anions, whereas phosphate and nitrite anions were less prevalent. Chloride concentrations correlate with proximity to oceans, which are rich in salt. Levels of  $\delta^{18}\text{O}$  are also dependent on geographic location, as well as altitude, the amount of rainfall, and distance from the source of the rain (usually an ocean). This data provides significant information on rainwater composition in the United States, but additional testing and analysis of our current samples as well as more samples from new locations, particularly for cations and  $\delta^{18}\text{O}$ , may reveal more about the nature of rainwater composition in the United States.

## **ACKNOWLEDGEMENTS**

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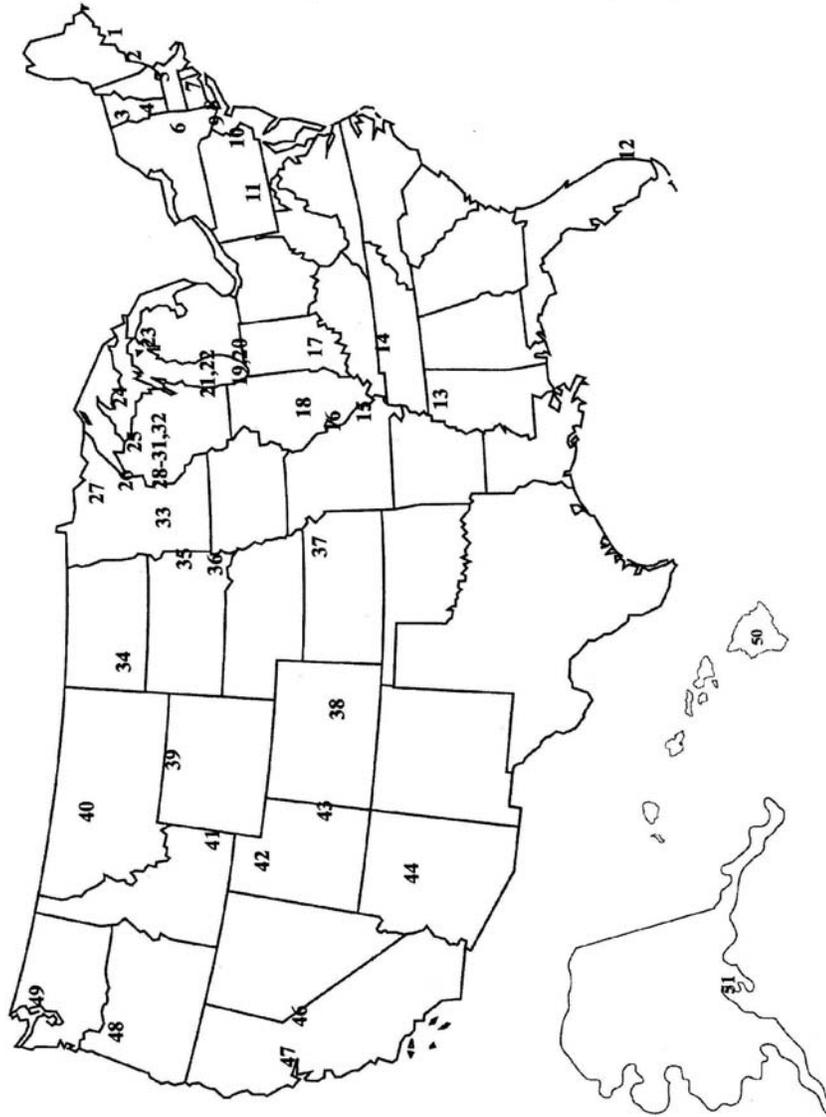
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# APPENDIX

## Distribution of Sampling Sites across the US



State	City	
1	Maine	Yarmouth
2	Maine	Plattsburgh
3	Vermont	Burlington
4	Vermont	Shrewsbury
5	Massachusetts	Concord
6	New York	Troy
7	Connecticut	Hamden
8	New York	Bronx
9	New Jersey	South Orange
10	Pennsylvania	Allentown
11	Pennsylvania	Huntington
12	Florida	Miami
13	Mississippi	Oxford
14	Tennessee	Oak Ridge
15	Missouri	Cape Girardeau
16	Missouri	St. Louis
17	Indiana	Bloomington
18	Illinois	Springfield
19	Illinois	Chicago
20	Illinois	Chicago
21	Wisconsin	Kenosha
22	Wisconsin	Milwaukee
23	Michigan	Northport
24	Michigan	Marquette
25	Wisconsin	Hurley
26	Minnesota	Renville
27	Minnesota	Cohasset
28	Minnesota	Northfield
29	Minnesota	Northfield
30	Minnesota	Northfield
31	Minnesota	Northfield
32	Minnesota	Minneapolis
33	Minnesota	Moose Lake
34	North Dakota	Dickinson
35	South Dakota	Lake Norden
36	South Dakota	Vermillion
37	Kansas	Manhattan
38	Colorado	Colorado Springs
39	Wyoming	Gresham
40	Montana	Great Falls
41	Idaho	Pocatello
42	Utah	Sandy
43	Utah	Moab
44	New Mexico	Albuquerque
45	Nevada	Las Vegas
46	California	Fair Oaks
47	California	Oakland
48	Oregon	Salem
49	Washington	Seattle
50	Hawaii	Kunihon
51	Alaska	Kenai

Fig. 1. Map of the United States and sampling sites.

## $\delta^{18}\text{O}$ Isotope Distribution in the US

Location	$\delta^{18}\text{O}$
Phippsburg, ME	-8.06
Yarmouth, ME	-8.43
Concord, MA	-8.76
Hamden, CT	-6.25
Bronx, NY	-5.64
South Orange, NJ	-7.42
Allentown, PA	-3.18
Huntingdon, PA	-3.37
Miami, FL	-2.86
Springfield, IL	-3.49
Chicago, IL	-8.49
Chicago, IL	-7.32
Milwaukee, WI	-8.51
Kenosha, WI	-5.23
Hurley, WI	-10.10
Renville, MN	-9.89
Cohasset, MN	-11.88
Northfield, MN	-5.38
Moose Lake, Mn	-7.71
Dickinson, ND	-8.72
Colorado Springs, CO	-8.54
Great Falls, MT	-18.66
Fair Oaks, CA	-2.44
Salem, OR	-11.51
Seattle, WA	-2.62
Kurtistown, HI	-1.92
Kenai, AK	-16.59



Fig. 2 Map of the United States and sampling sites for  $\delta^{18}\text{O}$ . Locations with a \* were obtained from hurricane-related storm systems.

### Sample Analysis Report

<b>Sample Name:</b>	Rain47	<b>Sample No.:</b>	674
<b>Sequence Name:</b>	WinterAnions	<b>Injection vol.:</b>	20.0
<b>Program Method:</b>	Geochem3	<b>Dilution Factor:</b>	1.0000
<b>Quantitation Method:</b>	Geochem3	<b>Sample Wt.:</b>	1.0000
<b>Date Time Collected:</b>	11/4/04 10:52 PM	<b>Sample Amt.:</b>	1.0000
<b>System Operator:</b>	Default		

Peak No.	Component Name	Retention Time	Area $\mu\text{S}\cdot\text{min}$	Height $\mu\text{S}$	Amount	Relative Amount %
1	Chloride	3.86	0.171	0.991	0.8705	14.75
2	Nitrate	6.28	0.169	0.666	1.5706	26.62
3	Sulfate	8.88	0.492	1.244	3.4588	58.63

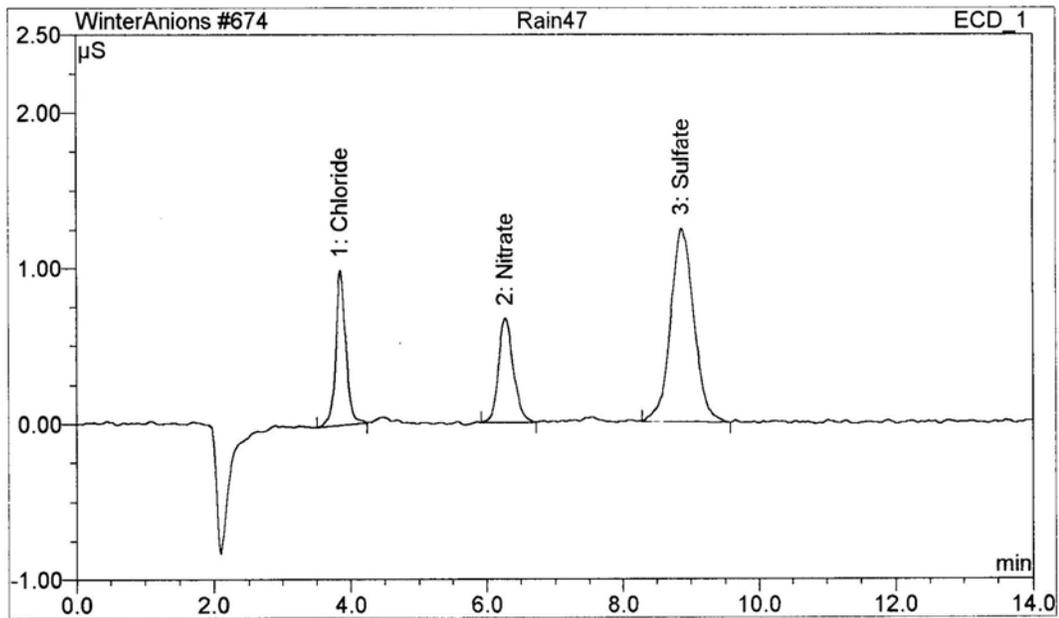


Fig. 3 Sample readoff from the Ion Chromatograph.

## Anion Concentrations

Sample ID see Fig. 1 for location	Fluoride	Chloride	Bromide	Nitrite	Nitrate	Phosphate	Sulfate
1	0	0.3516	0	0	0.2959	0	0.5296
2	0	0.8269	0	0	0.5305	0	0.6654
3	0	1.6465	0	0	0	0.971	2.4932
4	0	0.875	0	0	0.5814	0	0.4658
5	0	0.3625	0	0	0.7039	0	1.0345
6	0	0.3021	0	0	0.3374	0	0.2367
7	0	0.1922	0	0	0.1805	0	0.5225
8	0	0.2782	0	0	0.5478	0	0.8485
9	0	0.2402	0	0	0.3699	0	0.3601
10	0	2.1528	0	0.1369	4.4171	0	5.832
11	0	0.5977	0	0	2.3101	0	5.8884
12	0	0.75	0	0	0.4395	0	0.805
13	0	0.3423	0	0	0.622	0	1.2681
14	0	0.5747	0	0	0	0	1.1311
15	0	1.0009	0	0	0	0	2.7056
16	0	0.4079	0	0	0.5208	0	3.5007
17	0	0.117	0	0	0.5111	0	1.0348
18	0	0.7287	0	0	2.3446	0	4.5204
19	0	0.7537	0	0.117	2.8854	0	4.6738
20	0	0.2815	0	0	0.13408	0	2.1302
21	0	0.1722	0	0	0.7065	0	1.2082
22	0	0.1839	0	0	0.7159	0	0.9392
23	0	0.8936	0	0	8.1632	0	5.4755
24	0	0.8806	0	0	1.7896	0	1.9859
25	0	0.9282	0	0	2.598	0.5051	1.3935
26	0	0.4039	0	0	0.3935	0.7577	0.7253
27	0	0.2306	0	0	0.3363	0	0.5112
28	0	0.1088	0	0	0.8777	0	0.7849
29	0	0.1754	0	0	0.4201	0	0.5573
30	0	0.6046	0	0	0.16994	0	1.4412
31	0	0.3776	0	0	1.0176	0	1.2307
32	0	1.495	0	0.0933	4.8198	0	5.02
33	0	0.3683	0	0	1.5115	0	1.2679
34	0	0.5194	0	0.0943	4.766	0	7.33
35	0	0.1087	0	0	0.1312	0	0
36	0	0.8705	0	0	1.5706	0	3.4588
37	0	0.2218	0	0	0.2181	0	0.2546
38	0	1.2382	0	0	3.0644	0	2.1222
39	0	0.4879	0	0	0.449	0	0.3797
40	0	0.469	0	0	0	0	1.0166
41	0	0.3425	0	0	0.2643	0	0.3153
42	0	0.8546	0	0	1.5821	0	1.841
43	0	2.5376	0	0	0	0	2.5744
44	0	0.4612	0	0	1.0465	0.2569	2.2052
45	0	0.4314	0	0	0.1779	1.1432	0.6908
46	0	3.696	0	0.1537	2.5827	0.5478	2.333
47	0	2.02976	0	0	0	0.2626	1.295
48	0	0.134	0	0	0	0	0.162
49	0	0.3499	0	0	2.159	0	4.1267
50	0	0.7036	0	0	0.2209	0	0
51	0	4.2139	0	0	0	0	0.8609

Fig. 4 Table of Anion Concentrations.

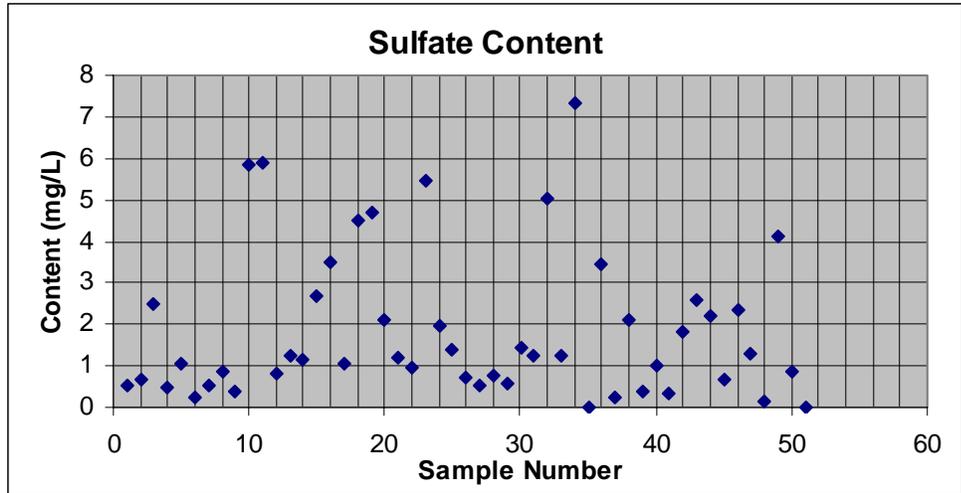


Fig. 6

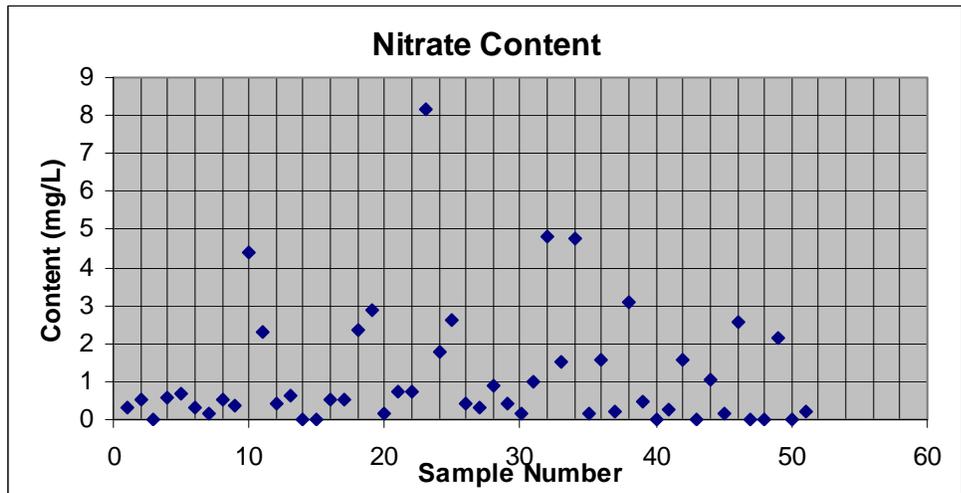


Fig. 7

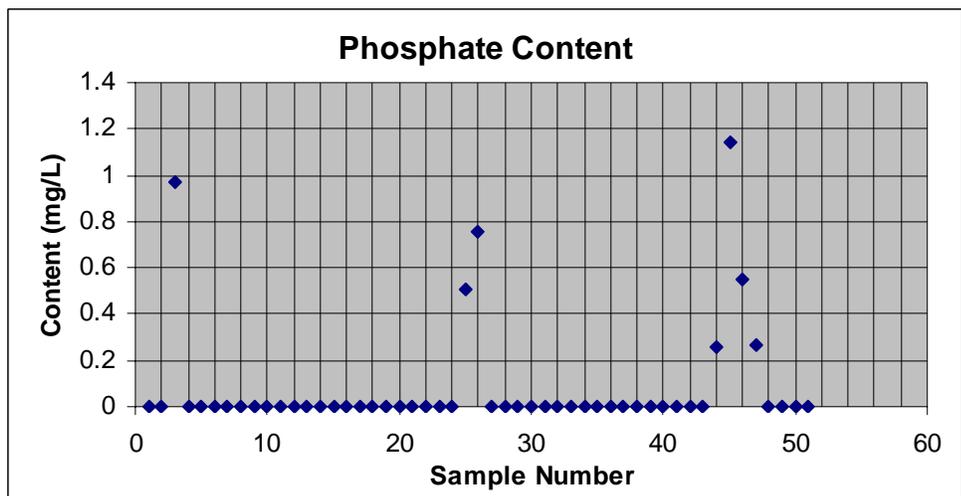


Fig. 8

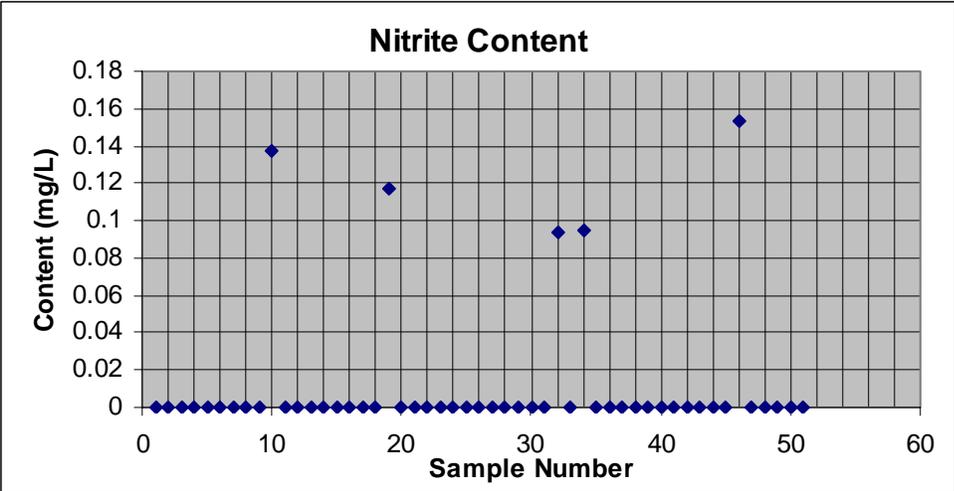


Fig. 9