

POTENTIAL OF A NEW TECHNIQUE FOR
REMOTE SENSING OF HYDROCARBON
ACCUMULATIONS AND BLIND URANIUM
DEPOSITS: BURIED LiF
THERMOLUMINESCENCE DOSIMETERS

F.R. SIEGEL, Department of Geology, The George Washington
University, Washington, D.C. 20052 USA
J.E. VAZ, Instituto Venezolano de Investigaciones Cientificas,
Apartado 1827, Caracas, Venezuela
R.C. LINDHOLM, Department of Geology, The George Washington
University, Washington, D.C. 20052 USA

ABSTRACT

Buried thermoluminescence dosimeters may be useful in remote sensing of petroleum and natural gas accumulations and blind uranium deposits. They act as integrating detectors that smooth out the effects of environmental variations that affect other measuring systems and result in irregularities and poor repeatability in measurements made during gas and radiometric surveys.

INTRODUCTION

The radiometric survey is the primary prospecting method for uranium deposits. The radiometric survey is an adjunct technique in prospecting for petroleum and natural gas accumulations and would be especially valuable in lowering the front end exploration or field extension costs.

In theory, gases generated by the radioactive decay of uranium or thorium ore ascend through the overlying rock cover and/or soil system. The gases radioactive signals are detected at the surface or from fixed wing or helicopter aircraft (with a gamma-ray spectrometer or scintillometer). When a measured radiation field exceeds the "normal" field by an arbitrarily selected concentration, the explorationist has a target area for further detailed exploration.

The depositional environment of accumulation for petroleum and natural gas precursors is one that allows for the accumulation of uranium compounds. The uranium materials associate with the petroleum and natural gas containing rocks. These hydrocarbon accumulations contain the gases from the radioactive decay process. In theory, these gases should rise through the overlying rock and soil system and give radioactive "hot spots" in a halo or fan-shape. Since the radioactivity highs are believed to mark the perimeter of a hydrocarbon accumulation, the associated low concentrations would mark the exploration targets. Figure 1 illustrates this situation.

In the case of radioactivity patterns associated with uranium and thorium accumulations, the target will manifest itself as an anomalous concentration directly above the radioactive mineralization unless structural geology characteristics (dip or fracture, a

joint-fault-crack-crevasse system), hydrodynamic conditions, or coning effects from diffusion result in "offset" anomalies (Figure 2).

The radioactivity concentration patterns that are related to petroleum and natural gas accumulations are peripheral to the accumulations or form halo-shaped targets enclosing or nearly enclosing them. The reason for the peripheral or halo-shaped target lies in the genesis of petroleum and natural gas deposits. In the subsurface, the hydrocarbons form from their precursors in a "source" rock unit. The petroleum and natural gas have a much lighter specific gravity than the enclosing rocks. If the rocks are porous and permeable (able to hold and transmit fluids), the hydrocarbons with their associated radioactive gases will be mobilized upward along the density gradient until they contact an impermeable rock unit or "trap". Accumulation will take place at the trap. Where accumulation stops, at the borders of the traps where there is porosity and permeability or cracks or fissures in the rock unit, the associated radioactive gases will escape and rise vertically to the surface where peripheral or halo-shaped radioactive concentrations in excess of the natural radioactive field will develop (Figure 3).

Radiometric prospecting for uranium and thorium ores and for petroleum and natural gas has a good theoretical base. The technique has been imminently successful in the exploration for radioactive minerals. Most major deposits have been targeted using this method especially when they are not covered by great thicknesses of rock or other overburden. In petroleum and natural gas exploration, the technique has not been as successful, and is considered unconventional. It is used only as an adjunct to the accepted approaches to exploration for hydrocarbons. The major problem that exists in radiometric prospecting is one of repeatability. If the results are not reproducible in assessing the radioactivity signals in a survey, something is wrong either in the theory or in the measuring technique or equipment. Since the theory is good, the lack of repeatability in measurement must lie in the measuring technique, timing, or the environment.

PROBLEMS

Although excellent instrumentation has been developed to measure the transient radioactivity field (portable, vehicle mounted and airborne gamma-ray spectrometers, scintillation counters, ionization chambers, and others), geochemical prospecting based on analysis of gases (e.g., Rn) or radiometric signal from the decay products from the uranium and thorium chains may be difficult to carry out. The problem of repeatability of measurement mentioned above affects confidence in results and subsequent exploration interpretation. This problem exists if insufficient amounts of gases are collected in short term sampling, if radioactivity signals are low, or if interferences result from short term environmental variations caused by meteorological and/or seasonal processes. Such variations can occur in air temperature, soil temperature, barometric pressure, time of sampling, wind, precipitation (rain, light snow, heavy snow), position of the water table, relative humidity, soil moisture, the frozen or thawed state of the soil, diurnal cycles, solid earth tide, orientation of slope sites, and thickness of

overburden (McCarthy, 1972; Klusman and Webster (1981)).

Klusman and Webster (1981) have demonstrated some of these effects beautifully in their study of free mercury vapor emission and radon emanation in a shallow instrument vault set and sealed in weathered regolith at a single non-mineralized site. The site is about 30 miles west of Denver, Colorado, at an elevation of 2640 m. Bedrock at the site is Precambrian gneiss. For example, Figure 4 compiled from Klusman and Webster (1981) shows the seasonal trends for Hg emission at the vault site and that Hg emission may vary by an order of magnitude (from <0.4 ng to >4.0 ng). Figure 4 also illustrates some multivariate interactions and phase lag effects in the system. The diurnal effect on Hg emission as a function of air temperature and daylight hours is shown in Figure 5 where there is a range of ng Hg from <0.25 to >1.5 .

Environmental variations that affect the precision of mercury emission measurements also affect gas-coupled radioactivity signal. Thus, it is obvious that geochemical exploration based on radioactivity measurements can result in false targets or can miss targets because of environmental variations or the other factors cited previously.

RESOLUTION OF THE PROBLEMS

The problems of insufficient signal measurable during short term sampling of surface radioactivity, and environmental interferences that affect repeatability of measurements and hence limit the usefulness of some exploration techniques must be resolved. Also, deep weathering or burial beneath a meter or more of colluvium or other transported cover masks the radioactivity from the underlying bedrock and hinders exploration. The problems may be obviated by the use of an integrating detector that can be left in the field for long periods of time. This could be used to establish background radioactivity for a near surface soil environment. With the background radioactivity level thus determined, and following arbitrary norms to fix radioactivity concentrations that encompass local and/or regional fluctuations, the "anomaly" radioactivity concentration can be set. In geochemical exploration, a measured value that exceeds the mean plus two standard deviations is often set as the threshold value above which a measurement may be considered an anomaly.

An integrating system has been used for soil gas radon determinations in uranium exploration using α -track cellulose nitrate film placed in inverted plastic cups. The cups are placed open end down in a hole 0.4 to 1.0 m deep for about three weeks. The number of α -track registered for a given area in a known period of time is a function of the amount of radon at the sample site. However, these cups may be sensitive to moisture condensing in them, and rainy weather and persistent seepage downward effectively prevent Rn from reaching the point of α -track registration. The unit cost of a cup and "reading" the α -tracks is about \$35. if a private company is used. The user must bury the cups and recover them.

We propose that thermoluminescence dosimetry is an integrating system that will work effectively in geochemical prospecting via

radioactivity measurement for uranium and thorium mineralization and for petroleum and natural gas accumulations. When semiconductors or insulating crystals interact with ionizing radiation, electrons are displaced from their ground state, or minimum energy levels, to higher, or activated, impurity or imperfection levels. The subsequent heating of these materials permits the displaced electrons to fall to a lower energy state or their ground state resulting in the emission of energy in the form of heat and light. The light is termed thermoluminescence. The amount of thermoluminescence accumulated is a function of the amount of radiation to which a crystal has been exposed.

Thermoluminescence dosimetry to establish near surface radiation levels has not been used in petroleum and natural gas prospecting. However, the potential of thermoluminescence for uranium prospecting is not new. Nielsen and Botter-Jensen (1973) used thermoluminescence to evaluate natural radiation over rock units in Greenland. Geologists on mapping projects carried $\text{CaSO}_4:\text{Dy}$ dosimeters in their pockets in the field for three months. The radioactivity levels measured represented integrated doses for the field areas traversed. An evaluation of the dose rates measured over different rock types (crystalline rocks, sedimentary rocks, and basalts) and their average radioactive element contents indicated that the method may be useful for large scale regional prospecting. In a study in Texas, thermoluminescence of the minerals quartz and feldspar gave a ratio of lower temperature thermoluminescence to higher temperature thermoluminescence that was consistently higher in ore and the reduced rock zone of a roll-type uranium deposit than in the oxidized rock zone (Spirakis et al., 1979).

We employed buried LiF thermoluminescence dosimeters to establish background radiation levels for environmental monitoring in northern Virginia (Siegel et al., in press). The dosimeters (3.2 x 3.2 x 0.9 mm in size) were buried at 45 cm (18") depth in waterproof plastic bags and covered with soil. Of the 101 dosimeters buried, 92 were recovered after about four months. The registered radioactivity was determined by comparing the accumulated thermoluminescence against a calibration curve which was made by exposing dosimeters to known amounts of ionizing radiation (from Co-60) and reading their thermoluminescence outputs (with an analyzer). The buried dosimeter integrated dose rates ranged from 0.06 to 1.08 mR per day (or 2.5 to 44.5 μR per hour), an 18 fold difference. In the study, background radiation levels were also determined with a gamma-ray spectrometer but are better defined with buried thermoluminescence dosimetry. Two anomalies were found using the dosimeters which were not indicated by the gamma-ray spectrometer data.

We may speculate why anomalies indicated by thermoluminescence were not indicated by gamma-ray spectrometry or why the correlation coefficient between the two sets of data was not better than the calculated +0.64. First, the minimum dose rate that can be registered by the dosimeters (in R per month or in the cited study, 2.5 μR per hour) is below the detection limits of most gamma-ray spectrometers (or for that matter scintillometers). Second, the field spectrometer detects only gamma rays with energies that fall within a specific count mode so that the loss of a parent isotope affects the concentration of the daughter products in the soil system. However, the thermoluminescence dosimeters register even

the less energetic gamma rays from the decay products of the uranium and thorium series. Third, the radioactivity measurements are more accurate with the buried thermoluminescence dosimeters than with the electronic field equipment. This is because the dosimeters act as integrating detectors that dilute the effects of short-term environmental changes cited previously. Measurements made with the gamma-ray spectrometer are responsive to these changes.

There is no doubt that for the reasons cited above and another we will give that this use of buried thermoluminescence dosimeters in geochemical prospecting for uranium mineralization and hydrocarbon accumulations should be field tested. This would be especially important where gamma-ray spectrometer or scintillometer methods have been used but have failed to define a uranium deposit or an oil field. For example, at the Number Three Orebody, Ranger One, Australia, various remote sensing techniques were tested in the exploration for this U_3O_8 deposit (Table 1). Some were successful, and some not (Sherrington et al., 1982). Airborne gamma radiometry and ground radiometrics gave positive responses to the Ranger One Complex. However, in the same Alligator River Province, the Jabiluka II body, the largest known uranium deposit in the world is essentially blind to all methods not involving drilling. It is also undefined on two sides (Nash et al., 1981). The Jabiluka II body would be an ideal place to test the buried thermoluminescence dosimeter technique for uranium prospecting.

The testing of the buried thermoluminescence dosimeter technique for oil and/or gas exploration could be done at any known field which has had or presently has production. Weart and Heimberg (1981) report excellent results of radiometric surveys in known producing fields using a vehicle mounted 5 1/2 foot long ionization chamber. Truck mounted gamma ray spectrometers with large detector crystals (to 2000 cubic inches) are also in use (Geoprofiles, 1982). These systems are useless in areas inaccessible to surface vehicles. Such areas are generally accessible to a ground field party. Where the radiometric survey is the principal method to find radioactive mineral deposits, it is used as part of an integrated program in petroleum exploration to lower front end costs. For example, it can be used to highlight limited areas in a large exploration zone for seismic study and thus avoid seismic work in the entire zone.

A fourth reason for establishing the technique is that since the thermoluminescence radiometric survey requires only the planting of unexposed dosimeters and later recovery of the exposed dosimeters, the survey can be carried out during periods of adverse field conditions. Also areas which are inaccessible to vehicle mounted equipment are accessible to the explorationist who can carry hundreds of the thermoluminescence dosimeters into the field. For example, in tropical rain forests, dosimeters can be planted before the rainy season begins and be collected a few months later when field conditions are favorable. At another extreme, we buried thermoluminescence dosimeters in the Antarctic during the past austral summer; these will be recovered and read next December or January.

Finally, we would like to note that the technique would seem to be cost effective and once the dosimeters are recovered, the data they carry can be obtained rapidly. The dosimeters cost about \$2.00 each and are reuseable. They can be read at a rate of about 100 per

Table 1. Remote Sensing Parameters Used in Exploration of the Number Three Uranium Orebody, Ranger One, Australia (compiled from Sherrington et al., 1982). The symbols "+", "+" and "-" summarize (with qualifications) the success of each method, from useful through equivocal to non-responsive.

AIRBORNE	Landsat Photographs - no mineralization related features	
	Aerial photography - no obvious correlation of any feature with mineralization	
	Airborne gamma radiometric surveys - give a very strong response (mineralization intersects the land surface)	
	Airborne magnetic surveys - do not delineate the bodies themselves	
GROUND	Regional gravity surveys: assist defining boundaries	
	Ground magnetics	-
	Ground radiometrics	+
	Gravity	<u>+</u>
	V.L.F. electromagnetics	-
	Transient electromagnetics	-
	Self potential	-
	Resistivity	-
	Surface pea-gravel geochemistry	+
	Soil geochemistry at 0.5-1 m deep	+
	1-2 m deep	+
	Soil geochemistry profiles 0-5 m deep	<u>+</u>
	Radon gas	+
	Helium gas	<u>+</u>
	Biogeochemistry	+
	Hydrogeochemistry	+
	Stream sediment sampling	<u>+</u>

day. The major cost in an exploration project is in manpower to plant and recover them. The cost is warranted if front-end exploration expenses are lowered, for example, in hydrocarbon exploration, or if uranium deposits are detected that can not be found by other methods.

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Figure Captions

- Figure 1. Hypothetical section across an oil-bearing structure. The pattern of molecular movement of hydrocarbon gases from the oil mass towards the surface and the movement of subsurface water with its natural radioactive component are portrayed. There are concentrations of radioactivity and hydrocarbon gases at the surface position vertical projection peripheral to the subsurface petroleum accumulation (From Merritt, 1959).
- Figure 2. Conceptual diagram showing the distribution of uranium and ^{214}Bi (eU or equivalent uranium) downslope from an oxidizing uranium deposit. Note the displaced eU anomaly where groundwater bearing radioactivity issues as a spring, and the false anomaly for uranium in the bog where sufficiently reducing conditions have resulted in the precipitation of uranium (After Bradshaw and Lett, 1980).
- Figure 3. Results of a radiometric survey at the Coyote Creek and Madison Pole Hills Field, Bowman County, North Dakota. Oil is found at about 9800 feet in the Ordovician Red River dolomite. Note the halo-shaped pattern for the radiometric high region (modified from Weart and Heimberg, 1981).
- Figure 4. Variations in Hg emission in relation to changes in soil temperature, soil moisture, and water table level as measured over a one year period. Gas-coupled radioactivity signals may be subject to the same variations (compiled from Klusman and Webster, 1981).
- Figure 5. Diurnal variation in Hg emission in relation to changes in outside temperature. Gas-coupled radioactivity signals may be subject to the same variation (after Klusman and Webster, 1981).

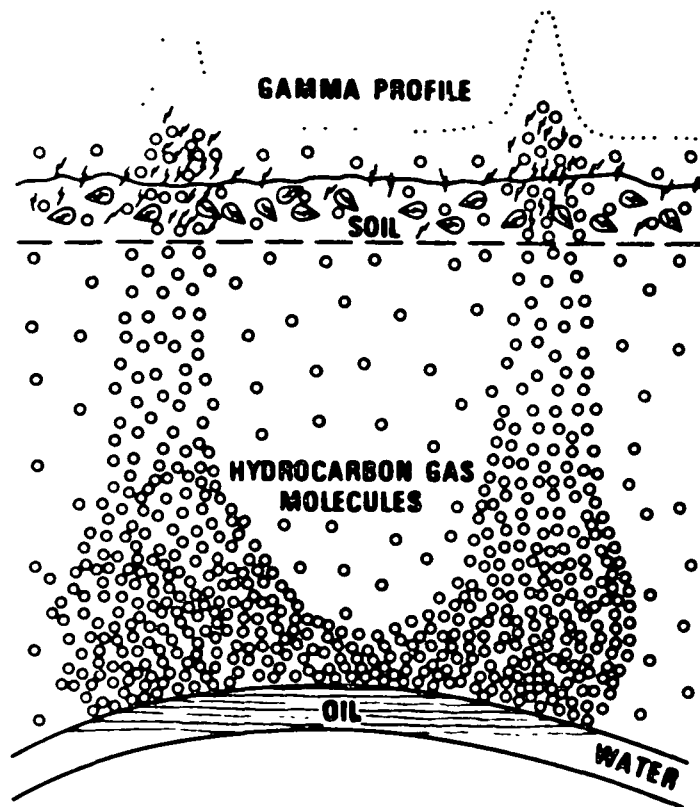


Figure 1

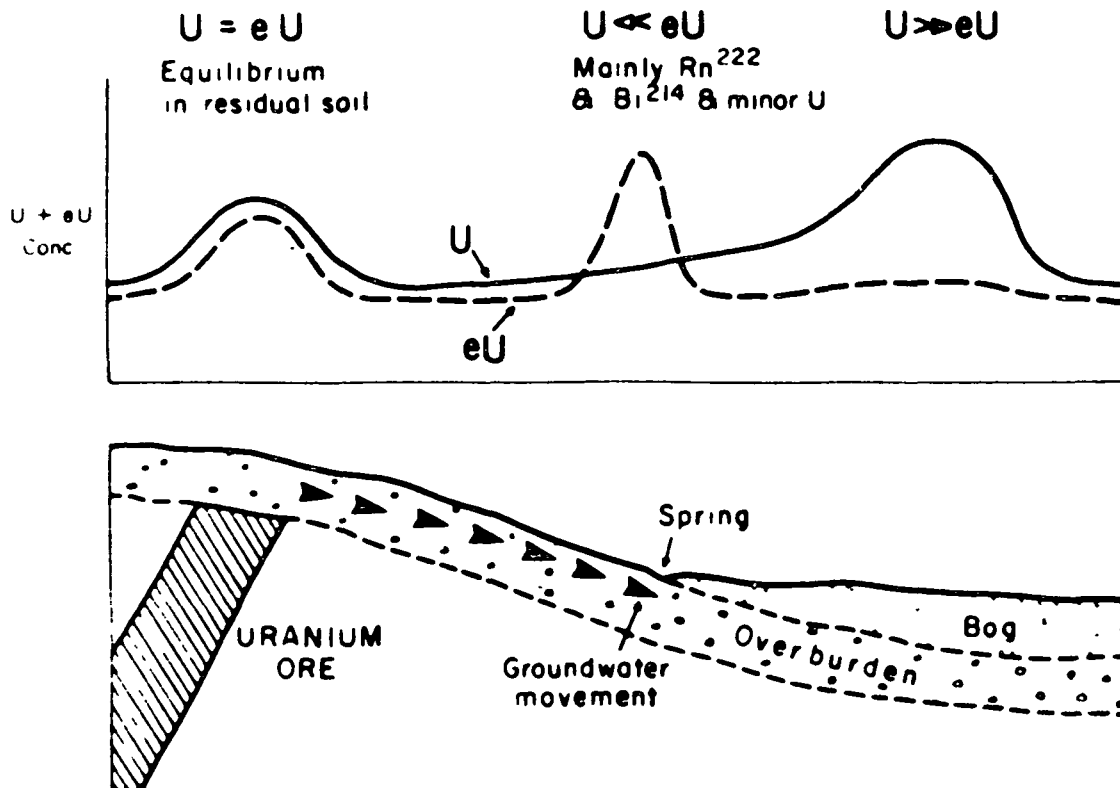


Figure 2

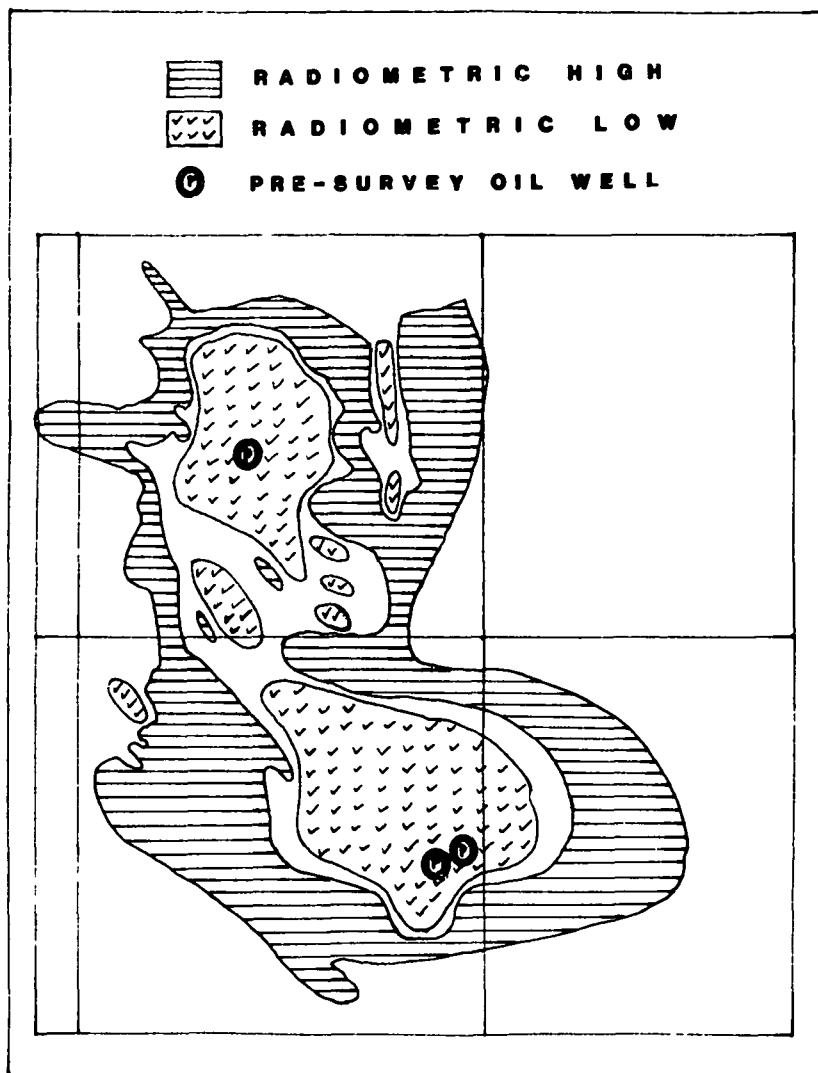


Figure 3

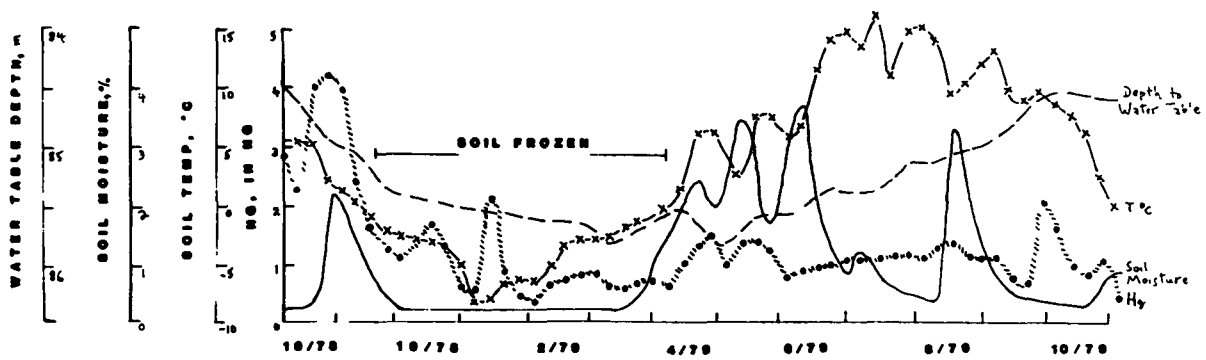


Figure 4

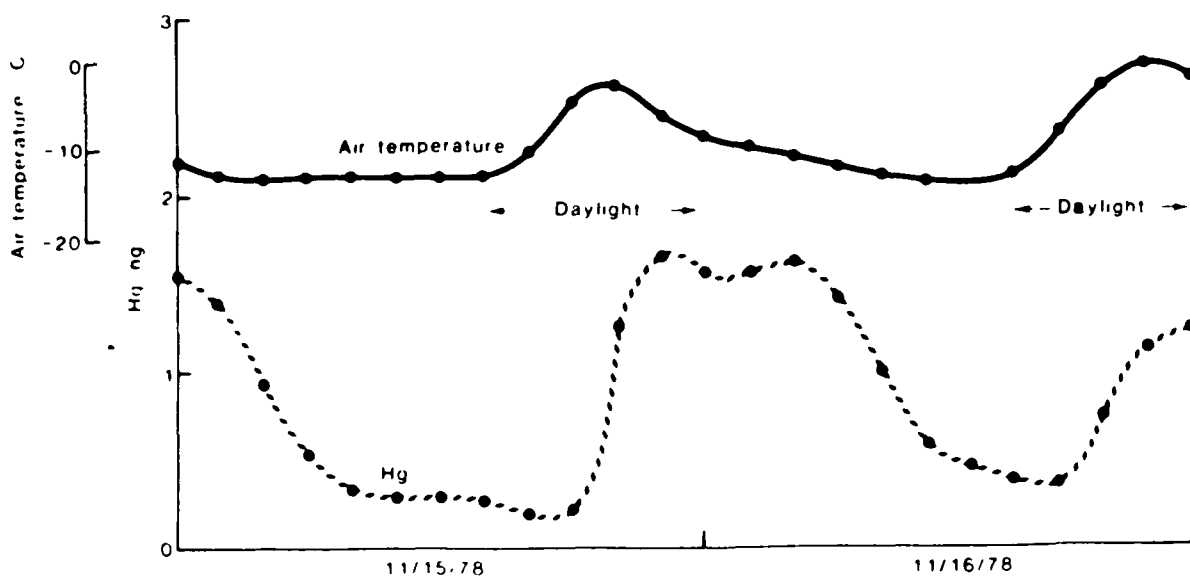


Figure 5