

ENVIRONMENTAL UPDATE

NATURAL RADIONUCLIDE CONTAMINATION OF POTABLE GROUNDWATER SUPPLIES

By Henry M. Wise, CPG

ABSTRACT

Anomalous amounts of uranium, radium, and radon are known to occur naturally in groundwater in numerous areas around the United States. Granites are well known sources of such elevated concentrations of radionuclides. However, many areas such as portions of the Gulf Coast and mid-continent which do not have granites, contain anomalous uranium, radium, and radon in their groundwater. These occurrences may be the result of "roll-front" type uranium mineralization in sand aquifers. The placement of water supply wells in or near these deposits will result in the withdrawal of radionuclide-contaminated water from the aquifer. Identification of these deposits will assist in the avoidance of them. Various methods of identification are also discussed.

INTRODUCTION

Federal law limits the maximum drinking water concentrations of several radionuclides, including gross alpha (excluding uranium and radon), and gross beta particle activity. Radon and uranium will soon be added to this list. As a result, numerous wells will have to be abandoned and re-drilled, or chemically treated.

It is advantageous to predict where unacceptable levels of radionuclides will occur prior to drilling. Many areas of the United States, primarily in the Northeast, are known to have high concentrations of radionuclides in groundwater associated with granites (Graves, 1987). Other areas, such as the Texas Gulf Coast and mid-continent, also have anomalous radionuclide concentrations, however the source has not been fully identified.

Radionuclides found in fresh-water, alluvial aquifers located at some distance from a granitic source typically occur in a "scattered" pattern. One well may contain anomalously high amounts of radionuclides while a nearby one may not. Also, the radionuclides may vary in concentration from well to well. This scattered pattern is typical of "roll-front" type uranium mineralization.

"Roll-front" type uranium mineralization is typically sinuous and narrow and results from geochemical reactions. Uranium is leached from source rocks (granite, ashfall tuffs, shales, etc.) by oxygen-rich groundwater and meteoric waters. Once leached, the soluble oxidized uranium migrates down-gradient until it reaches a reducing environment, which may be natural gas, oil, or coal. Uranium is insoluble under reducing conditions. If the oxidation-reduction (redox) boundary remains in place for any significant length of time, "roll-front" mineralization will form. If a water-supply well were to penetrate this deposit, a significant amount of radionuclides would be found in the groundwater supply. It is therefore important to understand

how and why these deposits occur so that they may be avoided. Sampling methods already exist which may be utilized to predict where these deposits may occur.

DESCRIPTION OF ROLL-FRONT URANIUM MINERALIZATION

Possible sources of uranium are granites, volcanic sequences, bentonitic and volcanoclastic sediments, and basement metamorphics (Adams and Smith, 1981). Black shales such as the Chattanooga Shale and its stratigraphic equivalents are also rich in uranium (Ahmad and Finlay, 1987). The uranium in these sequences is easily leached by any meteoric or oxygen-rich groundwater which penetrates it. Uranium levels in such groundwater are extremely low.

Uranium in the oxidized state typically exists in solution as either the uranyl ion, UO_2^{2+} , or as a carbonate complex (Levinson, 1974). The precipitation of uranium is a direct function of pH and Eh, as shown in Figure 1. Uranium becomes insoluble when the Eh becomes low or negative. The source of this reduction may be natural gas, hydrogen sulfide, oil, or coal (Levinson, 1980, and Adams and Smith, 1981). The redox boundary acts as a sieve, allowing the groundwater to pass through, but removing uranium in the process. If this boundary remains in place for a significant period of time, a build-up of uranium mineralization will occur.

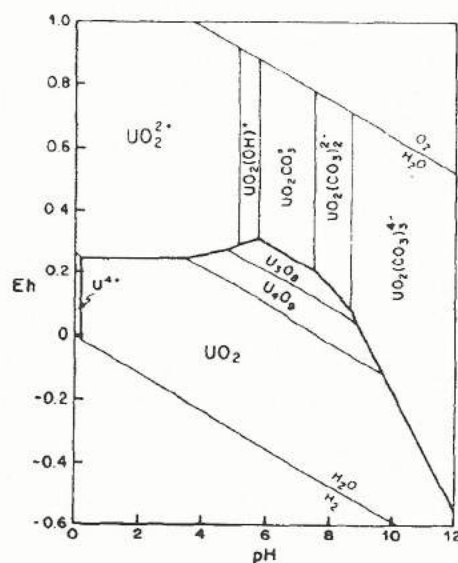


Figure 1. Eh-pH diagram for the system U-O-H-CO showing the mobility of the uranyl complexes, and the stability fields of three solid uranium oxides (uraninite or varieties), at 25 C. U = 1 ppm; P = 10 (Levinson, 1989).

With time, the oxidizing waters will deplete the area of reduction. As this occurs, precipitated uranium will redissolve, move down-gradient and reprecipitate. This movement causes the mineralization to eventually take on a roll-, or C-, shape because the groundwater will flow faster through the more permeable portions of the sand unit. Also, uranium will absorb onto the confining clays and therefore be further slowed along the edges of the aquifer (Levinson, 1980).

Adams and Smith (1981) have shown that mineralization distribution trends are controlled by broad-scale sedimentary features such as mega-channel systems or beach sands. Mineralization is often found along the flanks of these features. Roll-front mineralization typically extends for miles, but is often less than several hundred feet wide.

Figure 2 illustrates a typical cross section of a uranium roll-front deposit. Groundwater movement is from left to right. There are easily distinguished and definite patterns to roll-front mineralization, as indicated by color and mineral content.

Gamma-ray logs of drill holes show increasingly high and broad peaks until the main portion of the deposit, or "nose", is reached. Up-gradient of the "nose," the gamma peak separates into two peaks, or "tails." These "tails" are located at the top and bottom of the aquifer. Down-gradient, the gamma-ray peak remains broad but its intensity decreases.

Sediment color is another important method for determining the location of the well in relation to the mineralization. Because uranium precipitates under approximately the same conditions as iron changes oxidation states, the color of iron is a good marker for the in situ Eh. As shown in Figure 2, iron is commonly found as pyrite (FeS_2) in the reduced portions of the roll-front, as limonite at the redox boundary, and hematite between the "tails." Pyrite becomes increasingly pitted and tarnished towards the "tails" until it has completely dissolved.

The oxidation of iron from pyrite to hematite results in the characteristic sediment color change from green or blue to yellow, brown, or red. If the roll-front is moving quickly down-gradient, these colors may be diffuse, or overlap because it takes a set period of time for limonite to fully dehydrate to hematite.

Given sufficient time, uranium decays to lead, and in the process, give rise to a series of isotopes known as daughter products. The daughter products of most concern are radium and radon. Radium occurs as a solid, whereas radon occurs as an inert gas.

There are no daughter products associated with the uranium at its initial precipitation from the groundwater. According to Harshman and Adams (1981), if no further uranium mineralization occurs, 250,000 years will pass before the uranium in the deposit attains equilibrium with its daughter products. Once equilibrium is attained, the

Typical "roll-front" type uranium mineralization (Adapted from Campbell and Biddle, 1977).

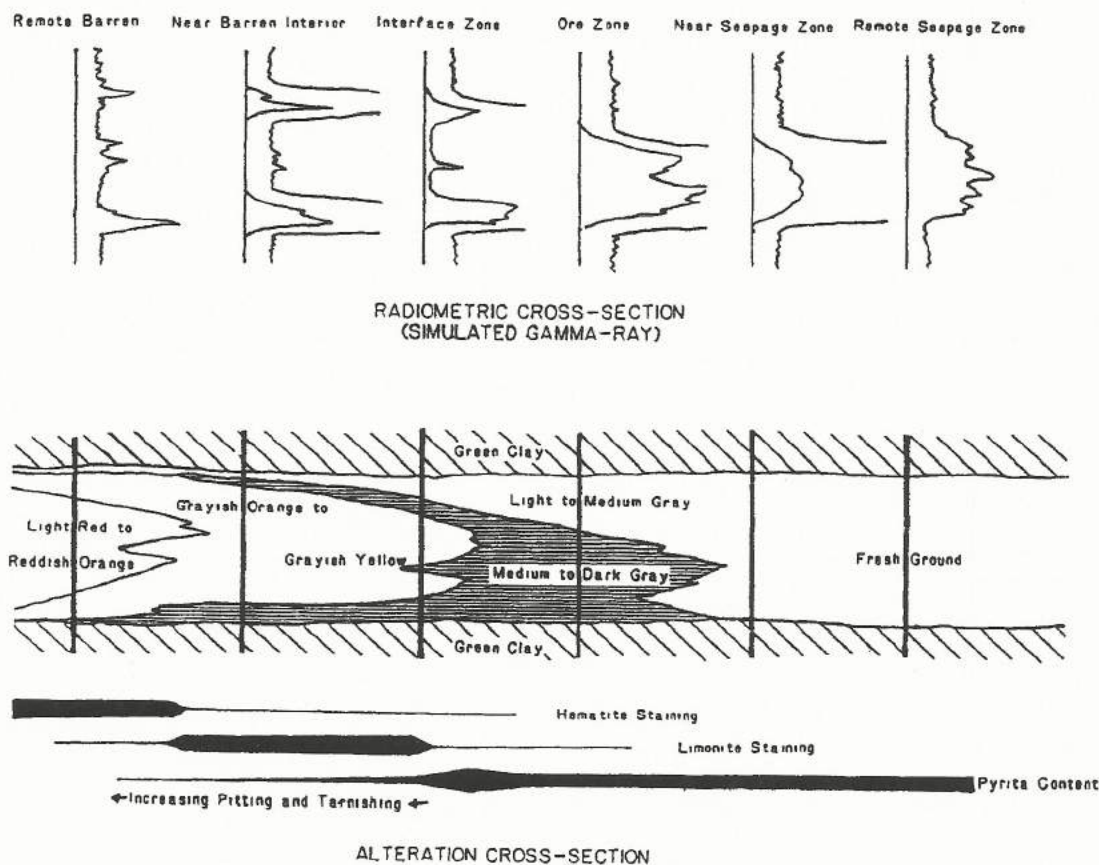


Figure 2. Typical "roll-front" type uranium mineralization (adapted from Campbell and Biddle, 1977).

amount of radiation shown on a gamma-log is proportional to the true amount of uranium available. Exact equilibrium is seldom attained in nature, however, because groundwater is constantly redissolving and reprecipitating the uranium. The net result is that the equilibrium of uranium fluctuates with time. In general, a negative equilibrium ($U_{\text{ACTUAL}}/U_{\text{CALCULATED}} < 1.0$) will occur in oxidized portions of the mineralization, while a positive equilibrium ($U_{\text{ACTUAL}}/U_{\text{CALCULATED}} > 1.0$) will be found at the redox boundary. Any precipitated uranium will be redissolved if oxidizing water should reach it. Since uranium is generally more readily oxidized than its daughter products, redissolution of uranium will result in the uranium moving down gradient, the daughter products remaining in-place, and an equilibrium less than 1.0.

Because uranium is mobile under oxidizing conditions while the daughters are not, there are often more daughter products than uranium in the barren, or "tails", area of the mineralization. Anomalously high uranium concentrations may be found in groundwater samples taken from this area. Radium, which is soluble under reducing conditions will be found as a precipitate rather than in solution (Morse and Cook, 1979). Radon concentrations increase directly with uranium or radium concentrations. It will be carried down gradient until it dissipates. According to Soonawala and Telford (1980) if only diffusion increases are active, radon will dissipate within one or two hundred feet of its generation. If however convective processes are active radon attenuation is decreased by a factor of as much as 800. As a result, it may be detected several thousand feet down gradient from the mineralization.

The "interface zone" of the mineralization is the most reactive portion of the roll-front and is located at the redox boundary. Here, uranium is actively remobilizing and reprecipitating. Radium is primarily found in solid form here, although some dissolution may take place. Radon concentrations are directly proportional to the concentrations of uranium and radium.

In the "nose", "seepage zone", or "front", areas, uranium exists primarily as a precipitate, as reducing conditions are now the rule. Radium, which is soluble under reducing conditions, may be found in anomalously high concentrations in the groundwater (Beck and Brown 1987; Szabo and Zapecza, 1987). Radon concentration is highest in the "nose" area of the mineralization as uranium and radium concentrations are highest here. All radionuclide concentrations fall dramatically in front of the "nose" area.

METHODS FOR IDENTIFICATION OF DEPOSITS

Various methods may be employed to delineate mineralized areas. Gamma-ray logs and chemical analyses of existing wells are two methods which usually have the information readily available. The gamma-ray log reveals the presence of any radionuclides. When properly interpreted, several logs can reveal the shape and nature of the mineralization. Useful chemical information includes uranium, radium, radon, gross alpha, gross beta, dissolved oxygen concentrations, and Eh. Water supply wells are analyzed for many of these parameters on a regular basis.

Additional information may be obtained by drilling exploration or production wells. Besides gamma-ray logging and analyzing the groundwater in the new well, the drill cuttings or core should be properly identified. Often there is

ADDITIONAL OPENINGS AVAILABLE

WELL LOG SEISMIC SEQUENCE STRATIGRAPHY COURSE

Peter R. Vail, Ph.D., Maurice Ewing Chair, Rice University
Walter W. Wornardt, Ph.D., Adjunct Prof., Rice Univ.; President, MICRO-STRAT INC.

Friday and Saturday, February 9-10, 1990, 8:00 am-12:00 pm 1:30 pm-5:30 pm
Doubletree Hotel at Post Oak, 2001 Post Oak Blvd., Houston, Texas

Friday and Saturday, March 23-24, 1990, 8:00 am-12:00 pm 1:30 pm-5:30 pm
Location to be announced, Dallas, Texas

Friday and Saturday, April 27-28, 1990, 8:00 am-12:00 pm 1:30 pm-5:30 pm
Location to be announced, New Orleans, Louisiana

Course Description:

- Identification of sequence boundaries, system tracts, condensed sections, geologic age, paleoenvironments, etc. on well logs from Gulf of Mexico data sets.
- Utility of high resolution micropaleontology in the determination of geologic age, paleoenvironments, condensed sections, system tracts, etc.
- Interpretation of paleoenvironments and lithofacies from well log patterns.
- Participation in an eight-hour, hands-on practical application workshop of Sequence Stratigraphy tied to a Seismic Record Section.

Registration fee is \$295 per person

Each session limited to 64 participants from oil companies
or independent geological/geophysical consultants.

A portion of your registration fee will be donated to the Rice University
Sequence Stratigraphy and Micropaleontology Research Fund.

For additional information, MICRO-STRAT INC., 5755 Bonhomme, Suite 406
or to register, contact: Houston, TX 77036, (713) 977-2120

sufficient information in the drill-cuttings samples obtained every five feet to estimate the in situ Eh at a particular depth.

DISCUSSION

Placement of a water supply well into an area of uranium mineralization is an important reason why anomalously high radionuclides are found in drinking water. The location of a well in relationship to the various zones of mineralization will determine which radionuclides may be expected to be found in the groundwater.

Figure 3 illustrates the types of radionuclides that may be expected in water supply wells pumping groundwater from "roll-front" type uranium deposits. Although wells A and C will have a lower radionuclide concentration than well B, wells D and E will have none. As wells A and C are located further from the "nose" of the mineralization, the concentration of the radionuclides in the groundwater will become less.

Well A draws water from the "tails" side of the mineralization. Groundwater here is oxidizing. Therefore, uranium and radon are the major radionuclides found in the groundwater. Anomalous concentrations of uranium and radon will increase as the position of well A moves closer to the "nose" area. This is because the area of influence of the cone of depression for A will cover an increasingly larger volume of the mineralization. As more mineralization is covered by the cone of depression, more of the withdrawn groundwater passes through the mineralization, redissolving and sweeping the radionuclides into the well bore.

Well B represents the position of a well where the maximum concentration of radionuclides would be expected. Maximum concentrations of uranium, radium, and radon in the groundwater occur here because this is the

area of maximum chemical activity in the mineralized area. The cone-of-depression for well B extends across both sides of the redox boundary, so both uranium and radium are being redissolved and precipitated.

As well placement changes in favor of the position of well C, the groundwater Eh becomes more reducing. The cone of depression therefore draws in increasingly reduced waters, so uranium concentrations decrease to zero and radium concentrations increase.

The concentration of all radionuclides in water wells farther from the area of mineralization drops until the wells are no longer influenced by the deposit, as illustrated by wells D and E.

CONCLUSIONS

"Roll-front" type uranium mineralization is an important factor in causing radionuclide contamination of groundwater. These deposits are characterized by changing in Eh from positive to negative in the down-gradient direction, being narrow and sinuous, and having a C-shape. The rate of change in Eh, as well as rate of groundwater flow, controls the concentration of the mineralization, as well as its width. A groundwater production well placed in or near one of these deposits will yield groundwater with varying degrees and types of radionuclide contamination. A knowledge of where and how these deposits occur is useful in the proper placement of water wells.

(REFERENCES)

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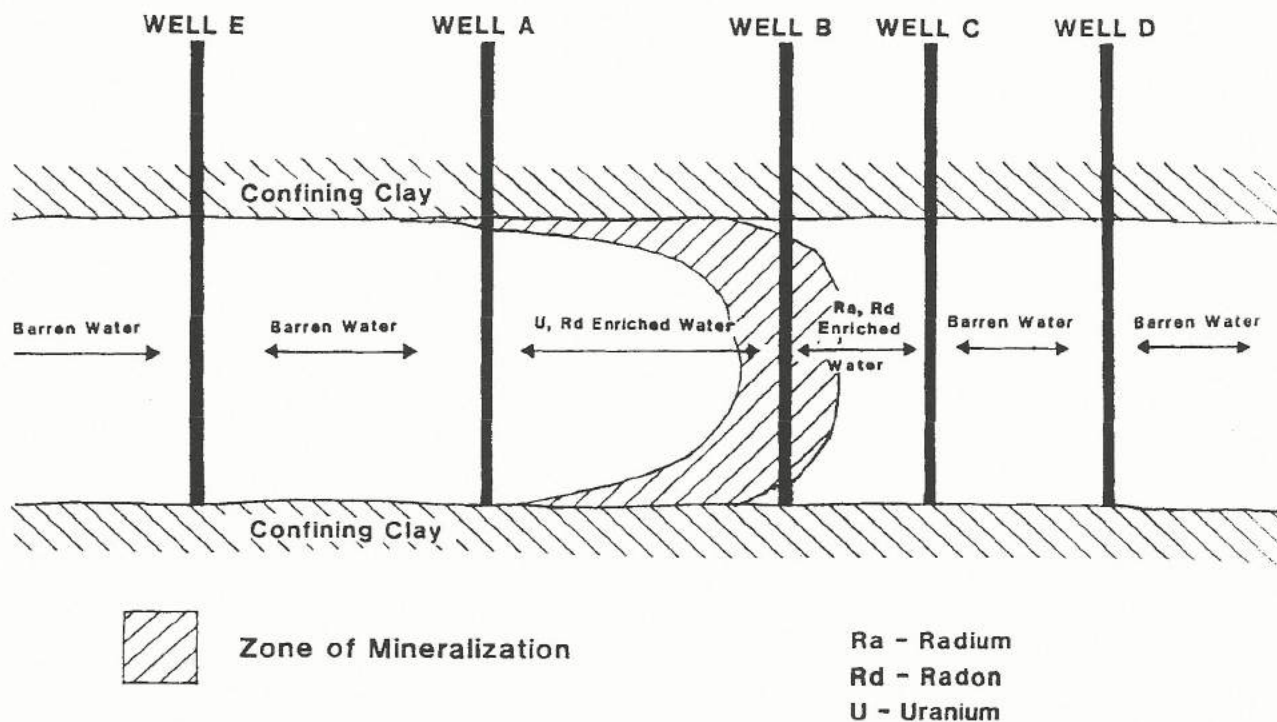


Figure 3. Importance of placement of water wells relative to "roll-front" type uranium mineralization.

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HOUSTON GEOLOGICAL SOCIETY ENVIRONMENTAL/ENGINEERING GEOLOGY COMMITTEE FIELD SEMINAR

LANDFILL AND LANDFARM TOUR, AND WATER SAMPLING TECHNIQUES ASSOCIATED WITH RCRA PERMITTING PROCESS FOR LANDFILL

- Date:** March 17, 1990, 9:00-NOON
- Location:** Gulf Coast Waste Disposal Authority
1600 Campbell Bayou Road, Texas City
- Scope:** A tour of the Gulf Coast Waste Disposal Authority facility (landfarm and landfill), noting construction and RCRA permitting process for hazardous and non-hazardous waste landfill, and RCRA closure process for landfarm. Demonstration of RCRA groundwater sampling techniques and procedures.
- Speakers:** Gulf Coast Waste Disposal Authority personnel
Groundwater Technology, Inc.
- Who Should Attend:** Anyone contemplating entering the environmental business, or anyone just interested in how a landfill/landfarm looks and operates.
- Cost:** \$5.00 HGS members
\$8.00 Non-HGS members

HOUSTON GEOLOGICAL SOCIETY ENVIRONMENTAL/ENGINEERING GEOLOGY COMMITTEE FIELD SEMINAR

DRILLING AND SOIL SAMPLING TECHNIQUES, AND MONITOR WELL INSTALLATION

- Date:** February 17, 1990, 10:00 - 2:00
- Location:** Meet at Environmental Drillers, Inc.: located at 9871 Tanner Road, Ste. E
(office is on Tanner between Beltway 8 and Campbell Road, west of Hempstead Highway)
- Scope:** An overview of monitor well drilling techniques and soil sampling methods commonly used in environmental site assessments.
- Demonstrators*:** Environmental Drillers, Inc.
Groundwater Technology, Inc.
*Well supplies provided by Hughes-Beard Company, Inc.
- Who Should Attend:** Anyone contemplating entering the environmental business.
- Lunch:** Food and refreshments will be provided by Environmental Drillers, Inc.
- Cost:** \$5.00 HGS members
\$8.00 Non-HGS members

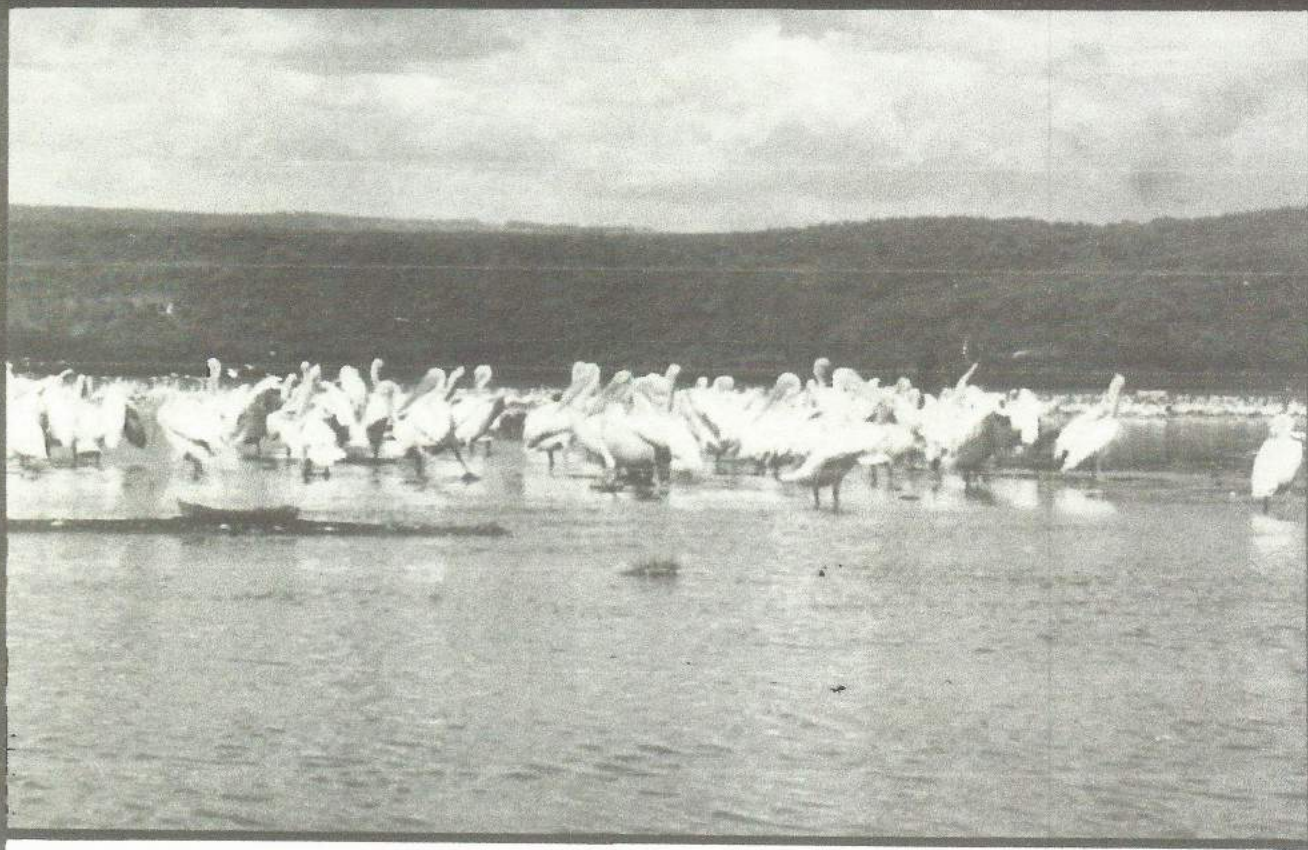


February, 1990

BULLETIN**HOUSTON GEOLOGICAL SOCIETY**

Volume 32

Number 6

***IN THIS ISSUE...***

- Have Passport, Will Travel Page 12
- Natural Radionuclide Contamination Page 17
- Early Exploration for Hydrocarbons in the
North American Arctic Page 22
- Let's Get Rid of A Dumb Exploration! Page 26
- Computer Standards Concepts in the AAPG Page 40

PLUS MORE!

(For February Events, see page 1 and Geoevents section, page 31)