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# Radiation Halos and Hydrocarbon Reservoirs: A Review

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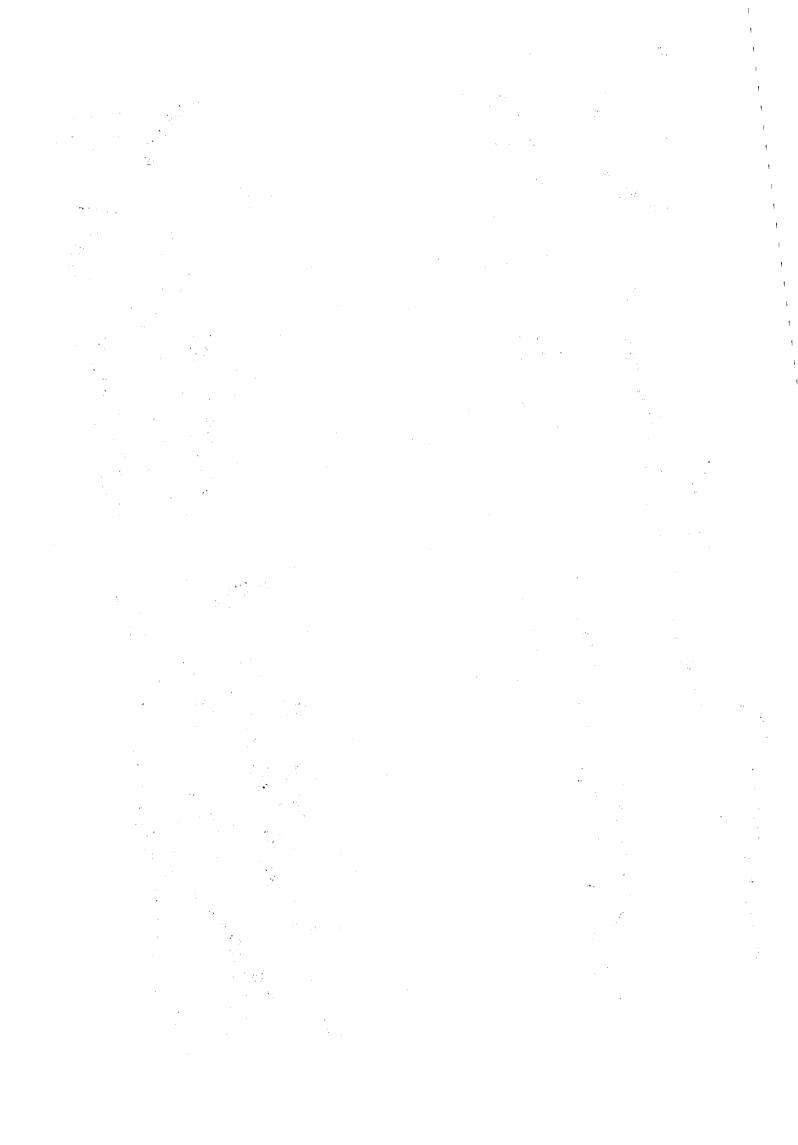
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#### RADIATION HALOS AND HYDROCARBON RESERVOIRS: A REVIEW

by

F. E. Armstrong 1 and R. J. Heemstra 2

#### ABSTRACT

This Bureau of Mines publication reviews 237 papers on radiometric prospecting, which is the measurement of naturally occurring radioactivity in the earth's surface and its application to petroleum and natural gas exploration. Many theories and documented surveys are listed in both support and refutation of this reported phenomenon, and the Bureau of Mines is conducting research to obtain direct field information in order to determine the statistical credibility of a radiometric method of predicting petroleum accumulation.

#### INTRODUCTION

This Bureau of Mines paper reviews the literature dealing with the possibility that a "radiation halo" surrounds oilfields and gasfields; proof of the existence of such a halo would obviously be a valuable step forward in petroleum exploration. Intimate knowledge of such a phenomenon would be a prerequisite to its general application as a prospecting tool for petroleum. Thus, an extensive search of the literature, complemented by personal communications, was made as a preparation for further endeavor in the field of radiometric prospecting.

Both the existence of such an anomalous radioactive background and the identification of the radionuclide species responsible for the radiation pattern relevant to the vicinity of a petroleum accumulation have been the subject of much speculation. Although the fact that 96 out of 113 observers report a gamma-radiation anomaly occurring in the vicinity of oil and gas deposits gives this reported phenomenon greater credibility  $(\underline{1-92})$ , it should also be noted that none of these same observers claim a significant lack of radiation anomalies over dry but otherwise promising petroleum structures. On the other hand, negative positions were expressed by 17 other investigators in varying degrees of confidence  $(\underline{10}, \underline{12-13}, \underline{19}, \underline{35}, \underline{39}, \underline{46}, \underline{71})$ .

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 $<sup>^{3}</sup>$ Underlined numbers in parentheses refer to items in the list of references at the end of this report.

Most of the results of gamma-radiation surveys relative to the search for hydrocarbons were published before the advent or wide usage of pulse height analysis of the total gamma energy spectrum. As a result, the more recent articles are directed to specific radionuclides and their detection with the use of sensitive scintillation crystals, multichannel pulse height analyzers, and elaborately corrected radiation profiles and isoradiation contours derived by means of automatic data processing.

The radiation halos discussed in this review are not to be confused with the radioactive disturbances found commonly in many mineral structures. Those pleochroic halos, such as are found in biotite, are caused by alpha-particles forming discoloration rings 10 to 40 microns in size (155). In fact, the radiation "halo" anomalies described are very often not the traditional ringshaped "halos" but are randomly arranged patterns. Some patterns are semicircular or even fan-shaped and are often several miles in magnitude.

In the early days of nuclear-radiation anomaly detection, the emphasis was on the development of radiation-counting equipment. Almost coincidental with the detection of radiation anomalies was the development of detection methods for geochemical anomalies in the vicinity of both ore bodies and petro-leum. Trace amounts of hydrocarbons, measured in surface soil samples, were among the first geochemical indicators correlated to deposits of gas and oil at great depth (140). The early methods of hydrocarbon detection were somewhat limited in sensitivity, however.

Today, just as with nuclear-radiation detection, advances have also been made in the detection and identification of trace amounts of gaseous and light hydrocarbons by gas-liquid chromatography. As a result of gas chromatography development, interest in methods utilizing these technological skills for the search for oil reserves has been increasing (124, 152). It seems only natural that theories explaining causative relationships between anomalous gammaradiation fields and hydrocarbon or any other geochemical anomalies would be prominent. Some of the general background is developed in this review to support the interrelationships between the various geochemical and geophysical phenomena.

#### Literature Survey

An intensive survey of the literature reveals that radiometry, which is the measurement of naturally occurring radioactivity in the earth's surface, has been actively studied and applied to petroleum and natural gas exploration since 1927. More than 50 different principal investigators and 115 authors, in at least 12 countries and five States in the United States, have contributed to the study. Many of the early workers combined their experimental efforts with an overt display of salesmanship in the cause of "nuclear application." This incentive contributed to a burgeoning list of publications on the subject until the early 1960's when a maximum of activity apparently was reached (fig. 1).

The marked drop in number of articles seen during 1963-64 probably reflects the high level of long-lived radioactive isotopes formed in the

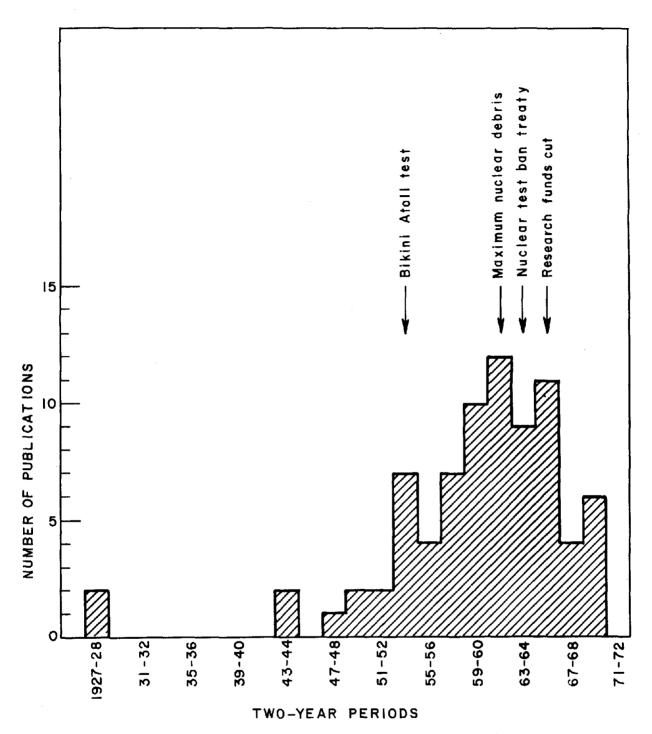


FIGURE 1. - Publication Frequency on Radiometric Oil Exploration.

atmosphere during the period of intense testing of nuclear weapons in 1961 and especially in 1962 ( $\underline{204}$ ). Then on August 3, 1963, the nuclear weapons test ban treaty was signed by the Soviet Union, the United States, and Great Britain. It permitted the radiation background level to decay sufficiently for further radiometric exploration work, as seen by the 1965-66 rise in

publication activity. A possible explanation for the sudden drop in research activity in 1968 is the 1966 cut in research funds nationally (231).

The bibliography includes a selected list of references to the literature of petroleum exploration by means of radiation surveys which were written between 1927 and 1971. Broad subject categories are arranged for convenience to the researcher.

Most references selected for this review were examined in detail, but some were not available for examination or were not translated, except for their abstracts.

# Early Research

The first investigator to attempt radiometric exploration for petroleum was Bogoyavlenskiy, who measured the gamma radiation of the Maikopsky oil region with an electrometer in the summer of 1926 (15) and that of the Ukhta oil-bearing region with a new 2,150-cm³ electrometer in 1927 (16). The Maikopsky field was an enormous tectonically undisturbed surface in a virgin forest region which was geologically well known. The Ukhta field was a great anticlinal fold. Bogoyavlenskiy observed sharp increases in intensity of penetrating radiation above oil lenses of light naphtha as well as above strata of heavy oil. He reasoned that oil absorbed large amounts of radium which gave off emanations through the earth.

Following Bogoyavlenskiy's reports, the first reports on the "radio-activity halo" phenomenon published before 1956 were by Stothart, 1943 (81); Sterrett, 1944 (79); Lang, 1950 (47); Lundberg, 1952 (54); Merritt, 1952 (62); Scherb, 1953 (73); Gott and Hill, 1953 (33); Pringle and coworkers, 1953 (70); Lobdell and Buckley, 1954 (52); Flerov and Alekseev, 1955 (27); and Alekseev and coworkers, 1955 (9). The next 16 years saw greater application of the more sophisticated instrumentation.

#### Reviews

Many review articles on radiometric prospecting have been written. Alekseev (5) outlined the state of the art in 1957. A review of radon migration by Tanner (84) includes applications of environmental radioactivity to petroleum as well as geophysical exploration. A review of radiometric surveys and of the many various theories on radiation "halo" anomalies around oilfields and gasfields is well presented by Sikka (76-77).

Broda and Schönfeld (18) included the radiometric method of prospecting in their well-documented list of radioactive applications. In the U.S.S.R., Polshkov (69) recognized radiometry as a possible structural exploration tool among the other well-established geophysical methods. A recent review by Foote (30) on radiometric techniques and a comprehensive article by Pirson and coworkers (68) on the geochemical concepts involved contribute much to an understanding and awareness of this unconventional method of exploration.

# Natural Radioactivity

The three principal sources of natural radioactivity in the earth are potassium-40 ( $K^{40}$ ), the uranium-radium series shown in figure 2, and the thorium-radium series shown in figure 3. The migration and fixation of various members of these series are responsible for natural radioactive anomalies as well as for general background radiation originating from the earth. The relative contribution of each species to the total intensity of radioactivity is influenced by its half-life, type of radioactivity, and energy of radiation.

The radiation energies, intensities, and half-lives are given in table 1 for the uranium series and in table 2 for the thorium series.

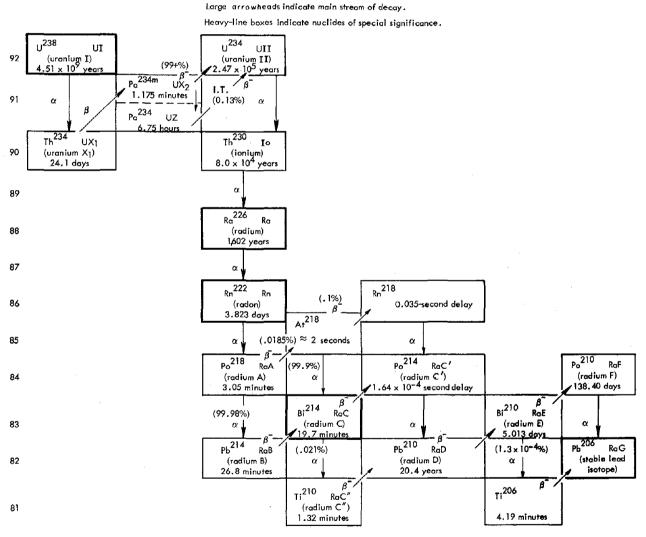


FIGURE 2. - Uranium-Radium (4n + 2) Series. Data compiled from Lederer (205).

TABLE 1. - Gamma radiation energies from the uranium (4n + 2) series occurring in natural background (205)

Name of nuclide	Classical symbol	Half-life	Energy, mev	Absolute intensity/decay, percent
Thorium-234	UX1	24.10 days	{0.064 .093	3.5 4
Radium-226	Ra	1,602 years	.186	4
Lead-214	RaB	26.8 min	.242 .295 .352	4 19 36
Bismuth-214	RaC	19.7 min	.609 .769 .935 1.120 1.238 1.378 1.40 1.728 1.764 2.204	47 5 3 17 6 5 4 3 17 5
Lead-210	RaD	20.4 years	.047	4

TABLE 2. - Gamma radiation energies from the thorium (4n) series and  $K^{40}$  occurring in natural background (205)

Name of nuclide	Classical symbol	Half-life	Energy, mev	Absolute intensity/decay, percent
Thorium-232	Th MsThl	1.41 x 10 <sup>10</sup> years 6.7 years	0.059 .010	<del>.</del>
Actinium-228	MsTh2	6.13 hr	.129 .270 .328 .338 .908 .960	4 3 4 11 25 } 20
Thorium-228	RdTh ThX	1.910 years 3.64 days	.084	1.6 3.7
Lead-212	ThB	10.64 hr	{ .239 .300	47 3.2
Bismuth-212	ThC	60.60 min	.727	7
Thallium-208	ThC''	3.10 min	277 .511 .583 .860 2.614	7 23 86 12 100
Potassium-40	K <sup>40</sup>	1.26 × 10 <sup>9</sup> years.	1.460	11

Large arrowheads indicate main stream of decay.

Heavy-line boxes indicate nuclides of special significance.

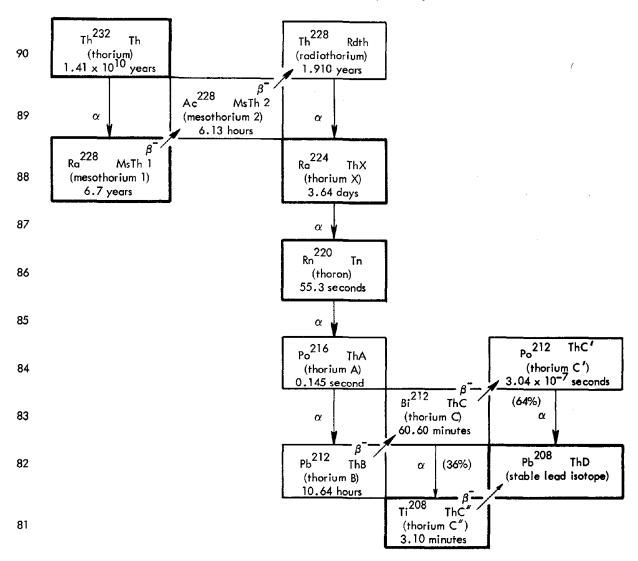


FIGURE 3. - Thorium-Radium (4n) Series. Data compiled from Lederer (205).

In the process of properly assessing the degree of atmospheric contamination by nuclear weapons and manmade nuclear waste products, radiation detection equipment was being developed with ever-increasing sensitivity and reliability. Studies of atmospheric radon radioactivity eventually led to the detection of these gamma-activity anomalies at specific surface locations. Meteorological factors modifying the anomalies contributed to the difficulty of locating the sources of emanation ( $\underline{134}$ ). Crews ( $\underline{20}$ ) recognized the influence of weather on the radiation profile. The presence of radon in the atmosphere was considered to be mainly influenced by the radium concentration in the surface soils of various classifications (164).

The airborne gamma-ray instrument system developed for radiometric surveying is of particular interest in that up to 75 percent of the radium decay gamma radiation measured may be caused by airborne radon decay ( $\underline{195}$ ). This radon decay activity must then be subtracted along with radioactivity from other background sources ( $\underline{179}$ ) to give the radiation response originating from the earth's surface only. Anomalies containing as little as 6 ppm thorium and 3 ppm uranium have been detected by use of airborne equipment ( $\underline{176}$ ).

#### ACKNOWLEDGMENTS

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#### GEOCHEMISTRY

# Geochemical Anomalies

A brief study of geochemistry should aid in discovering any similarity between certain geochemical anomalies and the observed radioactivity patterns. A geochemical anomaly is essentially a macroscopic nonrandom distribution of a particular sought element or geochemical species occurring on or in the earth's surface. Superjacent or lateral hydromorphic anomalies, or those lying immediately over the source, are termed epigenetic in nature because of the upward or horizontal movement of metal-bearing solutions and show the well-developed halo pattern. Lateral anomalies are formed by horizontal migration of ground water and are frequently influenced by local landforms. The mechanical dispersion of element materials, termed syngenetic anomalies, occurs in moraine deposits and often forms fan-shaped patterns. Hydrocarbon dispersion patterns originating superjacent and lateral to petroleum-bearing formations would be epigenetic in nature.

The migration of trace concentrations of heavy metals under favorable conditions of hydrolytic precipitation results in aqueous dispersion halos of these metals (146). This precipitation usually occurs along a natural geochemical barrier, such as an oxidation-reduction interface, or a pH barrier (113). Reducing conditions are normally formed in environments containing organic materials and are often accompanied by an increase in carbon dioxide and hydrogen sulfide contents and a negative Eh value.

Geochemical patterns can occur over concealed mineral deposits and are described as ranging in shape from halos to fans depending on lateral ground-water flow or upward movement of soil moisture (119). Anomalous subsurface occurrences of gold, silver, molybdenum, mercury, copper, lead, and zinc have been individually observed in drill hole cores taken from 100 to 1,200 feet below the surface to assess local mineral potentials (133). Sulfide ore

bodies are postulated to be natural galvanic cells which cause extensive local solution and transporting of metals and may be responsible for heavy-metal dispersion aureoles at the surface over ore bodies 100 to 150 meters deep ( $\underline{167}$ ). Even surface vegetation in areas of oxidized ores is markedly affected by the presence of uranium-vanadium deposits (102).

# Geochemical Prospecting

The chemical characteristics of subsurface waters in the vicinity of oil-fields and gasfields have long been sought in case they could be useful in petroleum exploration. Ammonium contents in ground water are said to increase 100 percent in the vicinity of oil and gas formations (158). Ground waters surrounding and underlying petroliferous regions of the Pre-Carpathian Downwarp and the Dneper-Donets Basin were found to have low silica contents relative to other ground waters (162). The silica content, however, did not correlate with total salinity, depth, or temperature of the aquifer, and it was felt that silica-poor ground waters near petroleum were also usually poor in sulfate ions but enriched in radium, ammonium, strontium, and volatile phenols. The trace element data of the Saratov-Volgograd region, U.S.S.R., show the concentrations of manganese, strontium, barium, and to a lesser extent lithium to be higher in ground water inside the oil-bearing contour (136). Strontium was three to five times more concentrated inside the immediate neighborhood of the oil deposit than outside.

Many geochemical anomalies have been observed, both on and beneath the surface of the earth, to have direct relationships to hydrocarbon deposits. The trace elements of manganese, vanadium, nickel, and copper as well as the radioactive elements uranium and radium were found by Alekseev, Gottikh, and Sundukova (7) to be adsorbed to a greater extent around the environs of the Kyurov-Dag oilfield in Azerbaijan than directly over the field. In this same basin, copper and manganese were shown to increase near oil and gas structures, probably as a result of a change in the local ground-water redox potential (117). Sikka (77) lists 25 oil and gas deposits found by chemical analysis of the soil. Preliminary results by Zak (174) failed to show any trace element anomaly resulting from migration of the trace elements of nickel, vanadium, zinc, chromium, copper, or uranium from the underlying oil reservoir to the surface in the soil samples over the Heletz oilfield in Israel. A hydrocarbon anomaly mentioned, however, probably contributed to the northward extension of the Heletz field, known as the Kokhav field.

A fluorometric method of analysis has been applied to petroleum exploration by examining sulfur compounds extracted from soil bitumens (132). In addition, a 35-percent success ratio was claimed during a 6-year period of field testing the surface-geochemical method involving heavy-metal detection. According to theory, heavy metals of low mobility, moved vertically by compaction fluids and deflected by the prospective hydrocarbon barrier, would form anomalous halos in mineralized chimneys surrounding the deposit.

Other explanations have been suggested for the association of the geochemical anomaly to a hydrocarbon deposit. Alekseev and coworkers proposed that overlying sediments were made oil-wet by hydrocarbon leakage from subsurface accumulations, thus reversing their adsorptive role for ions of heavy elements (4, 7). Shneyerson and Skosyreva (163) were able to characterize this water repellency of rock and soil quantitatively by a flocculation method. An interesting fact is that hexane molecules are relatively insensitive to the chemical nature of the surface and are held primarily by van der Waals forces (236). This places the greater importance on colloidal particle size in determining the surface properties of the rock materials and thus the presence or absence of trace elements.

Figure 4 illustrates the probable mechanism that accounts for the hydrocarbon and radioactive "halo" found encompassing an area above a petroleum reservoir. The phenomenon is explained by postulating globules of hydrocarbon carried with the mineralized water in the sand lens flowing around the oil barrier and up, creating a geochemical aureole. This basic concept has been modified by many and refuted by some, but the preponderance of evidence in the literature encourages further study.

Naturally occurring organic acids form stable colloids between pH 6 and pH 9, which cause chemical weathering of the host rock ( $\underline{148}$ ). The metal organic complexes could cause transport of minerals through the aqueous environment of a subsurface structure depending on Eh, pH, and concentration (120).

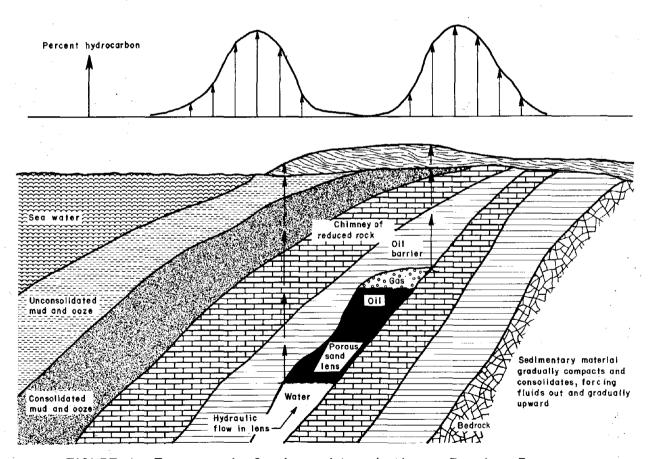


FIGURE 4. - Formation of a Geochemical Aureole Above a Petroleum Reservoir.

Unsaturated hydrocarbons found in trace quantities around petroleum reservoirs are suggested as being capable of forming complexes with heavy elements, including uranium, and moving upwards from reservoir edges by compaction to the surface (111), whereas methane, occurring in higher concentrations over the geologic apex of the deposit, is considered incapable of forming any complex (64). This places considerable importance on hydrocarbon weight distribution as an influence on a geochemical anomaly, if true.

# Hydrocarbon Anomalies and Prospecting

Light hydrocarbon gases, mainly methane, apparently diffuse vertically from high-pressure structural reservoirs through the semipermeable sedimentary column to the surface. Tripp (171) calculated that 376 cubic feet of methane diffuses to the surface per square yard per year from a Fort Collins anticline at 4,500-foot depth. This amounts to nearly 4,000 moles of methane per acre per year. This vertical migration of hydrocarbons, both as dissolved in ground water and as gases slowly leaking from oil and gas accumulations, is recognized by many investigators (103, 122, 124, 140, 143, 152, 169) as the mechanism giving rise to hydrocarbon anomalies observed lying over various hydrocarbon-bearing formations. Horvitz (123) has outlined some of the history of geochemical prospecting through the use of hydrocarbon-detection methods. Through his persistent investigation of a special case, Horvitz illustrated the temporary nature of a hydrocarbon anomaly. The hydrocarbon halo surrounding the Hastings oilfield, Texas (discovered in 1934), which had been measured in 1946, was again measured in 1968, after the field had undergone considerable depletion. The 1968 measurements indicated that the hydrocarbons originally present in the soil above the field had disappeared in in large sections of the area measured and that the main anomaly had left the soil. Quite possibly, the rate of hydrocarbon emanation is proportional to the bottom-hole pressure in the oil and gas reservoir, down to the prevailing hydrostatic pressure, after which emanation can be expected to cease altogether.

Most of the investigators surveyed caution against simple interpretation of the hydrocarbon measurements. Corrections for soil types, vegetation, bacteria, and other organic artifacts make interpretation a complex problem. Soil samples for hydrocarbon analysis should be taken at uniform depths exceeding that of vegetation growth in the survey area. Horvitz (125) thoroughly investigated the role of vegetation in hydrocarbon prospecting. Even carbon-12 to carbon-13 ratios in methane from samples taken from drill holes were used by Slack (165) to locate petroleum accumulations, on the basis of diffusion fractionation taking place in the lateral direction.

According to Mirchink and coworkers (147), the Soviet Union was operating 500 gas-logging (gasometry) units by 1965. This method was said to have contributed to proving oilfields and gasfields in the Near Volga-Urals area; however, no mention of radiometry was included in that encompassing report.

# Radioactive Anomalies

When a geochemical anomaly includes one or more radioactive elements as an integral part, either adsorbed on soil or in ground water solution, the anomaly becomes a radiometric one, and its sensitivity for detection is greatly increased. Small amounts of uranium in the soil can greatly influence radio-activity measurements made on the surface. Weak radioactivity anomalies have been caused by thin mudstone beds that contained only 0.002 percent  $\rm U_3O_8$  (100). Another anomaly of 10 times natural background resulted from an outcrop containing 0.028 percent  $\rm U_3O_8$ .

Experimental evidence shows that migrating ground water tends to acquire and differentiate uranium from the daughter products when passing through uranium ore bodies (98). Other investigations have shown that colloidal uranium is absent from the ground water in the vicinity of a uranium deposit but that the contained uranium occurs 95 to 98 percent in anion form (159). The uranium content in the ground water from a supergene uranium deposit correlates to total salinity in a nonlinear fashion. The solubility of uranium is shown to be also a function of the CO<sub>2</sub> content (104), but the  $Ra^{286}/U^{238}$  solubility ratio can vary widely, especially under the carbonate influence (170).

Generally speaking, radiation leakage anomalies are common above most buried uranium ore bodies. The position of this leakage halo or any mineral-ization of the soil, with respect to the buried ore deposit, depends on the local structural tectonic conditions.

#### SIGNIFICANCE OF THE URANIUM-HYDROCARBON ASSOCIATION

The history of radioactivity measurements associated with produced oil dates back to Bogoyavlenskiy (183), who pointed out the inverse relationship of ash activity to ash content in the oil. The inverse relationship shown to exist between the uranium and ash contents of petroleum demonstrated that the uranium is present among the organic components of petroleum (93). The presence of uranium is also shown to be genetically related to the content of organic matter in uranium-rich shales, and uranium enrichment is enhanced by a low rate of sedimentation which allows more organic material to accumulate (6, 160). Alekseev and coworkers (94) also found a strong correlation between the uranium content and the organic carbon content in clay.

Hydrocarbon deposits, especially those which are highly oxidizing in nature, tend to accumulate heavy metals (99, 106, 109, 141). Fossil bones that contained large amounts of organic matter accumulated unusually high concentrations of uranium from percolating ground waters (131). The high uranium concentration in peat is not derived from plants growing in uranium-bearing soils; instead, the humus tends to absorb uranium by direct contact with water (142). A lignite field in Japan contained up to 0.074 percent  $\rm U_3O_8$  (166).

The uranium contents of 24 crude oils from midcontinent, Rocky Mountain, and West Coast fields in the United States range from 0.00004 to 0.013 ppm (99). A clear relationship between the uranium content and the specific gravity of 23 oxidizing petroleums from the Fergana depression in the U.S.S.R. was established (141). As the petroleum becomes more oxidized or asphaltic, its capacity to extract uranium from ground-water solution increases.

Organic deposits accumulate certain metals in preference to others. Vanadium and nickel are associated generally with all heavy crude oils (107), and in a vesicular basalt from Rozelle Point, Box Elder County, Utah, containing 12.5 percent sulfur, the trace metal content includes a high amount of titanium. This linear increase of metal content with asphaltene concentration is also often true for vanadium and nickel (107-108).

Oil extracts from asphalt-bearing rocks contained 0.17 to 21 ppm uranium  $(\underline{118})$ , and those from petroliferous rocks contained 0.2 to 67 ppm uranium  $(\underline{109})$ . The uranium concentration in asphalt, however, ranged from 0.015 to 32,410 ppm (3.24 percent).

Deposits of petroleum and uranium tend to associate together in equivalent sedimentary terrains because both require transmissive host rocks and deposition traps of a similar nature (138). One major difference, however, is the fluvial origin of the uranium deposit as compared with the marine origin for petroleum. Uranium roll ore deposits are described as occurring along oxidation-reducing interfaces in sandstone introduced there in a "billowing manner" by oxygen-bearing ground water (112).

Reducing conditions were usually caused by the local presence of hydrocarbons, hydrogen sulfide, or unstable sulfur compounds. Both Eh and pH were considered important factors in the transport of uranium by ground water movement (113-114). Under these reducing conditions, uranium is believed to migrate from water into the oil, which explains a very high gamma activity measured at the contact zones in wells of the Mancharov and Arlan fields of Bashkirea, U.S.S.R. (135).

The reduction of uranium from solution to insoluble form occurs on the surface layer of the insoluble solid organic phase (96). While uranium (VI) is reduced in valency to uranium (IV), the organic matter involved--peat, wood, cellulose, oil shale kerogen, etc.--is oxidized and new reactive groups are formed to complex with more uranium. Although many uranium-organic associations are considered unstable, the complexes of uranium with ethylene, propylene, butylene, and other organics are thought to be very stable (111).

Breger and Deul outline the importance of the role of the organo-uranium association in the geochemical cycle of uranium (101). Although petroleums from nonuraniferous areas may contain very little uranium, petroleum still has the ability to transfer uranium from its original site to another (109-110). Analysis of the organic matrix of uranium ore points to a humic acid origin, and the presence of metals which hydrolyze easily in solution, such as thorium, titanium, and zirconium, indicates that these uranium-bearing materials were carried in a colloidal suspension by ground water to the deposition site (126, 129).

Since precipitation occurs along the border of the reducing zone or at the contact zone between petroleum and water, uranium and other elements, such as nickel, cobalt, molybdenum, silicon, iron, vanadium, lead, copper, and zinc, concentrate themselves in the surface-active fractions, which tend to adhere to the pore walls of the reservoir rock (105, 114, 151). Other metals

(titanium, vanadium, nickel, and aluminum) are said to be precipitated to the bottom of organic layers, especially in bog deposits ( $\underline{119}$ ). Epigenetic uranium mineralization can occur in either a productive or a vacated petroleum reservoir ( $\underline{114}$ ).

#### SIGNIFICANCE OF RADIOACTIVE GROUND WATER

Petroleum extracts uranium from ground water according to the chemical nature of the water in contact, but because of repeated redistribution between petroleum and water, some petroleums become more uranium enriched or impoverished than others. The determination of the uranium contents in 56 petroleum samples from six Azerbaijanian deposits in the Soviet Union supports this view (93). The Azov-Kuban, Emba, Turkenenia, and Ural-Volga region oilfields were found to contain formation waters rich in radium, but with insignificant amounts of uranium (111). The reason given for this was that the uranium forms stable organometallic complexes in association with the petroleum, but that the daughter product, radium, which does not form complexes with the organic substances retained by the petroleum, migrates into the formation waters, leaving much of the parent uranium behind. This would explain an earlier puzzle in which uranium-bearing brines did not correlate with brines depositing radium-bearing precipitates (33).

The question must still be answered whether radium in ground water increases because of the presence of hydrocarbons in the water or because the presence of both hydrocarbon and radium in the water has a common origin. Data for the radium content in ground waters from deep exploratory wells of the inner zone of the East Carpathian Downways of the Soviet Union and for their salinity were listed by Shchepak (161) in groups of both those waters not related to hydrocarbon deposits and those associated with hydrocarbon deposits. Correlation coefficients of 0.922 for waters not related and of 0.637 for petroleum-related waters can be computed from his data. Hydrocarbons apparently displace the influence of salinity on the radium content in ground water and, at the same time, relate to an increase in the overall radium content. Gutsalo (40) observed a direct linear relationship of the radium concentration to the partial pressure of hydrocarbons dissolved in water.

A total of 113 analyses by Gutsalo (116) also confirms a linear relation—ship between the radium content and the total salinity in ground water from the Paleozoic and Mesozoic Formations of the Dneper-Donets Basin. The radium content increases with the age of the rocks and also with proximity to oil and gas accumulations. Similar relationships are found in the Pre-Carpathian Downwarp.

These positive radiohydrogeological anomalies near oil deposits were considered by Shchepak also to be important prospecting clues in the outer zone of the East Carpathian Downwarp ( $\underline{161}$ ). The higher ratios of radium content to salinity for a particular formation relate very well to waters from natural gas and petroleum deposits but not to waters above or below these deposits. Mal'skaya ( $\underline{145}$ ) disagrees with Shchepak, except for certain gas deposits.

The increase in radium content of the ground-water horizon near the oilwater contact is credited to the high degree of metamorphism associated with reducing environment of deep, stagnant waters in isolated horizons  $(\underline{6})$ . Although some surface waters show an increase in radium concentration over the productive crests of oil pools, with the reverse being true for the uranium contents  $(\underline{95})$ , generally speaking, radium is not observed in the surface water within the oil and gas region  $(\underline{6})$ .

Emanation measurements of radium activity anomalies, which are often misconstrued as radon anomalies, can inaccurately reflect geologic features under thick overburden because of displacement by ground-water movement unless the features are very large (168). The radiation aureoles observed on the surface cannot have been produced solely by ground water carrying dissolved radium salts up from the vicinity of the accumulated petroleum and associated brines. Rothe (72) calculated that the surviving radium in ground water moving up at a rapid pace of about 1 inch per year from a depth of nearly 800 feet would be reduced to 2 percent of the original. Uranium, on the other hand, would survive such a trip.

Uranium and thorium can also be transported from their original sites under the influence of increased salinity, temperature, or pressure by fluids such as brines and petroleum  $(\underline{110})$ .

# The Question of Radon Diffusion

The general consensus is that greater than normal amounts of radon, helium, argon, radium, uranium, and thorium tend to occur in petroleum and natural gas deposits. The highest amounts of radon in helium-bearing natural gases from the Panhandle field, Texas, are found in reservoirs which also contain uraniferous asphaltite, radium-bearing brines, and gamma-ray anomalies in the rocks (151). The helium present in natural gases is believed to be derived from uranium and its daughter products. The radon level is easily lowered, however, by absorption into the petroleum which still may be present in the pore spaces of the gas reservoir.

Comparatively high radon concentrations were found in ground waters that flow through acidic rock formations, and low concentrations were found in basic rocks (137).

# Anomalous Radon

Several inert gases, both radioactive and nonradioactive, are produced by the disintegration of uranium, thorium, and potassium-40 isotopes. Being chemically inert and physically immobilized in the formations below the water table, these gases dissolve into the ground water. As the ground water reaches the water table, the gases leave the water, go into a vapor phase, and enter the pores of the rock or soil. A natural breathing action of the soil causes the inert gases to diffuse into the air at the surface, where the presence of their radioactive components is detected as a diurnal variation of the radiation background.

According to Hawkes and Webb  $(\underline{119})$ , this breathing action is caused by changes in the barometric pressure at the surface of the earth. Schroeder and coworkers  $(\underline{157})$  discuss radon pumping action at the earth's surface as being associated with continual microoscillations observed in the barometric pressure or in thermal instabilities such as in desert atmospheres. Other meteorological conditions, rain, for example, are said by Jacobi and Andre  $(\underline{128})$  to have a much greater influence on radon leakage to the atmosphere.

The occurrence of radon-222 in 307 natural gas wells was measured by Bunce and Sattler (186) to be in the range of 2.7 to 66.9 picocuries per liter. Upon disintegration, radon gas transmutes on through radium A ( $Po^{218}$ ) and radium B ( $Pb^{214}$ ) to the strongest gamma emitter, radium C ( $Bi^{214}$ ), as shown in figure 2.

# Radon Over Faults

A leakage pattern for radon will often occur along a fault area or over a more permeable zone. Because of its low diffusion coefficient, radon does not reach the earth's surface from below at very great distance. Budde ( $\underline{185}$ ) calculated the distance from which a buried point source can be detected from measurements of anomalous radon concentrations. This distance ranges from 1 meter in sand to 5 centimeters in boulder clay. His investigations also show that radon diffusion in unconsolidated overburden appears to be controlled mainly by grain size and water content. Thus, he considers any uranium deposit to be totally hidden below a 4-meter depth if detected from radon diffusion alone. Dyck ( $\underline{192}$ ) sets the detection of buried uranium by radon emanation at 20- to 30-foot depths.

Lang (47) advocates the radioactivity survey method for defining the presence of faults. Kondratenko (202) assumed that radon emanation anomalies, with B-activity anomalies lacking, were determined by tectonic disturbances. This assumption was later confirmed by mining operations. Subsurface faults located as deep as 5,000 feet below unfaulted formations were observed from gamma radiation increases gained with mobile ionization chambers of 350 liters at 30 atmospheres' inert gas pressure  $(\underline{228})$ . Test drillings later confirmed their existence.

Traditionally, faults have been normally considered barriers to, rather than conduits for, the migration of fluids or gases (237). Vogler (173) rejected the radon diffusion theory over fault zones and explained the activity increase as evaporation of uranium-bearing water moved upward by capillary osmosis. This principle of capillary osmosis of ground water could explain radiation anomalies over structures which accumulate petroleum. Oil deposits do not act as sources of radon diffusion reaching the surface, according to Marton and Stegena (209). The penetration of emanometric measurements seldom exceeds 5- to 10-meter depths.

#### RADIOMETRIC PROSPECTING

# Exploration Methods

Many investigators have produced results which, in their opinion, definitely show how to outline or find the presence of oil productive areas by means of the direct radiometric method of prospecting. Table 3 shows a partial list of some of the fields studied. Lang (47) reported that the Ceres pool, a shoestring sand without local structure in Noble County, Okla., was drilled on the basis of radioactivity measurements. He even felt that the productivity of an area was related to the value of a radioactivity count.

TABLE 3. - Partial list of fields having radiometric anomalies

Location and field	Principal author	Reference
Romania	Bisir	14
U.S.S.R., Ukhta region	Bogoyaklenskiy	16
Egypt, El Alamein	El Shazly	24
U.S.S.R., Volgograd district	Glagoleva	31
United States, southeastern Kansas	Gott	33
United States, California, Ten Section	Kellogg	43
Israel, Rift Valley	Mazor	60
U.S.S.R., Bukhara-Khiva Depression	Semenov	74
United States, California, Ten Section	Sikka	75
Colombia, La Cira	Trapp	86
U.S.S.R., western Fore-Caucasus	Us	87
U.S.S.R., west Turkmenia, Kum-Dag and Kizyl-Kum	Yermakov	89
U.S.S.R., Binagady-Khurdalan anticline	Zolotovitskaya	90-92
U.S.S.R., Kyurov-Dag	Alekseev	7
U.S.S.R., Lower Volga and Ciscaucasus	Afonin	2
U.S.S.R., Rybaki	Kopia	45
Canada, Álberta, Redwater	Pringle	70
Do	Sikka	76
Romania, Carpathian Outer-Depression	Gohn	32
U.S.S.R., Bashkiria, Mancharov and Arlan	Komarov	135
U.S.S.R., Turkmenia, Dzu-Dzu-Klinskou	Simon	78
United States, Texas, Darst Creek; Oklahoma, Seminole County.	Stothart	81
United States, Oklahoma, Ceres	Lang	47
United States, Texas, Van	Scherb	73

The gamma-ray contour map of Scherb (73), showing Nicar's survey of the Van field in Van Zandt County, Tex., indicates to him that a radiation low, which has drifted to the west of the center of production, could be successfully used to determine the productivity of an area.

Langford (48) claims that old oilfields show a more pronounced "halo" effect than do new fields. His reasoning is that wells drilled into old fields allow the release of radioactive molecules from the crest of the pool, causing a low-radiation zone.

A decrease in radioactivity derived from uranium was demonstrated by Flerov  $(\underline{26})$  over the Shkapovo oilfield. This uranium anomaly was thought to reflect a general geochemical pattern over the deposit, although thorium activity did not show this same low. A 60- to 70-percent chance of success for petroleum discovery was claimed after careful consideration of lithologic factors.

Negative gamma radiation anomalies were measured and mapped above oiland gas-bearing formations by many investigators. Activity profiles recorded with carborne equipment included those of the Binagady-Khurdalan anticline in Azerbayan by Zolotovitskaya (90-92); the Kum-Dag and Kizyl-Kum oilfields in western Turkmenia, U.S.S.R., by Yermakov and Shatsov (89); and the Cambay and Ankleshwar areas in India by Aithal (3).

The gamma field intensity detected by Us and Kripnevich ( $\underline{87}$ ) outside oil deposits in the western Fore-Caucasus measured 60 to 80 percent greater than that inside the deposit area and 40 to 60 percent greater than the average background.

Positive activity increases were shown by Stothart  $(\underline{80}-\underline{83})$  in radon measurements over the petroleum-producing fields of Darst Creek, in East Texas fields, in Fort Bend, Fisher, and Scurry Counties, Tex., and in Seminole County, Okla.

The limits of productivity for crude oil sources existing at depths to several thousand feet were determined with a claimed accuracy of 70 percent by measuring emanation crests from collected radon (80). From seven positive radioactive anomalies, four new fields were opened (82). Stations established at 100-foot intervals are monitored for 15 minutes by portable equipment carried by jeep (83).

The quantitative results from radon emanations, which defined existing shoestring formations in Kansas, indicated to Sterrett (79) the degree of thinning of the productive sand.

Lower contents of uranium were expected by Sikka  $(\underline{76})$  in lime-rich sandy soils (pH 7.5 to 8.5) and in acidic soils (pH 2.5 to 5.0), whereas maximum uranium was removed from solution by organic matter at pH 6.0 to 6.6. Gregory  $(\underline{35})$  related radioactive highs to some of the prominent alkaline areas, probably owing to precipitated radium sulfate.

In his table of correlations of radiation anomalies to oilfields by various investigators, Sikka (76) suggests that the failures outnumber the successes, but does not mention any ratio. He lists 28 investigators who claim low anomalies and 14 who claim high anomalies over oilfields. In one anticlinal field, the Ten Section, both a low and a high are observed. The difference between Kellogg's (43) high and Sikka's (77) low over the Ten Section is entirely due to soil corrections in the latter (figs. 5-8).

Aithal (3) has observed that the highs around an anomalous gamma field are mainly from radium activity and the lows in the center of the field are

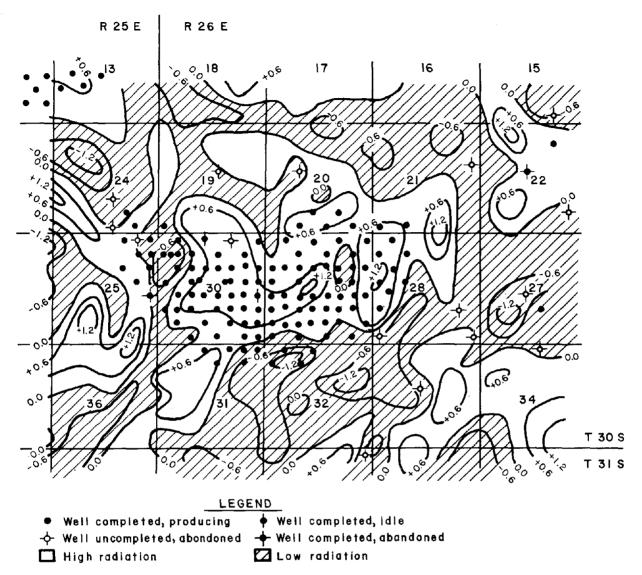


FIGURE 5. - Aerial Radioactivity Survey, Ten Section Oilfield, Kern County, Calif. Contours show standard deviations from mean. (After Kellogg, ref. 43.)

mostly from potassium-40. Also, normal background levels were found to be different in different areas. On the other hand, Alekseev and Gottikh  $(\underline{6})$  show that changes in the surface gamma field are mainly caused by changes in potassium content.

Soil samples were taken by Baranov and coworkers ( $\underline{11}$ ) from 25-centimeter depths over the Gekcha oil-bearing structure in western Turkmenia which exhibited a pronounced gamma anomaly. Results showed equal increases in concentration over the anomaly of the elements uranium, radium, thorium, and potassium. In this same oil-bearing structure, Karasev ( $\underline{42}$ ) found the uranium to be tightly bound to the soil and in isotopic equilibrium with both radium and other members of the uranium family, specifically  $\mathbf{U}^{\otimes 34}$ .

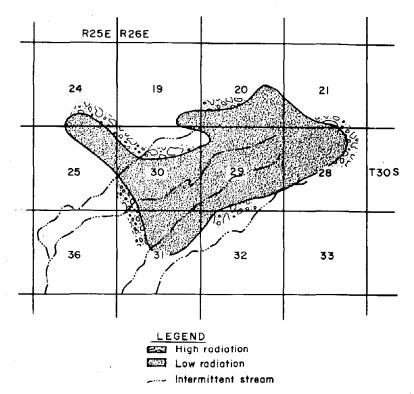


FIGURE 6. - Radiometric Survey of an Oilfield, Showing Anomalous Low. (After Sikka, ref. 77.)

This again indicates that the activity "halo" could not have been caused principally by radium precipitation around the oil pool periphery. Lundberg (54) supports this view and feels that the radium comes from uranium-bearing formations close to the surface. His calculations show that only 1 percent of the radium would remain after ascending 800 feet at 1 inch per year.

Sikka (76) discussed the different individual anomaly shapes over a reef, an anticline, and a fault. A reef, he maintained, shows scattered highs bordering a low over the pool; an anticline is the same but the scattered highs are farther from the edge of the pool, and the fault shows highs along its length.

Thomeer (85) observed similarity between hydrocarbon anomaly traces and radioactivity anomaly patterns. Miller (63) suggested that anomalous radiation patterns agreed with the general locations of hydrocarbon increases. Sikka (75) also found a radiometric anomaly to coincide with a hydrocarbon anomaly over the Ten Section oilfield, Kern County, Calif. Even the radiation anomaly pattern from alpha counts on soil samples taken from drill holes in Alberta are reported by Lundberg (54) to resemble those of the chloride and methane contents.

Many countries, other than the United States and the Soviet Union, have participated in petroleum exploration programs involving radiometry. Bisir  $(\underline{14})$  found that oil-productive areas in Romania were reflected through low-radioactivity anomalies from the uranium-radium family of nuclides. The direct correlation of gamma fields to nine Romanian oil deposits was summarized by Gohn and Bratasanu  $(\underline{32})$ . Four gamma profiles across the La Cira oil-field in Colombia were correlated by Trapp and Victoria  $(\underline{86})$ . In 1959, all Chinese organizations which could secure radiation-detection equipment were asked by Liu and coworkers  $(\underline{51})$  to make immediately a radiation survey of some area for petroleum exploration. A general gamma radiation low over the El Alamein oilfield in Egypt was described by El Shazly and coworkers  $(\underline{24})$ . Also, the Redwater oilfield in Alberta, Canada, was studied by Pringle and coworkers  $(\underline{70})$  and by Sikka  $(\underline{76})$ .

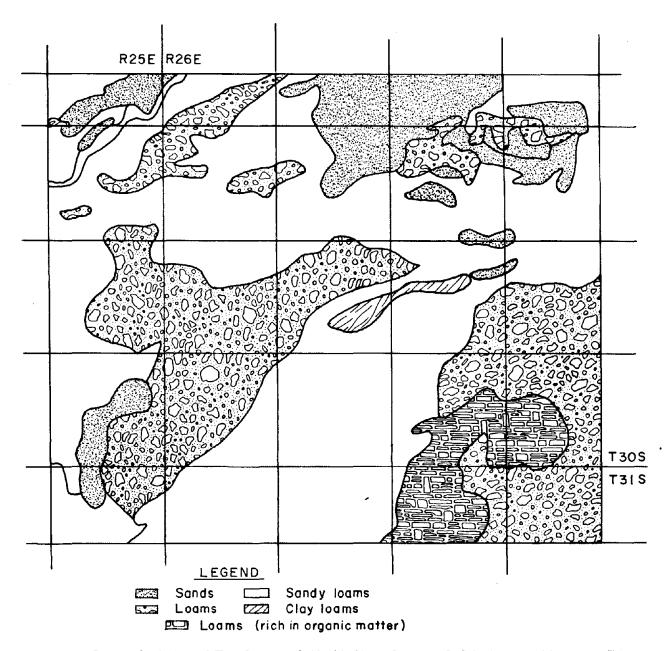


FIGURE 7. - Soil Map of Ten Section Oilfield, Kern County, Calif. (After Sikka, ref. 77.)

# Negative Observations

Although Cook  $(\underline{19})$  did place radiometric exploration into his category of scientific methods, all results obtained before 1954 at Southwest Research Institute were either negative or inconclusive.

Bêress  $(\underline{13})$  could correlate no gamma-ray anomaly with the Biharnagy-bajon oil structure in Hungary. Soft gamma radiation shows an inconclusive relation to reservoir structure. Vegetation was an important factor in his measurements.

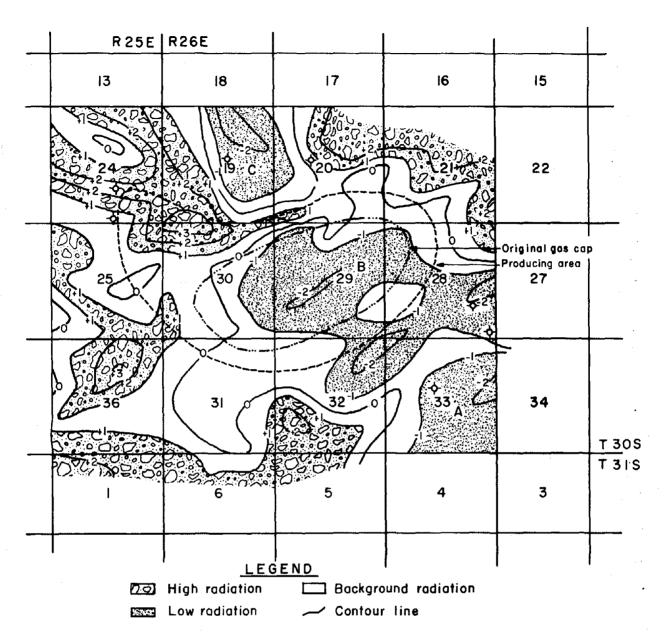


FIGURE 8. - Radiometric Survey of Ten Section Oilfield, Kern County, Calif. Contours show standard deviations from mean. (After Sikka, ref. 77.)

Gregory (35) maintains that the radiation lows are only coincidental with the petroleum reservoir outline; nevertheless, he associates the high activities with radium-bearing waters draining from petroliferous formations and accumulating upslope to alkaline areas.

Although many workers  $(\underline{56}, \underline{59}, \underline{63}, \underline{79}, \underline{82} - \underline{83})$  advocate the use of radiometry directly in outlining the field, it is not considered by Haddad  $(\underline{41})$  the answer to a direct surface geophysical method of petroleum exploration.

Radioactivity and geochemical anomalies are considered by Asadov and Shoykhet  $(\underline{10})$  to be related more to sediment grain size than to hydrocarbon migration.

Several authorities  $(\underline{12}, \underline{71})$  publishing in 1965 seem to show no awareness or recognition of radiometric prospecting as a working method.

# Theoretical Concepts

According to the theories of several investigators (57-58, 62, 65), the radioactive anomalies form because of the direct presence of hydrocarbons in the soil or rock. A linear system of fracture patterns beginning near the flanks of a structure, progressing upwards, and emerging to the surface away from the structure is theorized by Sikka (76). This feature would provide easy outlets to rising hydrocarbons, ground water, and radioactive salts.

Halo patterns from hydrocarbons, salt concentrations, and radioactivity can be explained by fluid expulsion and capillary barriers resulting from rock load  $(\underline{85})$ . These patterns are so distorted, however, that they might be useful only as supplementary information to other methods.

Merritt  $(\underline{62})$  states that an excess of gas movement or escape to the surface, leading to an excess of water evaporation, results in an accumulation of water-soluble radioactive substances which varies according to the different soil types and water saturation. From thermodynamic considerations, Tripp  $(\underline{171})$  has calculated that 81 percent of the total water evaporated at the surface comes from the upper 25 feet.

Foote  $(\underline{30})$  places the explanation for the existence of a radiation halo directly on the presence of hydrocarbons in the soil and lower formations in the vicinity of deposits of oil and gas. Sikka  $(\underline{77})$  mentions a temperature difference, proposed by Merritt in 1957, observed between petroleum wells drilled on the edge of the field and those drilled near the center, to explain the formation of the hydrocarbon halo anomaly and subsequently possibly cause the radiometric anomaly.

Alekseev and Gottikh (6), on the other hand, believe that the gamma field is related to the "progressively developing structure" which may or may not have accumulated a petroleum deposit over the course of its formation. Their studies show that the gamma field is not directly related to the presence of oil or gas and demonstrate this same "halo" to be present around the very promising, but nevertheless dry, Shchelkovo structure. Thus, Alekseev has painted a very convincing picture of the origin and explanation of the so-called "oilfield halo." This explanation is not convincing enough, however, for Pirson and coworkers (68), who question the unproductiveness of the dry structure or its past history of petroleum accumulations. They also question the actual grain-size variation over the studied structures, which is the basis for Alekseev's explanation of the radiation anomaly.

If a radiation anomaly is dependent on the presence of a hydrocarbon anomaly, then the limiting conditions that apply to the hydrocarbon anomaly

(that is, horizontal drift, soil gas interference, etc.) would also apply to the radiometric anomaly. The field observations of Kroepelin (139) pertaining to hydrocarbon patterns, therefore, must have bearing on radioactive pattern according to the theories of radiation origin from hydrocarbons.

Gregory (35) maintains that no correlation exists between radioactivity anomalies and deeply buried oil reservoirs. Instead, a direct correlation of the radioactivity distribution to the surface soil classification is pointed out. However, in early attempts to reconcile radiation data to geochemical data, today's sophisticated radiation counting techniques were not available.

Pirson and coworkers (68) view radioactive anomalies as originating from mineralization phenomena that occur during vertical formation water leakage from consolidation and compaction of sediments as the petroleum is being formed in geologic traps. In other words, a funnel of mineralization containing radioactive elements is formed by the vertical escape of compactionexpelled waters during the consolidation process while oil is being formed. These expelled waters carrying suspended hydrocarbons migrate upward, bypassing the oil pool traps, and form a "chimney" of reduced rocks extending to the earth's surface around the oil environment. This chemically reduced zone was identified recently by Pirson (153) as the cause for certain telluric currents which can be used to help locate oil. Pirson feels that the radioactivity lows form during the actual formation of the oil pools, and that they indicate the presence of primary oil accumulations at depth but are not expected to be associated with secondary oil accumulations where the oil has migrated from the original location. Also the radioactive halo does not disappear after the oil has left the region. From Pirson's reasoning, the radiation anomaly should take a large amount of time to form during the geological history of the oil pool.

#### RADIATION MEASUREMENTS

#### Detection Methods for Exploration

The alpha-particle electroscope was one of the earliest means of measuring radioactivity in petroleum  $(\underline{183})$  and measuring radon emanations over oilfields  $(\underline{81})$ . Alpha-radiation surveying of surface soil samples was even judged by MacElvain  $(\underline{57-58})$  to be far superior to gamma surveying. A much clearer halo pattern should be produced by the presence of stable lead-206 and lead-208. These daughter products from radon and thoron, he thought, may have been concentrated on the surface by ascending hydrocarbons.

The measurement of beta radiation on the earth's surface was included in the radiometric method of petroleum prospecting by Grumbkov and Marin-Fedorov  $(\underline{36})$ .

Bisir  $(\underline{14})$  found results by emanometric methods more conclusive than those by gamma radiometric measurements.

Early preliminary gamma-radiation surveys by Lang (47) in known anticlined producing fields indicated increased total gamma activity at the crest

of the structures. Sensitive Geiger-Mueller detectors were placed in stations 1/4 mile apart, and their activity was recorded for 6-month periods. Walking and carrying his counting equipment, Rothe (72) measured negative anomalies from background gamma radiation taken at 65-foot intervals across the old Fallstein oilfield and the Diesdorf-hadde-Kath field in East Germany. A high-sensitivity, 10,000-cubic-inch ionization chamber, 18 inches in diameter by 10 feet in length, was preferred by Miller  $(\underline{63})$  to scintillators for gamma detection because of sensitivity and slow but steady drift.

Drill holes have been used to monitor gamma-radiation profiles across oilfields and gasfields (45, 80), and gamma-radiation well-log records were used by Omes and coworkers (65) and Pirson and coworkers (67-68) to outline radioactive oilfield anomalies.

Where roads are available, many workers used the automobile to transport equipment (223). Distances covered included 10 miles per day (88), 40 miles per day (63), 15 to 20 kilometers per hour (89), and 15 kilometers per hour (91).

A scintillation counter was used for geological fieldwork for the first time in Canada by Pringle in 1949 (219). Both gamma-ray spectra and isoradiation plots of anomalous gamma fields were involved in this early work.

Whereas the use of an extremely sensitive electroscope by Stothart  $(\underline{81})$  illustrates the state of the art of detection in the 1940's, the scintillation counters employed by Trapp and Victoria  $(\underline{86})$  are typical of the 1960's.

# Airborne Radiometry

Lundberg (54) was one of the first oil seekers to use the airborne scintillation detector. He recorded radioactivity on flight lines over uranium deposits and oilfields in Texas and Canada in 1952. The airborne method involving scintillation equipment was applied to petroleum prospecting by many investigators (1, 4-5, 11, 24, 30-31, 38, 43, 50, 56, 59, 76, 78), and all reported successes.

Many other nonpetroleum-oriented, airborne radiation surveys were also conducted (176, 178, 180, 184, 188-190, 193-195, 198, 200, 207, 210, 217, 220).

MacFadyen and Guedes (207) studied the effect of height, spacing, and "atmospheric and soil humidity" in airborne radiometry on radioactive mineral deposits in Brazil. They found that 100- to 150-meter altitudes were optimum. Resolution decreased significantly above 200 meters. After a rain, 3 days were required before a similar radiation profile could be discerned.

The U.S. Atomic Energy Commission has developed equipment called ARMS (Aerial Radiological Measuring Survey) which will produce 360 counts per second from a 500-foot altitude over an  $\rm I^{131}$  source concentration of 1 microcurie per square meter (39, 217). Six 4-inch-diameter by 2-inch NaI(T1) crystals with photomultipliers were used, giving a natural gamma-ray background ranging from 100 to 1,000 counts per second. With this equipment, a geological correlation to activity was found to be similar to that calculated from use of subsurface gamma-ray logs.

Boyle  $(\underline{184})$  noted that gamma-ray penetration of rock is less than 1 foot and felt that airborne prospecting for uranium was not a substitute for prospecting on the ground, but rather an adjunct. However, flying altitudes of 100 to 200 feet still allowed coverage of 100 to 200 feet on each side of the flight path using a 3-1/2-inch NaI crystal.

The radiation measurements taken from the air must be reduced to a common level, usually the earth's surface (210). One of the tools for reducing the surface radiation to a lithologically normal value is the gamma spectrum of  $K^{40}$  (187).

Radioactivity mapping with a single gamma spectral line from radium, referred to as radium metallometry (203), is considered a less ambiguous indication of hydrocarbons in the ground than the total gamma-ray activity (230). Many others have concentrated their measurements also to specific nuclides in the gamma spectrum with the use of pulse height analyzers (194-195, 222). The quantitative separation of the gamma-ray spectra is accomplished by using certain suggested parameters. These parameters are the radium decay, as represented by the 1.12- and 1.76-mev levels of RaC; the thorium decay, as represented by the 2.614-mev level of ThC" and the 0.908- and 0.966-mev levels of MsTh2; and the potassium-40 presence, as represented by the 1.46-mev level.

#### Combined Methods

The coordination of radiometry with other geophysical methods may offer a promising application to petroleum exploration. In a general review of the geophysical achievements in Soviet petroleum geology, Mirchink and coworkers (147) state that the best results in the direct prospection for petroleum came from seismic and electric surveys combined with gravimetry. Lyon and Lee (206) advocate low-level aeromagnetic measurements simultaneous with potassium-40 gamma-ray measurements to give a cross-relationship in data. Jenny makes use of dual-level aeromagnetic measurements to detect microanomalies (233).

A combination of radiometry and gravimetry was used, as reported by Klestov and Orlov  $(\underline{44})$ , to discover the U.S.S.R. Razepinsk oilfield. Baranov  $(\underline{180})$  mentions work being conducted for applying combined aerogamma and aeromagnetic recordings to petroleum exploration. Airborne magnetometry coordinated with airborne radiation detection has been used by MacFadyen and Guedes to establish the presence of the whole geological structure associated with a radiation anomaly from a uranium ore body (207).

Still, Rankin (71) feels that magnetometry, especially with the use of the fluxgate sensing element, is the leading airborne geophysical method and that scintillation counter radiometry, as a geophysical method, is no longer showing significant application.

#### DATA PROCESSING: LITHOLOGICAL NORMALIZATION

Most investigators recommend that soil samples be taken and analyzed as a basis for radiation corrections. According to Merritt  $(\underline{61})$ , soil interference patterns present an almost insurmountable problem to gamma-ray surveying, and

all radiation values must be adjusted to a uniform soil basis before mapping. For instance, low activity values would be found over marshy ground, sandy soil, and thin soil (less than 12 inches thick) above bedrock; high activity values would be expected over granite washes, unweathered shales, and heavy argillaceous soils; and exceptionally high activities should occur over dry lake basins. The 50-percent increase in activity of the average clay soil over sandy soil might tend to obscure the 10- to 25-percent anomalous effect found by Alekseev (4) over certain petroleum fields.

Statistics on soil pattern variations and soil type differentiation were given by Purvis and Buckmeier ( $\underline{220}$ ) for  $K^{40}$ , using ground and airborne measurements. Bunker and Bush ( $\underline{187}$ ) established a practical detection limit for potassium in Pierre shale of about 0.10 percent, based on the obscuring of the  $K^{40}$  energy peak by other natural radioisotope peaks as the limiting factor. An average potassium concentration of 2.0 percent was found in all Pierre shale samples. The average thorium-to-uranium ratio in Mancos shale, according to Pliler and Adams ( $\underline{154}$ ), is 3.1 with an average concentration of 10.2 ppm thorium, 3.7 ppm uranium, and 1.0 percent potassium as the metal. A linear relationship of potassium to thorium was found, with the potassium-to-uranium ratio nearly constant at 2,200.

A geologic model of the earth's crust, including sedimentary and marine domains, was developed with the aid of a computer and abundance data for 65 elements ( $\underline{121}$ ). Computer-based data files for the earth sciences are available in some vicinities, which include information on sampling points, wells, latitude-longitude coordinates, geologic, hydrologic, and other earth-science factors ( $\underline{216}$ ,  $\underline{232}$ ). The parameter codes used allow data exchange with other data banks, such as in the petroleum industry.

An airborne remote-sensing device based on an induced pulse transient was used by Barringer and McNeill (181-182) to locate gravels and detect layering, fault zones, ground-water channeling, or other conductive anomalies in the earth's surface. The system's output is a complex function of the electrical conductivity of the surface overburden penetrating down many feet. A combined radiation and conductivity survey could be applied possibly to the normalization of radioactivity for soil classification.

Correction factors for radioactivity found in various soils of the Redwater oilfield, Alberta, are listed in table 4 by Sikka (75-76). With clay-loam beginning at a soil correction factor of 1.00, the other soils are corrected upwards with factors of loam, 1.07; sandy loam, 1.22; wooded sands, 1.28; and sands, 1.43.

Pirson  $(\underline{66})$ , using gamma well-log data, found the background activity of shale decreased by 50 percent when lying directly over oilfields.

Soil type	Parent material	Radioactivity,	Soil-correction factor
		incur.	140001
Clay loam	Lacustrian	220	1.00
Loam	Bedrock	215	1.02
Do	Alluvial lacustrian and till	211	1.04
Sandy loam	Silt till	217	1.01
Do	Aeolian	201	1.09
Meadow	dodo	-	1.09
Sand	dodo	195	1.13
	Aeolian-organic	191	1.15

TABLE 4. - Some soil factors of 1957 Ellingwood data after Sikka (75)

The problems in interpretation can be listed as follows:

- 1. Establishing a background value to the anomaly requires extending the range of sampling and considering the possible overlapping of anomaly sources.
- 2. Measuring the statistical significance of an anomaly must be realized, as must recognition of nonsignificant anomalies.
- 3. A quantitative appraisal of the anomaly must be made in terms of the productive-to-dry hole probabilities and in terms of the anomaly-over-background magnitudes, including the size and shape, and many geologic parameters such as faults and folds.
- 4. Finding the lateral or superjacent relationship of the hydrocarbon deposit to the measured anomaly is necessary because laterally displaced anomalies often show close correlation to surface geology.

# CONCLUSIONS

Much of the difficulty in evaluating the actual reliability of the radiation survey method stems from the relative lack of access to the total survey data. Many records of oil and gas companies are unavailable to the public for analysis because of the competitive nature of the petroleum business. In addition, Crews (20) has expressed concern over the "promotional" aspects of the radiation survey method that are occasioned by the ready availability of inexpensive, portable, gamma-ray detection equipment.

Kroepelin (139) reviews some of the success-to-failure ratios of several geochemical prospectors. The history of geochemistry, as outlined since 1942, and its high overall success rate should have made it an integral exploration tool today, but this is not the case. There seems to be a genuine suspicion of geochemical methods which require comprehensive statistical processing of large quantities of data. This would be even more true in radiometric prospecting.

The search for documented oil discoveries stemming from previously surveyed radioactivity anomalies is a very tedious one. One application of the

hypothesis concerning the projected presence of oil and gas from the radio-activity of produced underground waters may be found in the Rift Valley area of Israel as proposed by Mazor (60). Filonov (25) also used this hypothesis as an indirect approach to estimating an oil-bearing potential. However, many petroleum prospectors have apparently retreated into a noninnovative attitude by what might be called a "divining rod syndrome" (fear of just another divining rod method), and thus may fail to examine many new exploration techniques which are not immediately understood and of proven capability.

Two of the problems have been that application of the method has always preceded final development and that information concerning negative results tends to be discarded.

The future of petroleum exploration in the United States does not seem to include the field of gamma-radiation detection in any list of the new tools in prospect, nor has the field been given any role by the United States in the past (234). In 1965, Polshkov (69) reviewed many of the latest developments of geophysical and geochemical prospecting for oil and gas in the Soviet Union, and radiometry was among them. The natural gamma field was admitted to be greater over oilfields.

A comprehensive analysis of the whole system of variables is extremely complicated and most likely would completely drain any reservoir of patience and money designated to the purpose of uncovering a practical working method, <u>unless</u> the diagnostic approach of systems analysis were vigorously applied.

While the high cost of exploratory drilling makes a systematic and direct approach to the determination of the specificity of radiation halos to the presence of hydrocarbon reservoirs an economically impossible one, the secretive nature of oil exploration imposes great difficulties in attempting a systematic radiation survey of potential reservoirs before drilling commences. Nevertheless, the existence of radiation anomalies over earth surfaces including hydrocarbon reservoirs is an observed fact, although the specific and unique nature of such halos is open to question.

On the strength of the investigations studied to date, the Bureau of Mines is endeavoring to obtain direct information through original fieldwork in order to determine the statistical credibility of a radiometric method for predicting the subsurface presence of oil or gas accumulations. The validity of the assertion that the existence of a radiation halo at the earth's surface is indicative of the presence of hydrocarbons below can be proved practically only by determination of the mechanism of its formation. Without question, this mechanism concerns the general phenomenon of ion migration in the earth's crust. To reduce such a vast and complex system to known and predictable parameters is a difficult undertaking, but the potential value of such knowledge extends beyond that of the possible development of a direct method of petroleum exploration into other areas of mining as well.

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