CHARACTERIZATION OF NATURAL GAS, OIL AND COAL DEPOSITS WITH REGARD TO DISTRIBUTION DEPTH, AND TRACE ELEMENTS CONTENT

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CHARACTERIZATION OF NATURAL GAS, OIL AND COAL DEPOSITS WITH REGARD TO DISTRIBUTION DEPTH, AND TRACE ELEMENTS CONTENT

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Nearly all geologists and geochemists agree that natural gas and hydrocarbon fuels have been derived from organic matter. Increasingly, however, more and more geologists and geochemists are questioning the biological origin of hydrocarbon fuels. Evidence given in support of non-biological origin is as follows:

1. Methane rises from hydrothermal vents on the East Pacific Rise and from the Nyirogong lava lake in East Africa.

2. When hot carbon comes in contact with $H_2O$ at high pressure and low oxygen concentration, methane, CO and $CO_2$ are produced.

3. Gas trapped in diamond crystals found deep in the crust includes methane, $CO$, $CO_2$, ethane, propane and butane.

4. One geologist estimates that 100,000 tons of abiogenic methane is being released into the oceans every year.

5. Another group of geologists give an estimate of $1.4 \times 10^{12}$ barrels of abiogenic oil being produced every million years for the past 700 million years.

6. The distribution of hydrocarbon fuels with regard to depth within the crust appears to correlate with coal being near the surface, oil in deeper wells and natural gas in deepest reservoirs.

7. The oldest oilfield is more than three times older than the oldest coal fields (1500 million to approximately 400 million years ago).

8. Some geologists correlate on a worldwide basis petroleum accumulations with regions that experienced diastrophism. This demonstrates a direct relationship between plate tectonics and petroleum accumulation.
Evidence given in support of a biological origin of hydrocarbons is as follows:

1. Nearly all accumulations of oil and coal are found in sedimentary rocks rich in carbon derived from the remains of living matter.

2. Petroleum contains "optically active" compounds, i.e., those which rotate a beam of light. Carbon compounds formed biologically are optically active whereas abiological compounds are not.

3. The presence of vanadyl and nickel porphyrins in certain oil is another strong point in favor of biological origin.

Giardini and Helton, two University of Georgia geologists, argue that all evidence in support of biological origin is inadequate and insist that the non-biological origin of oil is preferred.

In conclusion, it appears that neither proponents nor opponents of the biological origin of hydrocarbon fuel have adequate hard data to unequivocally establish beyond a shadow of a doubt a firm foundation for either case. The momentum, however, appears to be shifting toward more and more evidence for a non-biological origin.

With regard to radioisotope dating of coal, it appears that using uranium-thorium to lead decay is possible. One needs quantitative analysis of the relative amounts of $^{U_{238}}, U_{235}, thorium_{232}, Pb_{206}, Pu_{207}$ and $Pb_{208}$ to determine the age of the coal.
I. INTRODUCTION

Nearly all geologists and geochemists agree that natural gas and hydrocarbon fuels have been derived from organic matter. In the case of coal, some scientists of Argonne National Laboratory claim to have discovered how nature made coal from lignin, the woody tissue in plant cell walls, and other organic matter such as lipids (fatty and waxlike substances from plants or algae). The general consensus regarding the biogenic origin of hydrocarbon fuels is not universally shared; however, some geologists and scientists, very few in number, are beginning to question the biogenic origin. Porfir'ev published a paper in 1974 on the Inorganic Origin of Petroleum, in the Bulletin of the American Association of Petroleum Geologists. Gold and Gold and Soter have hypothesized that during the accretion of the earth material, 4.5 billion years ago, methane and other hydrocarbons were trapped and have been migrating from the depth to the surface ever since. Giardini and Melton published several articles based on the analysis of gas trapped in diamonds obtained from different geographical locations on the earth. They not only question the biogenic origin of hydrocarbon fuels, they argue for large accumulations of non-biogenic petroleum.

Interest in abiogenic methane has increased significantly after Welham and Craig observed that methane was rising from hydrothermal vents on the East Pacific Rise. Also, Gerlack observed that volcanic gases from Nyiragongo lava lake near Lake Kivu in east Africa was saturated with methane. Both sets of observations suggest that methane can be produced through magnetic processes. David Osborne in an article in the Atlantic Monthly reports that Craig and colleagues have found methane streaming out of every ocean rift
and out of several underwater volcanoes. Craig estimates that a 100,000 tons of abiogenic methane is released into the oceans every year. The ratio of CO$_2$ to CH$_4$ streaming out of ocean rifts is about 100 to 1 according to Craig.

MacDonald (1) recently reviewed arguments for the biological origin of oil. He concluded that nearly all accumulations of oil are found in sedimentary rocks rich in carbon derived from remains of living matter. Also many oils contain "optically-active" compounds, i.e., those which rotate a beam of polarized light. Carbon compounds formed biologically are optically active whereas abiologically compounds are not. The highest quantities of optically active compounds are found in the youngest oils and the least in the older oils. The presence of vanadyl and nickel porphyrins in certain oil is another strong point in favor of biological origin since porphyrins have been identified in pigments of both plants and animals. Finally, according to MacDonald (1) the predominance in oils of compounds with an odd number of carbon atoms is strong evidence of biological origin because the biochemical synthesis favors such an arrangement.

On the other hand, Giardini and Helton (8) offer explanations that none of the arguments cited by MacDonald (and others) are valid "proof" of the biological origin of oils. For example, they state that the sedimentary rocks can easily contain traces of biologically recycled carbon compounds and that these rocks are permeable formations which act as migration channels for petroleum. Petroleum reservoirs are formed of sediments that have been deposited since the appearance of life on earth. Giardini and Melton assert that petroleum in general is composed predominantly of compounds which are not optically active. The presence of traces of optically active matter is not proof of a biological origin of petroleum.
With regard to the presence of nickel and vanadium porphyrins in petroleum, Giardini and Melton attribute this to the sedimentary environment which contained biological remnants; this should not be interpreted as proof of biological origin.

Other technical arguments with regard to the ratio of $^{12}C$ to $^{13}C$ are equally refuted by Giardini and Melton.

It appears that not enough data exist to totally refute the biogenic or abiogenic origins of hydrocarbon fuels. We know for a fact that in the case of methane some of it comes from non-biological origin. Does it all come from non-biological origins? We do not know. And what about oil and coal? Giardini and Melton found in diamond hydrocarbon chains up to $C_4H_{10}$ which are attributed to a non-biological origin. This fact points to the possibility that petroleum also could come from polymerization of smaller chains of hydrocarbon. Again, we do not know how much if any oil comes from non-biological origins but the possibility exists. With regard to coal the picture is murkier. We have not been able to uncover any relationship between coal and oil or coal and gas. Two facts however point to the possibility that even coal could have a non-biological origin:

1. As a function of depth, it appears that coal is on top, petroleum is in the middle and natural gas is at the bottom.

2. The distinction between some thick oils and asphalt decreases as the volatiles in oil escape. There might be a similar relationship between asphalt and coal.

Now, if polymerization played a part in the creation of oil from natural gas then it is reasonable to speculate that where geologic formations allow, in so far as permeability, temperature, pressure and local physical and chemical conditions, the volatile components of the petroleum to escape, the resulting
residuals would be tending toward the thicker oils, asphalt, coal, and maybe even graphite. The fact that coal beds tend to be closest to the surface where it is relatively easier for volatiles to escape is one reason why the possibility for a connection between methane and coal through polymerization and other chemical reactions cannot be excluded.

The general acceptance of a biological origin, which has been firmly entrenched for a long time, is slowly giving way to more scrutiny and increasingly we see more and more people question the biological origin.

With regard to where or how hydrocarbons came into being, the opinions differ widely. For example, T. Gold (11) believes that all of it is premontial in origin, trapped within the earth from the beginning of time and is being released from the depth to the surface ever since. Mr. Gold further believes that natural gas is the result of heavier oils breaking up under the effects of high temperature into simpler structures principally methane and residuals like coal or pure carbon. This hypothesis is opposite of that which puts methane as the building block and the polymerization and other chemical reactions produces both oil and coal.

A.A. Scarborough (12) has published his work on the origin of the solar system and has hypothesized that hydrocarbon fuels are being made continuously by a nuclear furnace in the core of the earth. Mr. Scarborough does not offer any terrestrial evidence supporting his hypothesis.

MacDonald alluded to the fact that natural gas can be produced by magmatic processes and gave evidence that when hot carbon comes in contact with $H_2O$ under high pressure, methane, CO and $CO_2$ are produced. By analogy, when hot steam is brought into contact with graphite or coal, in the absence of oxygen, methane and oil are produced. This is called "synfuel."
Giardini and Melton (8) proposed a model in which continuous outgassing of
H₂, CO, CO₂, alcohol, and low order hydrocarbons, up to butane are
entrained over appropriate catalysts to form a variety of hydrocarbons. The
flow channels for these products from the mantle to crustal traps follow faults
traceable to lithosphere fracture. Petroleum bearing regions are found in
association with land masses that experienced rifting, orogeny by subduction or
collision. Adjacent regions for sediment generation and accumulation are
produced by the resulting differential elevation of surface. Tectonically
active regions undergo diastrophism, which produces reservoir structures in the
sediments. Tectonically produced faults and fissures tend to generate deep
seated fractures that penetrate the lithosphere and enable flow of the mantle
fluids to the sedimentary reservoirs.

Giardini and Helton (8) estimate that the upper limit for petroleum
production per million years for the past 700 million years is 2.1 x 10¹¹
 tons or 1.4 x 10¹² barrels/million years. Of this amount only 0.2% is
retained in reservoirs.

Another fundamental question with regard to migration of carbon from the
mantle to the surface is whether or not elemental carbon exists in the magma.
We know that diamond exists in diamond mines, therefore elemental carbon must
also exist. Under elevated temperatures and high pressure, carbon can strip
the hydrogen from water molecules to make natural gas. Under this scenario one
is able to explain better the fact that natural gas fields tend to be deeper
than those of oil and coal.

In summary, our survey of the literature shows that increasingly more and
more geologists and geochemists are questioning the biological origin of
hydrocarbon fuels. The non-biological origin is also being increasingly
promoted to explain natural phenomena which could not be explained otherwise.
In this report, the characterization of natural gas and oil wells and coal beds is reviewed with emphasis on Pennsylvania's coal, petroleum, and natural gas. We also review possible methods for radiometric dating of coal and petroleum.

II. NATURAL GAS AND OIL WELLS AND COAL BEDS CHARACTERIZATION

Coal, petroleum, and natural gas are widely distributed on the face of the earth. Because of the economic importance of these fuels, there is a vast amount of detailed information about their occurrences available in the open literature. Additionally, a great deal of proprietary information is held by companies involved in exploration for and extraction of the fuels, and many foreign governments have data about their fuel deposits that is not freely available.

Because of the great interest in the world's fuel resources, there are many articles and books that provide summaries of the available information. For example, the Encyclopaedia Brittanica (15th Edition, 1983) has authoritative articles entitled "Coals" and "Petroleum," which provide excellent, brief introductions to the character of these materials and their occurrences in the earth's crust. Examples of recent books that provide more thorough summaries of the character and distribution of fuels in the earth are those by Ward (13) and Tiratsoo (14). Both coal and petroleum fields usually have natural gas associated with them, but there are many other occurrences of natural gas. MacDonald (1) has written a thorough review of the origins of natural gas.

Because of the vast amount of information available about the occurrence of fuels in the earth's crust, it is obviously not possible for this report to be complete as a compilation of such data. Nor would it be appropriate for the authors to attempt a broad summary of the available information, for there are
already available summaries of this sort written by experts who have devoted their careers to evaluation of fuel resources. The sponsor of the research that this report represents is interested in the possibility that some of the fuels within the earth may not be of biological origin. For this reason, the focus of this report will be on certain aspects of the occurrence of natural fuels that relate particularly to the possibility of non-biological origin of fuels.

We will look at the geographical distribution of the fuels at several different levels, beginning with a brief summary of the worldwide patterns of their occurrences, which should convey several important ideas:

- Fuels in the earth's crust are very broadly distributed in the continental portions of the earth's crust (including the submerged continental margins) but coal and petroleum have not been found in the oceanic crust.
- Although broadly distributed in the continental crust, the fuels are distributed in ways that are far from uniform.
- Although some regions have both coal and petroleum, these two kinds of fuels are not generally associated with one another.

Along with the summary of the worldwide distribution of fuels, it is quite important to consider the geological aspects of the occurrence of the fuels. It is, of course, the geological associations of these fuels with particular kinds of rocks, formed within restricted ranges of time, that appear to govern the geographical distribution of the fuels. Important generalizations are:

- Fuels have been found almost exclusively in association with sedimentary rocks.
Coal has not been found in rocks older than 400 million years and oil has not been found in rocks older than 1500 million years. Most major occurrences of these fuels are in rocks formed within the last 350 million years of earth history.

We will also give a brief summary of the geographical distribution of the fuels in the 48 conterminous states of the U.S., since the geography of these states is likely to be much more familiar to those who will read the report than is world geography. But in this case, as for the world as a whole, the information is far too vast for a detailed accounting. Consequently, we will focus on one state, Pennsylvania, which has major occurrences of coal, petroleum, and natural gas.

II.1 Worldwide Distribution of Coal, Oil, and Natural Gas: A Summary

Coal, petroleum and natural gas are very broadly distributed in the continental part of the earth's crust, which includes the continents, the submerged continental margins (continental shelves), and all of the world's large islands except Iceland and Hawaii. These two islands are part of the earth's oceanic crust, which comprises all of the deep ocean floor as well as most of the small oceanic islands.

Although oceanic crust underlies more than half of the earth's surface, it is not known to contain any coal deposits nor any economically useful accumulations of petroleum. However, the high pressure and low temperature of deep ocean water permit methane hydrates to be stable in deep ocean sediments, and there is growing evidence that large quantities of methane may be present in such sediments [MacDonald(1)]. Methane is also known to be present in volcanic gases and hot waters associated with the volcanic activity that is continually producing new oceanic crust. The concentrations of methane associated with volcanic activity are low and do not appear to be of any economic importance, however.
Our knowledge of the character of the oceanic crust has vastly increased in the past four decades because of intense study of the sea floor by remote sensing methods, because of direct sampling of the sea floor by shallow coring and dredging and later by deep-sea drilling, and because of detailed geological studies of islands where oceanic crust reaches above sea level. Nevertheless, we have far less information about the oceanic crust than about the continental crust. For this reason, and because coal and petroleum appear to be limited in their occurrences to the continental crust, the remainder of this summary deals with the distribution of fuels in the continental crust.

II.1.1 World Coal Resources

Coal occurs in all of the continents (even in Antarctica, although it is not produced there) and in many of the world's large islands [Ward(13)]. It is not uniformly distributed, however, but is concentrated in certain sedimentary basins whose areal extent is a small fraction of the total land area of the earth. Most of the world's coal is in three large countries, the USSR, the USA, and China. Africa and South America have much less coal than do the northern continents.

The world's coal resources have been estimated to be greater than 10 trillion tons, of which about 1 trillion tons are recoverable under present conditions [Ward(13)]. For comparison, the total production of coal in 1981 was about 4 billion tons, so recoverable reserves are several hundred times greater than current annual production. The USA has long been the world leader in coal production. It produced more than 700 million tons of coal in 1981, but the USSR and China were not far behind. In terms of proved reserves, coal is much more abundant than petroleum, and even though past estimates of the world's total petroleum resources have been notoriously inaccurate, there is little reason to doubt that coal is an order of magnitude more abundant than petroleum in the earth's crust.
II.1.2 Worldwide Petroleum Resources

Petroleum is more broadly distributed than is coal in the earth's continental crust. The broad distribution is evident by examination of a table showing world oil production by country in 1962 [Tiratsoo (14)] Table 1/6 in which all of the larger countries of each continent except Africa (and, of course, Antarctica) are listed as significant oil producers. Although petroleum is broadly distributed, it is by no means uniformly distributed. About half of the world's proved recoverable oil reserves are in the Persian Gulf area within a region roughly a million square kilometers in area, about 0.2 percent of the earth's entire surface area [Tiratsoo (14), Table A/9 and figure 4/1].

As is coal, petroleum is found in sedimentary basins, rather than in portions of the earth's crust where uplift and erosion have exposed large areas of plutonic igneous and metamorphic rocks. Tiratsoo (14) [Table 1/19] has listed 580 known sedimentary basins, of which 364 have no production of petroleum. Neither coal nor petroleum is ubiquitous in sedimentary basins, and, although some basins have both coal and petroleum, many basins have one but not the other. For example, coal is insignificant in the Persian Gulf area where petroleum is so abundant, while South Africa has major coal resources and no petroleum.

The proved recoverable oil reserves of the world total about 700 billion barrels. The U.S. Geological Survey has recently begun an effort to estimate the undiscovered recoverable resources of petroleum in every sedimentary basin of the world that is considered to have petroleum potential [Masters (15)]. There are necessarily wide ranges in estimates of this sort, but the most liberal estimates (assigned a five percent probability by the U.S. Geological Survey) provide a total for such undiscovered petroleum that is similar to the total of proved reserves, more than 500 billion barrels but less than one
trillion barrels. These estimates do not include the massive amounts of heavy hydrocarbons that occur as (currently) unproducible heavy oils or tar sands. More than one trillion barrels of such heavy hydrocarbons occur in Venezuela alone. The Athabaska tar sands of western Canada contain hydrocarbons in an amount comparable to the world's proved reserves of recoverable petroleum, about 700 billion barrels.

The United States was the world leader in petroleum production until about 1970 when its production was surpassed by that in the Middle East. Subsequently the Soviet Union has become the world leader in petroleum production. Unlike the Middle East and the Soviet Union, where proved reserves are far greater than total production to date, the United States has much less petroleum remaining in proved reserves than the total of its production during the past. Although in total (production plus proved reserves) the U.S. has had more petroleum than Mexico, Mexico now has greater reserves than the U.S. and follows the Middle East and the USSR in terms of proved recoverable reserves.

II.1.3 The Worldwide Distribution of Natural Gas

As noted earlier, natural gas normally occurs in association with accumulations of petroleum and in coal beds. Of these two kinds of occurrence, the gas associated with petroleum has been by far the most important economically; about 30 percent of the commercially useful natural gas in the world occurs in association with oil [MacDonald¹]. The methane associated with coal beds has been primarily an unwanted byproduct of coal mining, but serious efforts are underway to utilize coalbed methane resources in the U.S., particularly in certain cases where the methane is abundant but the coal cannot be produced economically [Rightmire¹⁶ and others]. The geographical distribution of these kinds of natural gas resources is of course the same as that of the petroleum and coal with which the gas is associated, which has
already been described. In addition, there are many occurrences of natural gas that is not associated either with petroleum or coal, and these occurrences are quantitatively the most important.

The largest gasfields of the world are in the West Siberian Basin of the USSR, where identified recoverable gas reserves are about 700 trillion cubic feet [Tiratsoo (14)]. A similar quantity of natural gas remains undiscovered in this basin, according to Clarke (17) and others. Thus, this basin holds a substantial fraction of the world's total recoverable resources of natural gas, which have been estimated by MacDonald (18) to be about 10 quadrillion cubic feet. The Middle East also has a substantial fraction of the world's gas resources.

Hunt (19) has estimated that the amount of gas dispersed in sedimentary rocks is 10 to 200 times that which occurs in economically useful accumulations, and MacDonald (18) has suggested that the amount of methane stored in methane hydrates may be more than one thousand times greater than the world's reserves of recoverable natural gas. It should be noted that even greater quantities of solid organic material occur dispersed in the world's sedimentary rocks. This material is neither coal nor petroleum nor natural gas, but it is the predominant form of reduced carbon in the crust of the earth. The amount of carbon dispersed in sedimentary rocks is estimated to be of the order of $10^{16}$ tons, roughly a thousand times more than that present in economically useful concentrations as coal, petroleum, or natural gas.

II.1.4 Age and Character of the Rocks Associated with Natural Fuels

Virtually all of the economically useful accumulations of natural fuels that have been found in the earth are in sedimentary basins, and so the association of fuels with sedimentary rocks is a very strong one. The fuels sometimes occur within or in contact with igneous rocks, but in nearly all such
cases the igneous rocks have been intruded into or have been extruded over pre-existing sedimentary rocks; such cases do not affect the generality of the association of fuels with sedimentary rocks.

Coal is found only in sequences of sedimentary rock (including metamorphic rocks that have been formed by metamorphism of sedimentary rocks) and is considered by geologists to be itself a sedimentary rock. The geological evidence for this genetic classification of the material is not simply the universal association of coal with sedimentary rocks, but rather the evidence derives from the structural and stratigraphic relationships between the coal and the sedimentary material that surrounds the coal, and from the petrographic characteristics of the coal itself. Some of the more important aspects of this evidence are as follows:

- "Hardly any coal seam consists of pure coal substance; nearly everywhere the coal is interlayered with stratified impurities (usually shale)." [van Krevelen (20)] The character of these non-coal 'bands' or 'partings' and their stratigraphic relationship to the coal are strong evidence for the sedimentary origin of coal [Ward (13)].

- On a larger scale, the commonly observed splitting and coalescence of coal seams and the discontinuous nature of many coal seams are features consistent with their origin in a complex sedimentary environment (for example, a major river delta) where there were continual changes in the local environment as a consequence of changes in positions of river channels and shorelines.

- Unlike volcanic rocks formed from extruded lava, which are often associated with dikes or plugs that clearly cut across the structure of the rocks through which the molten magma moved, coal is not observed to be associated with cross-cutting bodies of similar composition. (On
the other hand, asphalt commonly occurs in veins that cut across pre-existing rocks [Hunt (21) and others], evidence that it has moved as a liquid. In cases where magma has intruded across and into coal seams, the coal has been quite distinctly altered by loss of volatile constituents [Ward (13)], which suggests that the coal had never been exposed to high temperatures except where it is in proximity to the igneous rock.

- The fine structure of coal shows it to have a texture that is consistent with a sedimentary origin. The texture is quite different from the texture of rocks that have demonstrably formed by solidification of a liquid. One of the most important observations of coal petrography is the common occurrence of the well-preserved structures of plant cells, spores, and algae. Unlike macroscopic plant fossils, which are generally more abundant in shaly partings than in the coal itself, the microscopic plant remains are part of the coal macerals—the essential substances of the coal itself [van Krevelen (20)].

The occurrence of coal is notably restricted in time, that is, in respect to the ages of the rock sequences that contain coal. (Table 1 is a recent version of the geological time scale, reproduced here to show the relation of the various geological time periods to one another and the approximate absolute time interval with which each is associated.) The oldest coal-bearing strata were deposited in the Devonian Period, not more than about 400 million years ago. (A carbonaceous material called shungite occurs in older metasedimentary rocks in Fennoscandia, but this material is thought to have been asphalt originally rather than coal.) Nearly all coal is found in rocks formed during or after the Carboniferous Period, which began about 350 million years ago.
The absence of coal in rocks older than 400 million years has been interpreted as a consequence of the virtual absence of land plants on earth prior to that time. Coal is not evenly distributed in rock younger than 350 million years; rather it is much more abundant in rocks of late Paleozoic age (roughly from 250 to 350 million years old) and in rocks of late Cretaceous and Tertiary age (less than 100 million years old) than it is in rocks formed in the period between these age groups.

Petroleum and natural gas are fluids that migrate through permeable rocks, and the evidence that these materials may have originated from organic sedimentary material is quite different in character from the evidence that coal is a sedimentary rock. Since the fluid fuels have generally moved through permeable rock until trapped by some sort of relatively impermeable barrier, the structure and texture of the rock that contains the fuels—the reservoir rock—are usually unrelated to the origin of the fuels. For example, natural gas has been produced from volcanic rocks (vesicular basalts) of the Columbia Plateau in Washington; the gas presumably originated in coal-bearing sediments below the basalt [Swain (22)].

Traditional geological methods cannot pinpoint the origin of the fluid fuels. The belief held by most geologists that petroleum and most natural gas deposits originate from organic material in sedimentary source rocks is based on a complex set of data that includes a great deal of chemical and isotopic information as well as a vast amount of information from geological and engineering studies in the oilfields. The fact that these fuels have been found in significant quantity only in sedimentary basins is an important part of the case for their sedimentary origin but is by no means the whole story. One might question this part of the evidence on the ground that virtually no exploration for petroleum has occurred outside of the world's sedimentary
basins, but it should be noted that discovery of many oilfields, particularly in the early days of the petroleum industry, was aided by the presence of natural seeps. Such occurrences have not been noted outside of the sedimentary basins.

Petroleum and natural gas are often found in rocks older than the oldest coal-bearing rocks, and in a few cases petroleum-bearing rocks are of Precambrian age, more than 600 million years old. The world's oldest oil may be that found in Australian rocks about 1500 million years old [Cook (23) and others]. Significant accumulations of gas and petroleum have been found in Precambrian rocks in Eastern Siberia. Of course, the age of a reservoir rock is not the same as the age of the petroleum or gas that it contains. But since these fluids generally move upward through the earth's crust, and since in sedimentary basins older rocks are normally below younger rocks, there is reason to believe that the source of an accumulation of petroleum or natural gas is generally as old as or older than the reservoir rock. Natural gas is rather commonly found and petroleum is sometimes found in rocks of early Paleozoic age, but the fuels are more abundant in younger rocks. Rocks of Mesozoic age are particularly important as producers of petroleum (in contrast to their relatively small importance for coal).

In summary, the portions of the earth's crust that are known to contain coal and petroleum are restricted in geographical distribution and in the character and age of the rocks in which the fuels occur. Our known coal and petroleum resources occur only in sedimentary basins, coal in rocks not more than 400 million years old and petroleum in rocks usually less than 400 million years old but sometimes in older rocks (as much as 1500 million years old). These fuels are unknown in portions of the continental crust comprising plutonic rocks or very old sedimentary rocks, as well as in the entire oceanic
crust. Natural gas may be somewhat more broadly distributed, particularly as methane hydrates in oceanic sediments, but the gas known to be recoverable by present technology is restricted in its distribution in the same way as petroleum.

II.2 Coal, Petroleum, and Natural Gas in the United States: A Brief Summary

II.2.1 Coal

Ward (13) has given a brief summary of the geographical and geological distribution of coal in the United States and has cited examples of recent literature that give detailed information on specific areas. A map by Averitt (24) showing the distribution of coalfields in the conterminous United States is reproduced here as Figure 1. Coal-bearing strata occur in 37 states and underlie about 13 percent of the land area of the U.S. The major coalfields are in the Appalachian Mountain region, the Rocky Mountain region, and a few basins in the continental interior. Although there are numerous exceptions, there is a general decrease in rank of coal going from east to west in the United States. Also, the coals of the eastern U.S. are mostly in strata of Pennsylvanian age, whereas those of the western states are mostly in younger (Cretaceous and Tertiary) strata.

Anthracite occurs in the Appalachian fold belt where strong tectonic activity subjected the coal to high temperature as a consequence of deformation and deep burial. The major anthracite fields are in eastern Pennsylvania and in the Narragansett Basin of Rhode Island and Massachusetts. Great quantities of bituminous coal occur in the Appalachian Plateau region and in the Illinois Basin. Higher rank (low-volatile) bituminous coal is found in the eastern part of the Appalachian Plateau and in some adjoining parts of the Appalachian Ridge and Valley province where moderate tectonic activity caused the coal to be affected by somewhat elevated temperatures. High-volatile bituminous coal
Figure 1

Distribution of coalfields in the conterminous United States: ■ Anthracite and semi-anthracite; □ low volatile bituminous coal; △ medium and high volatile bituminous coal; ♂ sub-bituminous coal; ± lignite (Averitt 1975).
predominates in the western part of the Appalachian Basin and in the Illinois Basin. Although the abundant coals of the eastern U.S. are of late Paleozoic (mostly Pennsylvanian) age, there are some younger coals in this part of the country. Low-rank coal occurs in rocks of early Mesozoic age in small sedimentary basins within the Appalachian Piedmont region, and lignite is widespread in Tertiary sedimentary rocks of the Gulf Coastal Plain.

Coals of the western U.S. and Alaska are generally of lower rank than the Appalachian deposits. The most extensive coals of the western U.S. are in beds of late Cretaceous age in several large basins and numerous small basins in the Rocky Mountain region. These are mostly of high-volatile bituminous and sub-bituminous rank. In some of these basins, coal seams are present in beds of Tertiary age overlying the Cretaceous deposits; coal and lignite of Tertiary age also occur in other basins in the western U.S. The most important area of Tertiary coals is the western Washington coal region, the only major area of coal occurrence in the far-western states.

None of the major coal deposits of the eastern U.S. are deeply buried. The bituminous coals of the Appalachian Basin and the Illinois Basin are distributed throughout the uppermost sedimentary rocks of these basins, extending downward only a few thousand feet. Except at the southern end of the Appalachians in Alabama, where coal-bearing strata may be deeply buried under thick sediments of the Gulf Coastal Plain, and in the anthracite-bearing Appalachian fold belt, where strong tectonic deformation has caused portions of the coal beds to be deeply down-folded, the deepest Appalachian coal beds are only about 2300 feet below the surface. In the Illinois Basin, all the coal seams are within 3000 feet of the surface and the major coals are within 1500 feet of the surface [Archer(25) and Kerr].
The tectonic character of the Rocky Mountain region has led to thick accumulations of sediments in the intermontane basins. Consequently, even though the coal-bearing strata of this region are younger than those in the Appalachians, the coal in some of the Rocky Mountain basins extends to much greater depths than does the coal in the main part of the Appalachian Basin. Overburden as thick as 10,000 feet occurs over coal in parts of the Piceance Basin of Colorado [Choate (26) and others]. Such deeply buried coal beds may become quite important for the methane they contain, even though they are too deeply buried for coal mining to be practical [Rightmire (16) and others].

II.2.2 Petroleum

As in the world as a whole, petroleum in the United States is more broadly distributed than is coal. Petroleum accumulations are found in most of the coal-bearing basins, but the greatest oilfields are not associated with coal. The major oilfields of the U.S. are not in the basins where coal is important. Ten of the twelve largest oilfields of the U.S. are in Texas and California where coal is not significant [Tiratsoo (14), Tables 7/8 and 7/18]. The other two (of the top twelve) are the great Prudhoe Bay field in Alaska and one in southern Oklahoma; neither of these is in a coal-bearing area. The Louisiana Gulf Coast is another major region of oil production that is not associated with coal.

Major petroleum accumulations are generally at greater depths than the major coal deposits of the U.S., although some of the earliest-discovered oilfields extend quite close to the surface. According to Tiratsoo (14) most oils and gas accumulations are at depths of 2000 to 10,000 feet, but sustained production has been established to depths as great as five miles. The ages of the rocks containing petroleum also vary more broadly than those of coal-bearing rocks. Particularly in the Appalachian region and the
midcontinent, petroleum often occurs in reservoir rocks that are older than the oldest coal-bearing strata. Petroleum reservoirs of Devonian and Silurian age are common in the Appalachian region, and even older (Ordovician) strata are important reservoirs in the midcontinent. In sharp contrast are major oilfields in California in thick, very young (Pliocene) sequences of sediments formed by rapid sedimentation in a tectonically very active region.

II.2.3 Natural Gas

Natural gas is always present in association with coal and petroleum, except where they are so close to the earth's surface that the gas has been able to escape. Hence, the foregoing summaries of the distribution of coal and petroleum in the United States are also pertinent to the distribution of natural gas. But natural gas has many occurrences where it is not associated with coal or petroleum.

As was noted above the natural gas has been obtained from volcanic rocks in Washington State, but the normal occurrence of natural gas that is not associated with oil or coal is in the sedimentary basins. It often occurs as a gas phase within permeable sedimentary rocks that are capped by impermeable rocks that prevent the gas from escaping. Such gas reservoirs are structurally like petroleum reservoirs. There is a variety of reasons why such reservoirs may often contain gas but no oil, but the factor most universally recognized as important is the effect of depth and the geothermal gradient (that is, the increase of temperature with increasing depth in the earth). Natural gas is commonly found at greater depth than is oil within a given petrolierous region, presumably because gas is stable at temperatures that are (or were) too high for petroleum to exist. The common occurrence of gas at greater depth than petroleum is not to be confused with the vertical relationship of oil and free (that is, undissolved) gas in any particular petroleum pool where both
occur; in such cases the free gas is always above the oil because the oil is more dense. The same reasoning is also applicable to gas pools in rocks that now are fairly close to the surface but were once deeply buried, as in parts of the Appalachian Mountains where erosion has removed a great deal of the original sedimentary cover.

The accumulation of sediments is not the only way in which rocks become deeply buried. Important occurrences of natural gas occur in rocks that have been buried by the action of great thrust faults where thick sheets of rock have been pushed laterally over sedimentary layers that were originally near the surface. Both petroleum and gas occur in the Western "overthrust belt," including an interesting recent discovery of a major gas pool below crystalline Precambrian rocks that have been thrust over younger sedimentary rocks. There is considerable interest in the possibility that similar occurrences might be present in the "eastern overthrust belt," which includes at least a part of the Appalachian Piedmont region, once thought to be outside the realm of exploration for oil and gas.

In addition to the natural gas that is present in reservoirs from which it is recoverable by present technology, the United States may have vast quantities of natural gas in other forms. These include methane hydrates in deep waters offshore and perhaps in the Alaskan tundra, gas dissolved in formation water under high pressure deep underground, and possibly significant amounts of gas from deep within the earth, which might have accumulated in regions that have not been explored for fuels.

II.3 Coal, Petroleum, and Natural Gas in Pennsylvania

Pennsylvania has been a world leader in coal production for well over a century and has been a significant producer of oil and natural gas since the world's first modern oil well was drilled there in 1859. In a large area of
western Pennsylvania, there are important accumulations of all three fuels, but
the vertical distributions of each of the three fuels differs from that of the
others. In the state as a whole, there are also important differences in the
area distributions of the fuels.

As noted in the preface, there is a vast amount of information on the
worldwide distribution of natural fuels, and so this report will use
information about the distribution of fuels in Pennsylvania as a convenient
example of the complexity of the geographical and geological distribution of the
materials. Even for this one state, the amount of information available is far
too great for a report of practical size to be complete. For example, there
are more than 25,000 wells in the giant Bradford oil field (which extends from
Pennsylvania into New York State) alone [Tiratsoo (14)]. So this part of the
report, on fuel occurrences in Pennsylvania, is also necessarily a summary.

II.3.1 Coal

Coal is broadly distributed in Pennsylvania, from anthracite fields in the
eastern part of the state to the bituminous fields that occupy most of the
Appalachian Plateau in the western part of the state (Keystone Coal Industry
Manual, 1974, Figure 1). In comparison to the main bituminous field, the
anthracite fields are small and discontinuous, a consequence of the effect of
severe folding of the sedimentary strata in the eastern part of the state (at
the close of the Paleozoic Era) and subsequent erosion of much of the original
rock. There are scattered coal fields in the central part of the state, where
the strata are more gently folded than those further east. In the western
third of the state—the Appalachian Plateau province—the coal-bearing strata
were beyond the direct influence of the tectonic stresses that folded the rocks
further to the east, so the sedimentary strata are little changed in
orientation from the nearly horizontal attitude in which they were deposited.
But this area, the main bituminous coal field, is not structureless; there are gentle troughs and swells and the entire region forms a structural basin that deepens southwestward. The deepest part of this "spoon-shaped" basin, which extends across western West Virginia and eastern Ohio, is in the southwestern corner of Pennsylvania and the adjoining portion of West Virginia. The deepest coal seams there are about 2300 feet below the surface. These coal seams, the lowermost ones in the stratigraphic sequence, are found at generally shallower depths as one moves northward across the western part of the state until, in the counties that border on New York State, there are only scattered occurrences of coal, close to the surface.

In the major bituminous fields there are dozens of individual coal seams, variable in thickness but typically reaching maximum thicknesses of three to six feet. These seams are distributed within a sequence of sedimentary rock that is more than 2000 feet thick in the southwestern corner of the state. The Keystone Coal Industry Manual (1981) shows 39 coal seams in the main bituminous field, distributed within a sequence of rocks whose cumulative average thickness is 2400 feet. Such a listing of coal seams is a generalization that focuses on the major seams that can be identified by correlation over broad areas; it does not include many minor occurrences of coal, not does it indicate the complexity that is characteristic of coal distribution—the tendency of coal beds to be discontinuous or at least to be quite variable in thickness, and the common splitting and coalescence of coal seams. For example, the oldest coal-bearing series of strata in Pennsylvania, the Pottsville Group, has "... a sporadic occurrence of coals with highly variable thicknesses and limited lateral extents. The main coals of this group are, in ascending order: the Sharon, Quakertown, and Lower and Upper Mercer" [Adams (27) and others]. Only the four main coals of the Pottsville Group are shown in the Keystone compilation.
The rank of coal varies systematically from anthracite in eastern Pennsylvania to high-volatile bituminous coal in the western part of the state. There is also some variability in rank to be expected with depth in the westernmost part of the state where there has been little tectonic disturbance. But the east-to-west variability in rank is much greater than any vertical variability, a consequence of the history of tectonic deformation and erosion that has brought once deeply buried coals to the surface in eastern and central Pennsylvania.

Most of the coals in Pennsylvania are in strata that were deposited during the Pennsylvanian Period. In the southwestern corner of the state, however, coal occurs in the Dunkard Group, which is at least partly of Permian age. The sedimentary rocks that contain the coals include both marine and nonmarine strata. These sandstones, shales, and limestones record a complex history of sedimentation that includes alternating periods of marine transgression and of fluvial delta growth seaward as clastic sediments were carried westward from the early Appalachian Mountains.

### II.3.2 Oil

Oilfields are spread throughout the western portion of the main bituminous coal field in Pennsylvania, but there are only scattered occurrences of oil in the eastern portion of this large coal field. Except in the extreme southwestern corner of the state, where oil occurs in sandstones of the Pennsylvanian Conemaugh Group, wherever oil and coal occur in the same area of the state, the oil is in strata that are older, and thus below, the coal-bearing strata [Ver Wiebe (29)]. The oilfields are not limited to the coal-bearing part of the basin; they extend northward beyond the coal fields into southwestern New York State. The giant Bradford oilfield is in this area north of the coal fields, where the rocks now exposed at the surface are older than the coal-bearing rocks.
The oilfields of Pennsylvania are generally associated with swells--very gently folded anticlinal structures--in the Appalachian Plateau region, where oil is trapped in permeable sandstones beneath impermeable formations. However, there are many cases where oil has been found in isolated sand bodies surrounded by impermeable rock, which forms a stratigraphic trap. Such oil pools are found in synclinal areas as well as anticlinal areas. The principal oil reservoirs are of Devonian age, roughly 50 to 100 million years older than the coal-bearing strata. The depth of the oil reservoirs is quite variable, but there is a general trend to increasing depth southwestward, in correspondence with the general structure of this part of the Appalachian Basin.

II.3.3 Natural Gas

Natural gas occurs in association with petroleum and with coal in Pennsylvania, as it does worldwide. There are also important gas pools in western Pennsylvania that are outside of the coal- and oil-producing areas, at least vertically (that is, by being deeper) if not areally. For example, oil is virtually absent from the eastern portion of the main bituminous coal field, but there is a major belt of gas reservoirs in that area; however, many of the gas reservoirs are deeper than the coal beds. As do the oilfields, the gas fields extend further north than the coal fields, but the distribution of gas is not the same as that of the oil. There are deep gas reservoirs in the northern tier of counties both to the east (as far as Tioga County) and to the west (to the Ohio border) of the main oil-producing belt. Some of the gas reservoirs are rocks of Silurian age, older than the important oil-producing sandstones. The Oil and Gas Map of Pennsylvania (1973) shows one gas field in Lackawanna County, in eastern Pennsylvania, close to part of the anthracite coal field, and some deep gas pools in Bedford County in the Appalachian Ridge and Valley Province.
Although coal is much more abundant in Pennsylvania than either oil or natural gas, gas occurs almost as widely (but not as continuously) across the state as does coal. In the vertical sense (and thus in the sense of geological age), gas is much more broadly distributed than is coal, with major occurrences in strata from Silurian age upward throughout the younger Paleozoic rocks.

III. AN EVALUATION OF POSSIBLE METHODS FOR RADIOMETRIC DATING OF COAL AND PETROLEUM

III.1 General Considerations

The insensitivity of nuclear decay constants to changes in physical and chemical conditions and the occurrence of a variety of radionuclides in most natural materials provide the basis for the radiometric dating methods, which have proved to be the only methods capable of providing accurate values for the absolute ages of materials over the whole range of geologic time. The radiometric dating methods may be classified in three groups:

1. Methods based on the decrease in abundance of a radionuclide from the time a material was formed to the present. Such a method requires measurement of the present amount of the radionuclide, and the original amount of the radionuclide must be established in some way. Radiocarbon dating is the best known and most widely used method in this category.

2. Methods based on the increase in amount of a daughter nuclide formed by decay of a radionuclide. In such a method, the present amounts of the parent and the daughter must be measured, and the initial amount of the daughter must be established in some way. The potassium-argon method is the most widely used method of this kind, but there are several other parent-daughter pairs that are of comparable importance in geochronology.
3. Methods based on the amount of radiation damage that has accumulated in a material. Such a method requires measurement of the amount of accumulated damage and a means of establishing the rate of accumulation of the radiation damage. Fission-track dating is the most widely used method of this kind.

Methods belonging to the first of these three categories may be applied to fluid materials as well as to solids, but methods in the second and third categories are generally applicable only to solids. Consequently, although there are a number of possible methods for radiometric dating of coal, the radiocarbon method is the only one that might reasonably be used to date oil and natural gas. Unfortunately, radiocarbon is relatively short-lived; materials older than 100,000 years will not have a measurable amount of $^{14}$C remaining. Quite generally, the geological environments in which petroleum accumulations are found imply that the petroleum is much older than 100,000 years. The same is true for significant accumulations of natural gas, although there are some near-surface environments in which methane has formed in the recent past (and is being formed today). Radiocarbon dating is not useful for dating petroleum and major accumulations of natural gas because any radiocarbon that may once have been associated with these materials (or their precursors) has long since decayed away.

Coal cannot be dated by the radiocarbon method for the same reason the method is not applicable to petroleum. All known coal deposits are presumed to be much older than 100,000 years, and there is no evidence that radiocarbon is present in any coal. But since coal is a solid material, there are a number of other radiometric dating methods that might be used for dating coal deposits. In comparison to other kinds of rocks, very little consideration has been given to the possibility of dating coal by radiometric methods, because none of the
long-lived radionuclides that are so useful for dating silicate rocks is part of the essential constituents of coal. But these long-lived radionuclides are generally present as trace constituents of coal, either as part of the mineral impurities in the coal or as trace constituents of the substance of the coal itself.

Because of the great sensitivity of the methods currently used for isotopic measurement of radionuclides and their stable daughters, there is no doubt that techniques for accurate measurement of these trace constituents of coal could be devised. But there may be considerable doubt about the significance of such measurements, because of the heterogeneity and porosity of coal.

The long-lived radionuclides that are useful for dating rocks are isotopes of the metallic elements potassium, rubidium, samarium, thorium and uranium, elements that are present in the earth's crust primarily as constituents of silicate minerals. Silicate minerals are ubiquitous in coals, largely as the very fine-grained materials known as clay minerals. In most if not all cases, there will be a considerably greater proportion of the long-lived radionuclides in the clay minerals than in the organic matter of coal. Unless an effective technique is developed to measure the amounts of a radionuclide and its daughter in the organic matter, separately from the larger amounts in the mineral matter, it is unlikely that useful information about the age of coal could be obtained by any radiometric dating method. The reason the mineral matter must be excluded from the measurements is that the minerals are generally detrital materials, which were formed in some other environment and transported to the site of deposition of the coal. Generally, such detrital material is considerably older than the coal that contains it, so the proportion of a radionuclide and its daughter in the detrital material is unrelated to the age of the coal. There may be an important exception to this
generalization, however. There is considerable evidence that clays may be altered during the formation of coal. If the degree of alteration of the clay is sufficiently great, then (some of) the clay minerals in or below a coal bed may in effect be new minerals having essentially the same age as the coal. In such a case, radiometric dating of the clay may be quite useful.

A major problem that may affect any attempt to directly date the organic matter of coal is the high degree of porosity of the material. Coals have intricate internal structures with a high percentage of very fine pores [Gan (29) and others]. Gas-adsorption studies have been used to show the nature of the porosity of coal and to gain information about the way in which the micropores are interconnected. The low density of coal and the high surface areas determined by the gas-adsorption studies suggest that coal is not likely to meet an essential criterion for radiometric dating based on the accumulation of a stable daughter nuclide, that is, that the material should have been a "closed system" in respect to the radionuclide and its daughter since the material formed. The problem may be particularly serious in cases where a daughter element is a gas, but ions of metallic elements may also migrate through coal since coal in place underground is usually saturated with water.

Following is an evaluation of each of the more commonly used radiometric methods of dating rocks in respect to its potential usefulness for dating coal.

III.2 The Potassium-Argon Method

The potassium-argon method is normally much more sensitive than the other radiometric dating methods, because the daughter element is an inert gas that is not normally incorporated into minerals when they are formed. The virtual absence of any of the daughter element initially means that the potassium-argon method may be useful for materials that contain very small amounts of
potassium. Dalrymple and Lanphere (30) gave a comprehensive description of the method, and their book is the best source of information on the principles of conventional potassium-argon dating. A variation of the potassium-argon method known as $^{40}\text{Ar}/^{39}\text{Ar}$ dating has come into common use since their book was published (see Dallmeyer 31). In most applications, the $^{40}\text{Ar}/^{39}\text{Ar}$ technique can provide more useful information about the relation of potassium and argon than can conventional potassium-argon measurements.

The abundance of potassium in coals is more than sufficient for potassium-argon dating of the bulk material, but it is likely that nearly all of the potassium is part of mineral impurities rather than part of the coal. If, however, there is a small percentage of potassium in the organic material, then it is possible that potassium-argon measurements could be used to determine when the coal reached its present form (presumably some time after the actual deposition of the organic material).

For potassium-argon dating of coal to be successful, it is necessary that the coal has retained virtually all of the radiogenic argon that has formed within it, that any extraneous argon in the coal must have a definitive isotopic composition that would allow it to be distinguished from the radiogenic argon, that any such extraneous argon not be so abundant as to overwhelm the small amount of radiogenic argon that may be present, and that the potassium and argon in the organic part of the coal can be separated for measurement from potassium and argon in potassium-bearing minerals that are present in the coal.

The potassium-bearing minerals in coal, mostly clay minerals, are largely of detrital origin, which means that they would have already contained some radiogenic argon at the time they were incorporated into the coal. Such "inherited" radiogenic argon is related to materials and events that predate the coal, so the apparent potassium-argon age of the clay minerals within a
coal bed is likely to be greater than the age of the coal. But clay minerals are susceptible to alteration during and after their deposition in sedimentary layers, so one cannot make specific predictions about the apparent potassium-argon ages of sedimentary clays without some knowledge of the history of the rocks that contain them. Conversely, potassium-argon studies of clay minerals in sedimentary rocks of known age can provide a great deal of information about the history of the rocks [Sedivy and others(32)].

Recent work at Georgia Tech (not yet published) has shown that potassium-argon dating of rock salt having as little as 0.005% potassium is possible. This work was done by a new technique for potassium-argon dating, which takes advantage of the solubility of salt and the insolubility of silicate minerals in water. If a method could be found to release argon and potassium from the organic matter of coal without affecting the silicate minerals in the coal, then dating of coal may be possible even when there is very little potassium in the organic material. But the extent and nature of the porosity of coal may preclude satisfactory results. It is unlikely that the radiogenic argon formed within coal is quantitatively retained in the material, because of the opportunity provided by the network of micropores for such molecules to escape. The network of micropores also provides a path for extraneous argon to permeate the coal, and it may not be possible to get all such extraneous argon out of the coal before analysis. In this respect, coal is quite unlike rock salt, which appears to be essentially impermeable to argon. Unless virtually all of the atmospheric argon which permeates a coal sample can be removed in the laboratory, the K-Ar method would not have for coal the great sensitivity that makes it useful for dating rock salt of low potassium content.
An alternative approach would be to use the $^{40}$Ar/$^{39}$Ar method, if the organic material releases argon under vacuum in a temperature range that is different from that at which the silicates release argon. [In the $^{40}$Ar/$^{39}$Ar method some of the $^{39}$K in a specimen is converted to $^{39}$Ar by fast-neutron irradiation, and the amount of $^{39}$Ar is then measured instead of measuring potassium directly. Since all of the isotopes of argon are released from a particular kind of material under the same conditions, potassium–argon apparent ages for different portions of a specimen may be obtained by releasing argon from the specimen in steps, usually by heating at progressively higher temperatures.) In principle, the information obtained by stepwise heating of material that has been irradiated for $^{40}$Ar/$^{39}$Ar study can be used in favorable cases to get around the problem of atmospheric contamination and the problem of partial loss of the radiogenic argon within a material. But in a material with a thoroughly penetrative network of very fine pores, it is likely that recoil during formation of the $^{39}$Ar will lead to considerable loss of $^{39}$Ar from the material. Unless some domains exist in the coal that are impermeable to argon and at least one micrometer in diameter, it is not likely that $^{40}$Ar/$^{39}$Ar studies of coal would be useful for dating. There is no reason to believe that such domains exist in the organic material of coal.

In summary, there appear to be some formidable problems in the application of potassium–argon dating to the organic material of coal, either by conventional methods or by the $^{40}$Ar/$^{39}$Ar technique. On the other hand, the clay minerals in coal certainly can be dated by the potassium–argon technique, and if the process by which the coal formed had a sufficiently strong effect on the clay, either by causing chemical alteration or possibly by causing thermal alteration, then such dating of the clay should provide useful information about the origin of the coal.
III.3 The Rubidium-Strontium Method

$^{87}\text{Rb}$ decays to $^{87}\text{Sr}$ with a half-life of about 49 billion years. Since rubidium is quite similar to potassium in its geochemical behavior, the Rb-Sr method is applicable to many of the materials for which potassium-argon dating is used. It has one great advantage over the K-Ar method: the daughter element is often retained in rocks under conditions of elevated temperature where argon will be lost. On the other hand, the fact that argon is a gas gives the K-Ar method a great advantage over the Rb-Sr method; in many cases the original amount of argon in a rock is nil, so the K-Ar method is generally much more sensitive than the Rb-Sr method. Strontium is always present as an original constituent of rubidium-bearing minerals and rocks, usually in sufficient quantity that the increase in $^{87}\text{Sr}$ content caused by decay of $^{87}\text{Rb}$ cannot be measured accurately unless the material is very old or relatively rich in rubidium. (For a similar reason, the decay of $^{40}\text{K}$ to $^{40}\text{Ca}$ is not used in radiometric dating. The K-Ca method is even much less sensitive than the Rb-Sr method.)

Like potassium, most of the rubidium in coal is presumably associated with the detrital minerals present in the coal. Consequently, Rb-Sr measurements on whole-rock samples of coal are not expected to provide useful information about the age of the coal (unless the detrital clay minerals were thoroughly altered during formation of the coal). As in the case of the K-Ar method, if a way could be found to measure the trace constituents of the organic matter in coal without any influence from the detrital minerals, then Rb-Sr measurements could perhaps provide information about the time when the coal formed. But because the rubidium content of the organic matter is likely to be extremely low, it is unlikely that the Rb-Sr method would be sufficiently sensitive in this application. The microporosity of coal is likely to be as severe a problem for
Rb-Sr dating of coal as it would be for K-Ar dating, since strontium is likely to be as mobile as (if not more mobile than) argon in water-saturated coal.

In summary, the possibility of dating coal by the rubidium-strontium method appears to be very slight, because the rubidium content of the organic material in coal is probably very low, because the separation of such trace metals in the organic material from what is present in detrital minerals is likely to be very difficult, and because the microporosity of the organic material makes it unlikely that the material has behaved as a closed system for rubidium and strontium. Although the same problems apply in the case of the potassium-argon method, the sensitivity of the K-Ar method is normally so much greater than that of the Rb-Sr method that any work done on coal for possible dating by these methods should first be focused on the K-Ar method.

111.3 The Samarium-Neodymium Method

$^{147}\text{Sm}$ decays to $^{143}\text{Nd}$ with a half life of about 107 billion years. Because samarium and neodymium are both lanthanides, quite similar to one another in their chemical properties, the degree to which the two elements are separated from one another during geochemical processes is slight. For this reason and because of the smallness of the decay constant, the Sm-Nd relationship was not useful for dating rocks until the advent of methods for very high precision mass spectrometry of metallic elements in the late 1960's. After successful application to studies of meteorites in the early 1970's, Sm-Nd dating of terrestrial rocks began about a decade ago. The method has been quite useful for studies of many very old rocks that cannot be dated by other methods because of alteration of the rocks. The Sm-Nd relationship is much less sensitive to the effects of alteration than are the more commonly used parent-daughter pairs because of the geochemical character of the lanthanides. Like other Group III elements, these elements have very little
mobility in most natural fluids, so a small volume of rock may have behaved as a closed system in respect to these elements even though more mobile elements may have moved into or out of the rock.

It is probable that virtually all of the samarium and neodymium in coal is part of the detrital mineral impurities in the coal. Unless there were some way to effect an almost perfect separation of the organic matter from the mineral impurities, an analysis of samarium and neodymium would likely be dominated by the influence of the detrital minerals. Even if a perfect physical separation could be done, and even if there were enough Sm and Nd in the organic material for accurate measurement (neither of which is likely), the Sm-Nd method would not be favorable for dating coals. The method is best suited for rocks that are billions of years old, but all known coal deposits are within rocks that are less than half a billion years old.

III.4 The Uranium-Lead Method

Uranium has two characteristics, not shared by the other long-lived radioelements used in radiometric dating, that may in some cases allow it to be used for dating coal. First, uranium has two different oxidation states in the earth's crust. Under the reducing conditions that prevail in many environments where organic materials are abundant, uranium may be immobilized by reduction to $^{IV}$. Consequently, uranium is sometimes highly enriched in materials that contain a lot of organic matter. Second, uranium has two long-lived isotopes, each of which decays via intermediates to a stable isotope of lead. This fortunate circumstance--two different parent-daughter pairs involving the same two elements--sometimes allows radiometric dating of uranium-bearing materials even if the materials have not been closed systems for uranium and lead.

The applicability of the uranium-lead method to organic-rich shales was evaluated by Cobb and Kulp. They found evidence that some of the elements
in the uranium decay series as well as the stable lead isotopes formed in the last step of decay had moved within the porous rock, but they still were able to infer values for the ages of the rocks that have proved to be approximately correct. Since that time the uranium-lead method has been developed into a quite reliable method for dating certain minerals, particularly zircon, in spite of the fact that such minerals normally have lost some of their radiogenic lead. The theoretical basis for establishing the age of a material that has lost lead was first given by Wetherill.

Although uranium is commonly enriched in marine black shales, which form in anoxic environments, the uranium content of coal is not usually notably greater than that of ordinary rocks. Some coals have an enrichment of uranium, however, and there is considerable evidence that uranium is associated with the organic material in coal (Van der Flier and Fyfe). Some of the uranium in coal must occur as a trace constituent of the detrital minerals, but in many cases a comparable or greater amount of uranium may be in the organic matter.

If uranium-lead dating of coal is to be successful, it will be necessary to find and isolate material that has a favorable ratio of uranium to original lead. Since lead is also sometimes enriched in rocks that formed in a reducing environment, because of its insolubility as the sulfide, it may be that most coals do not have a favorable ratio of uranium to lead. Perhaps, if the organic material of coal could be effectively separated from sulfide minerals as well as silicate minerals before analysis, the amount of original lead would not be a serious problem.

Because there are a series of energetic decay steps leading from uranium to lead and because of the porous nature of coal, it is unlikely that coal behaves as a closed system for uranium and lead or for some of the intermediate nuclides in the decay series. Nevertheless, there remains a possibility for
dating such open material if certain conditions have been met. Most important of these conditions is that lead should not have moved into the coal from the outside since the coal formed (but it is permissible for some of the radiogenic lead formed within the coal to have been lost to the outside). Another important condition is that there should not have been a severe loss of intermediates in the $^{238}\text{U}$ decay series. The most likely candidate for loss is the relatively long-lived $^{226}\text{Ra}$.

In summary, uranium is the only one of the long-lived radioelements used for radiometric dating that might generally be more abundant in the organic matter of coal than in the mineral impurities in the coal. But the concentration of uranium in most coals is very low, and unless the content of ordinary lead is correspondingly low, uranium-lead dating of the material is not likely to be possible. Even in cases where coal may have a favorable ratio of uranium to lead, the porosity of the coal may have allowed continual movement of uranium, lead, and perhaps radium into and out of the coal to such an extent that U-Pb measurements will not provide useful information about the age of the coal.
As an example of U- and thorium-lead dating consider the following decay schemes:

1. $^{238}U$ decays ultimately into $^{206}Pb$ (stable)  
   (See Table II.)

2. $^{235}U$ decays ultimately into $^{207}Pb$ (stable)  
   (See Table III.)

3. $^{232}Th$ decays ultimately into $^{208}Pb$ (stable)  
   (See Table IV.)

The differential equations governing the decay of $^{238}U$, $^{235}U$, and $^{232}Th$ are respectively:

For lead the following differential equations apply to U to Pb decay

$$\frac{d \text{Pb}^{206}}{dt} = \lambda \text{N}^{238}(t) \text{N}^{238}(t); \frac{d \text{N}^{238}}{dt} = \lambda \text{N}^{235}(t); \frac{d \text{N}^{235}}{dt} = \lambda \text{N}^{232}(t)$$

where $N(t)$ is the number of atoms at time $t$

$\lambda$ is the decay constant.

The solutions for these equations as a function of time is:

$$N^{238}(t) = N^{238}(o)e^{-\lambda t}; N^{235}(t) = N^{235}(o)e^{-\lambda t} \text{ and}$$

$$N^{232}(t) = N^{232}(o)e^{-\lambda t}$$

For lead the following differential equations apply to U to Pb decay

$$\frac{d \text{Pb}^{206}}{dt} = \lambda \text{N}^{238}(t) \text{N}^{238}(t); \frac{d \text{N}^{238}}{dt} = \lambda \text{N}^{235}(t); \frac{d \text{N}^{235}}{dt} = \lambda \text{N}^{232}(t)$$

The solution for this equation is

$$\text{Pb}^{206}(t) - \text{Pb}^{206}(o) = \text{N}^{238}(o) \left(1 - e^{-\lambda t}\right)$$

Similarly for Pb$^{207}$ the solution is:

$$\text{Pb}^{207}(t) - \text{Pb}^{207}(o) = \text{N}^{235}(o) \left(1 - e^{-\lambda t}\right)$$
and for $\text{Pb}^{208}$ the solution is

$$\text{Pb}^{208}(t) - \text{Pb}^{208}(o) = N^{232}(o)\left(1 - e^{-\lambda^{232} t}\right).$$

If we define the following

$$R_1 = U^{238}(o)/U^{235}(o),$$

$$R_2 = U^{238}(o)/\text{Th}^{232}(o),$$

$$R_3 = U^{235}(o)/\text{Th}^{232}(o) = R_2/R_1$$

and assume that $t = 4.5 \times 10^9$ years.

Then

$$\frac{\text{Pb}^{206}(t) - \text{Pb}^{206}(o)}{\text{Pb}^{207}(t) - \text{Pb}^{207}(o)} = 0.5055 R_1,$$

$$\frac{\text{Pb}^{206}(t) - \text{Pb}^{206}(o)}{\text{Pb}^{208}(t) - \text{Pb}^{208}(o)} = 2.5157 R_2,$$

$$\frac{\text{Pb}^{207}(t) - \text{Pb}^{207}(o)}{\text{Pb}^{208}(t) - \text{Pb}^{208}(o)} = 4.9763 R_3$$

By trial and error one finds $R_1 = 1$

$R_3 = R_2 = 0.176149$ and $\text{Pb}^{206}(o) = 0.0277$

$\text{Pb}^{207}(o) = 0.0$ and $\text{Pb}^{208}(o) = 0.0277$

This suggests that by measuring in coal isotopic abundance of $U^{238}$, $U^{235}$ and $\text{Th}^{232}$ as well as $\text{Pb}^{206}$, $\text{Pb}^{207}$ and $\text{Pb}^{208}$, it should be possible to date when these substances came together as coal.
**TABLE 2. RADIOACTIVE DECAY SCHEME OF URANIUM-238 (1)**

<table>
<thead>
<tr>
<th>Element</th>
<th>Decay Pathway</th>
<th>Decay Mode</th>
<th>Half-Life (y)</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-238</td>
<td>α</td>
<td>4.465</td>
<td></td>
</tr>
<tr>
<td>U-234</td>
<td>α</td>
<td>3.77</td>
<td></td>
</tr>
<tr>
<td>Pa-234</td>
<td>α, β</td>
<td>2.27</td>
<td></td>
</tr>
<tr>
<td>Th-234</td>
<td>α</td>
<td>9.39</td>
<td></td>
</tr>
<tr>
<td>Th-230</td>
<td>α</td>
<td>1.39</td>
<td></td>
</tr>
<tr>
<td>Ra-226</td>
<td>α</td>
<td>6.65</td>
<td></td>
</tr>
<tr>
<td>Ra-222</td>
<td>α</td>
<td>3.82</td>
<td></td>
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<tr>
<td>Rn-221</td>
<td>α</td>
<td>1.00</td>
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</tr>
<tr>
<td>At-218</td>
<td>α</td>
<td>27.4</td>
<td></td>
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<tr>
<td>Po-218</td>
<td>α</td>
<td>3.09</td>
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<tr>
<td>Bi-214</td>
<td>α</td>
<td>5.56</td>
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<tr>
<td>Pb-214</td>
<td>α</td>
<td>20.8</td>
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<td>Tl-208</td>
<td>α</td>
<td>2.04</td>
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*For this reference see the list at the end of Additional Data Section.*
TABLE 3. RADIOACTIVE DECAY SCHEME OF URANIUM-235 (1)*

<table>
<thead>
<tr>
<th>Element</th>
<th>Decay Scheme</th>
<th>Half-Life [s]</th>
<th>Activity [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>( ^{235}U )</td>
<td>( ^{235}U \to ^{231}Pa ) (Protactinium)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( ^{231}Pa )</td>
<td>( ^{231}Pa \to ^{227}Ac ) (Actinium)</td>
<td>( 1.1 \times 10^{-5} ) s</td>
<td>99.67%</td>
</tr>
<tr>
<td>( ^{227}Ac )</td>
<td>( ^{227}Ac \to ^{223}Ra ) (Radium)</td>
<td>( 1.2 \times 10^{-5} ) s</td>
<td>99.93%</td>
</tr>
<tr>
<td>( ^{223}Ra )</td>
<td>( ^{223}Ra \to ^{219}Fr ) (Francium)</td>
<td>( 1.2 \times 10^{-5} ) s</td>
<td>98.8%</td>
</tr>
<tr>
<td>( ^{219}Fr )</td>
<td>( ^{219}Fr \to ^{215}At ) (Actinium)</td>
<td>( 1.1 \times 10^{-5} ) s</td>
<td>99.99%</td>
</tr>
<tr>
<td>( ^{215}At )</td>
<td>( ^{215}At \to ^{211}Bi ) (Bismuth)</td>
<td>( 1.1 \times 10^{-5} ) s</td>
<td>98.8%</td>
</tr>
<tr>
<td>( ^{211}Bi )</td>
<td>( ^{211}Bi \to ^{211}Po ) (Polonium)</td>
<td>( 1.1 \times 10^{-5} ) s</td>
<td>99.99%</td>
</tr>
<tr>
<td>( ^{211}Po )</td>
<td>( ^{211}Po \to ^{211}Pb ) (Bismuth)</td>
<td>( 1.1 \times 10^{-5} ) s</td>
<td>99.99%</td>
</tr>
<tr>
<td>( ^{211}Pb )</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>( ^{211}Pb \to ^{211}Bi ) (Bismuth)</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>( ^{211}Bi )</td>
<td></td>
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<tr>
<td>( ^{211}Bi \to ^{211}Po ) (Polonium)</td>
<td></td>
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<tr>
<td>( ^{211}Po )</td>
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<td></td>
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<tr>
<td>( ^{211}Po \to ^{211}Pb ) (Bismuth)</td>
<td></td>
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<tr>
<td>( ^{211}Pb )</td>
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<tr>
<td>( ^{211}Pb \to ^{211}Bi ) (Bismuth)</td>
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<td>( ^{211}Bi )</td>
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<tr>
<td>( ^{211}Bi \to ^{211}Po ) (Polonium)</td>
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<td>( ^{211}Po \to ^{211}Pb ) (Bismuth)</td>
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<td>( ^{211}Po \to ^{211}Pb ) (Bismuth)</td>
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<td>( ^{211}Pb )</td>
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<tr>
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<tr>
<td>( ^{211}Bi )</td>
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<tr>
<td>( ^{211}Bi \to ^{211}Po ) (Polonium)</td>
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<td></td>
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<tr>
<td>( ^{211}Po )</td>
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</table>
**TABLE 4. RADIOACTIVE DECAY SCHEME OF THORIUM-232 (1)***

<table>
<thead>
<tr>
<th>Th</th>
<th>Th$^{222}$, Th (thorium)</th>
<th>Th$^{229}$, RdTh (radiothorium)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ac</td>
<td>2</td>
<td>Ac$^{223}$, MsTh$_1$ (mesothorium 1)</td>
</tr>
<tr>
<td>Ra</td>
<td>3</td>
<td>Ra$^{226}$, ThX (thorium X)</td>
</tr>
<tr>
<td>Fr</td>
<td>2</td>
<td></td>
</tr>
<tr>
<td>Rn</td>
<td>2</td>
<td>Rn$^{222}$, Th (thorium)</td>
</tr>
<tr>
<td>At</td>
<td>2</td>
<td>At$^{218}$</td>
</tr>
<tr>
<td>Po</td>
<td>2</td>
<td>Po$^{214}$, ThA (thorium A)</td>
</tr>
<tr>
<td>Br</td>
<td>2</td>
<td>Br$^{212}$, ThC (thorium C)</td>
</tr>
<tr>
<td>Pb</td>
<td>2</td>
<td>Pb$^{214}$, ThB (thorium B)</td>
</tr>
<tr>
<td>Tl</td>
<td>2</td>
<td>Tl$^{205}$, ThC$^<em>$ (thorium C$^</em>$)</td>
</tr>
</tbody>
</table>

*For this reference see the list at the end of Additional Data Section.*
REFERENCES


28. W.A. Ver Wiebe, 1950, North American and Middle East Oil Fields, Wichita, Kansas (Published by the author), 259 p.


31. R.D. Dallmeyer, 1979, $^{40}$Ar/$^{39}$Ar Dating: Principles, Techniques, and Applications in Orgogenic Terranes, pp. 77-104 in Lectures in Isotope Geology, E. Jager and J.C. Hunziker (editors), Berlin, Springer-Verlag.


ADDITIONAL DATA
Table 1. Long Lived Radioactive Nuclides

<table>
<thead>
<tr>
<th>Parent</th>
<th>abundance (%)</th>
<th>Daughter</th>
<th>abundance (%)</th>
<th>Half-Life (yr)</th>
<th>Type of Decay</th>
</tr>
</thead>
<tbody>
<tr>
<td>Potassium-40</td>
<td>0.01181</td>
<td>Argon-40</td>
<td>99.6</td>
<td>1.3 x 10^9</td>
<td>E.C.**</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Calcium-40</td>
<td>99.97</td>
<td>(total)</td>
<td>Beta</td>
</tr>
<tr>
<td>Vanadium-50</td>
<td>0.24</td>
<td>Titanium-50</td>
<td>5.34</td>
<td>~6 x 10^15</td>
<td>E.C.</td>
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<tr>
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<td></td>
<td>Chromium-50</td>
<td>4.35</td>
<td>(total)</td>
<td>Beta</td>
</tr>
<tr>
<td>Ruodium-87</td>
<td>27.85</td>
<td>Strontium-87</td>
<td>7.02</td>
<td>4.7 x 10^10</td>
<td>Beta</td>
</tr>
<tr>
<td>Indium-115</td>
<td>95.72</td>
<td>Tin-115</td>
<td>0.35</td>
<td>5 x 10^14</td>
<td>Beta</td>
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<tr>
<td>Tellerium-123</td>
<td>0.87</td>
<td>Antimony-123</td>
<td>42.75</td>
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</tr>
<tr>
<td>Lanthanum-138</td>
<td>0.0395</td>
<td>Barium-138</td>
<td>71.66</td>
<td>1.1 x 10^11</td>
<td>E.C.</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Cerium-138</td>
<td>0.25</td>
<td>(total)</td>
<td>Beta</td>
</tr>
<tr>
<td>Neodymium-144</td>
<td>23.85</td>
<td>Cerium-140</td>
<td>88.48</td>
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<td>Alpha</td>
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<tr>
<td>Samarium-147</td>
<td>14.9</td>
<td>Neodymium-153</td>
<td>12.17</td>
<td>1.06 x 10^11</td>
<td>Alpha</td>
</tr>
<tr>
<td>Hafnium-174</td>
<td>0.18</td>
<td>Ytterbium-170</td>
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<td>4.3 x 10^15</td>
<td>Alpha</td>
</tr>
<tr>
<td>Lutetium-176</td>
<td>2.5</td>
<td>Hafnium-176</td>
<td>5.2</td>
<td>2.2 x 10^10</td>
<td>Beta</td>
</tr>
<tr>
<td>Rhenium-187</td>
<td>0.293</td>
<td>Osmium-187</td>
<td>1.64</td>
<td>4 x 10^10</td>
<td>Beta</td>
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<tr>
<td>Platinum-190</td>
<td>0.0127</td>
<td>Osmium-186</td>
<td>1.59</td>
<td>7 x 10^11</td>
<td>Alpha</td>
</tr>
<tr>
<td>Thorium-232</td>
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<td>Lead-208</td>
<td>52.3</td>
<td>1.4 x 10^10</td>
<td>6 alpha + 4 Be</td>
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<tr>
<td>Uranium-235</td>
<td>0.7205</td>
<td>Lead-207</td>
<td>22.6</td>
<td>7.13 x 10^8</td>
<td>7 alpha + 4 Be</td>
</tr>
<tr>
<td>Uranium-238</td>
<td>99.274</td>
<td>Lead-206</td>
<td>23.6</td>
<td>4.51 x 10^9</td>
<td>8 alpha + 6 Be</td>
</tr>
</tbody>
</table>

* Reference (2)

** Reference (3)

*** E.C electron capture
Table 2
RUBIDIUM CONTENT OF PLANT FOODSTUFFS AND FODDERS (PPM DW) [4],[5],[6]

<table>
<thead>
<tr>
<th>Plant</th>
<th>Tissue samples</th>
<th>Mean content</th>
</tr>
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<td>Cereal</td>
<td>Grains</td>
<td>4</td>
</tr>
<tr>
<td>Corn</td>
<td>Grains</td>
<td>3</td>
</tr>
<tr>
<td>Onion</td>
<td>Bulbs</td>
<td>1</td>
</tr>
<tr>
<td>Lettuce</td>
<td>Leaves</td>
<td>14</td>
</tr>
<tr>
<td>Cabbage</td>
<td>Leaves</td>
<td>12</td>
</tr>
<tr>
<td>Bean</td>
<td>Pods</td>
<td>51</td>
</tr>
<tr>
<td>Soybean</td>
<td>Seeds</td>
<td>220</td>
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<tr>
<td>Apple</td>
<td>Fruits</td>
<td>50</td>
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<tr>
<td>Avocado</td>
<td>Fruits</td>
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<td>Clover</td>
<td>Tops</td>
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<td>Lucerne (alfalfa)</td>
<td>Tops</td>
<td>98</td>
</tr>
<tr>
<td>Grass</td>
<td>Tops</td>
<td>130</td>
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</table>

Table 3
MEAN LEVELS AND RANGES OF STRONTIUM IN FOOD AND FEED PLANTS (PPM DW) [4],[7],[8],[9],[10],[11]

<table>
<thead>
<tr>
<th>Plant</th>
<th>Tissue sample</th>
<th>Range</th>
<th>Mean</th>
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<tbody>
<tr>
<td>Wheat</td>
<td>Grains</td>
<td>0.48—2.3</td>
<td>1.5</td>
</tr>
<tr>
<td>Oats</td>
<td>Grains</td>
<td>1.8—3.2</td>
<td>2.5</td>
</tr>
<tr>
<td>Green tops</td>
<td>9—31</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>Corn</td>
<td>Grains</td>
<td>0.06—0.4</td>
<td>—</td>
</tr>
<tr>
<td>Lettuce</td>
<td>Leaves</td>
<td>—</td>
<td>74</td>
</tr>
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<td>Spinach</td>
<td>Leaves</td>
<td>45—70</td>
<td>—</td>
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<td>Leaves</td>
<td>1.2—150</td>
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<tr>
<td>Bean</td>
<td>Pods</td>
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<tr>
<td>Soybean</td>
<td>Leaves</td>
<td>58—89</td>
<td>—</td>
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<tr>
<td>Carrot</td>
<td>Roots</td>
<td>1.5—131</td>
<td>25</td>
</tr>
<tr>
<td>Onion</td>
<td>Bulbs</td>
<td>10—88</td>
<td>50</td>
</tr>
<tr>
<td>Potato</td>
<td>Tubers</td>
<td>—</td>
<td>2.6</td>
</tr>
<tr>
<td>Tomato</td>
<td>Fruits</td>
<td>0.4—91</td>
<td>9</td>
</tr>
<tr>
<td>Apple</td>
<td>Fruits</td>
<td>0.5—1.7</td>
<td>0.9</td>
</tr>
<tr>
<td>Orange</td>
<td>Fruits</td>
<td>—</td>
<td>0.5</td>
</tr>
<tr>
<td>Clover</td>
<td>Tops</td>
<td>95—850</td>
<td>219</td>
</tr>
<tr>
<td>Lucerne (alfalfa)</td>
<td>Tops</td>
<td>50—1500</td>
<td>662</td>
</tr>
<tr>
<td>Grass</td>
<td>Tops</td>
<td>6—37</td>
<td>24</td>
</tr>
</tbody>
</table>
### Table 4
**RUBIDIUM CONTENT OF SURFACE SOILS OF THE U.S. (PPM DW)** [12]

<table>
<thead>
<tr>
<th>Soil</th>
<th>Range</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sandy soils and lithosols on sandstones</td>
<td>&lt;20—120</td>
<td>50</td>
</tr>
<tr>
<td>Light loamy soils</td>
<td>30—100</td>
<td>60</td>
</tr>
<tr>
<td>Loess and soils on silt deposits</td>
<td>45—100</td>
<td>75</td>
</tr>
<tr>
<td>Clay and clay-loamy soils</td>
<td>45—120</td>
<td>80</td>
</tr>
<tr>
<td>Alluvial soils</td>
<td>55—140</td>
<td>100</td>
</tr>
<tr>
<td>Soils over granites and gneisses</td>
<td>&lt;20—210</td>
<td>120</td>
</tr>
<tr>
<td>Soils over volcanic rocks</td>
<td>20—115</td>
<td>65</td>
</tr>
<tr>
<td>Soils over limestones and calcareous rocks</td>
<td>50—100</td>
<td>75</td>
</tr>
<tr>
<td>Soils on glacial till and drift</td>
<td>30—80</td>
<td>60</td>
</tr>
<tr>
<td>Light desert soils</td>
<td>70—120</td>
<td>95</td>
</tr>
<tr>
<td>Silty prairie soils</td>
<td>50—100</td>
<td>65</td>
</tr>
<tr>
<td>Chernozems and dark prairie soils</td>
<td>55—115</td>
<td>80</td>
</tr>
<tr>
<td>Organic light soils</td>
<td>&lt;20—70</td>
<td>30</td>
</tr>
<tr>
<td>Forest soils</td>
<td>&lt;20—120</td>
<td>55</td>
</tr>
</tbody>
</table>

### Table 5
**STRONTIUM CONTENT OF SURFACE SOILS OF THE U.S. (PPM DW)** [12]

<table>
<thead>
<tr>
<th>Soil</th>
<th>Range</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sandy soils and lithosols on sandstones</td>
<td>5—1000</td>
<td>125</td>
</tr>
<tr>
<td>Light loamy soils</td>
<td>10—500</td>
<td>175</td>
</tr>
<tr>
<td>Loess and soils on silt deposits</td>
<td>20—1000</td>
<td>305</td>
</tr>
<tr>
<td>Clay and clay-loamy soils</td>
<td>15—300</td>
<td>120</td>
</tr>
<tr>
<td>Alluvial soils</td>
<td>50—700</td>
<td>295</td>
</tr>
<tr>
<td>Soils over granites and gneisses</td>
<td>50—1000</td>
<td>420</td>
</tr>
<tr>
<td>Soils over volcanic rocks</td>
<td>50—1000</td>
<td>445</td>
</tr>
<tr>
<td>Soils over limestones and calcareous rocks</td>
<td>15—1000</td>
<td>195</td>
</tr>
<tr>
<td>Soils on glacial till and drift</td>
<td>100—300</td>
<td>190</td>
</tr>
<tr>
<td>Light desert soils</td>
<td>70—2000</td>
<td>490</td>
</tr>
<tr>
<td>Silty prairie soils</td>
<td>70—500</td>
<td>215</td>
</tr>
<tr>
<td>Chernozems and dark prairie soils</td>
<td>70—500</td>
<td>170</td>
</tr>
<tr>
<td>Organic light soils</td>
<td>5—300</td>
<td>110</td>
</tr>
<tr>
<td>Forest soils</td>
<td>20—500</td>
<td>150</td>
</tr>
<tr>
<td>Various soils</td>
<td>7—1000</td>
<td>200</td>
</tr>
</tbody>
</table>
### Table 6
STRONTIUM CONTENT OF SURFACE SOIL OF DIFFERENT COUNTRIES (PPM DW) [13]

<table>
<thead>
<tr>
<th>Soil</th>
<th>Country</th>
<th>Range</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Podzols and sandy soils</td>
<td>Australia</td>
<td>—</td>
<td>118</td>
</tr>
<tr>
<td></td>
<td>New Zealand</td>
<td>350—570‡</td>
<td>—</td>
</tr>
<tr>
<td>Loess and silty soils</td>
<td>New Zealand</td>
<td>220—380‡</td>
<td>—</td>
</tr>
<tr>
<td>Loamy and clay soils</td>
<td>New Zealand</td>
<td>18—86‡</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>U.S.S.R.</td>
<td>280—310</td>
<td>295</td>
</tr>
<tr>
<td>Soils on glacial till</td>
<td>Denmark</td>
<td>—</td>
<td>14.7</td>
</tr>
<tr>
<td>Kastanozems</td>
<td>U.S.S.R.</td>
<td>—</td>
<td>280</td>
</tr>
<tr>
<td>Chernozems</td>
<td>U.S.S.R.</td>
<td>520—3500</td>
<td>—</td>
</tr>
<tr>
<td>Prairien and meadow soils</td>
<td>U.S.S.R.</td>
<td>150—500</td>
<td>300</td>
</tr>
<tr>
<td>Histosols and other organic soils</td>
<td>Denmark</td>
<td>—</td>
<td>92</td>
</tr>
<tr>
<td>Forest soils</td>
<td>U.S.S.R.</td>
<td>—</td>
<td>675</td>
</tr>
<tr>
<td>Various soils</td>
<td>Canada</td>
<td>30—500‡</td>
<td>210</td>
</tr>
<tr>
<td></td>
<td>Denmark</td>
<td>—</td>
<td>17.2</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>—</td>
<td>264</td>
</tr>
</tbody>
</table>

* Soil derived from basalts and andesites.
* Data for whole soil profiles.

### Table 7
INTERACTIONS BETWEEN MAJOR ELEMENTS AND TRACE ELEMENTS IN PLANTS[4], [14], [15],[16]

<table>
<thead>
<tr>
<th>Major element</th>
<th>Antagonistic elements</th>
<th>Synergistic elements</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>Al, B, Ba, Be, Cd, Co, Cr, Cs, Cu, F, Fe, Li, Mn, Ni, Pb, Sr, and Zn</td>
<td>Cu, Mn, and Zn</td>
</tr>
<tr>
<td>Mg</td>
<td>Al, Be, Ba, Cr, Mn, F, Zn, Ni, Co, Cu, and Fe†</td>
<td>Al and Zn</td>
</tr>
<tr>
<td>P</td>
<td>Al, As, B, Be, Cd, Cr, Cu, F, Fe, Hg, Mo, Mn, Ni, Pb, Rh, Se, Si, Sr, and Zn</td>
<td>Al, B, Cu, F, Fe, Mo, Mn, and Zn</td>
</tr>
<tr>
<td>K</td>
<td>Al, B, Hg, Cd, Cr, F, Mo, Mn, and Rb</td>
<td>—</td>
</tr>
<tr>
<td>S</td>
<td>As, Ba, Fe, Mo, Pb, and Se</td>
<td>Pb and Fe</td>
</tr>
<tr>
<td>N</td>
<td>B, F, and Cu</td>
<td>B, Cu, Fe, and Mo</td>
</tr>
<tr>
<td>Cl</td>
<td>Br and I</td>
<td>—</td>
</tr>
</tbody>
</table>

* Reported for microorganisms.
* Mutual pollution causes significant injury.
### Table 8
CONCENTRATION OF RARE EARTH ELEMENTS IN SOME U.S. COALS [17], [18]

<table>
<thead>
<tr>
<th>Element</th>
<th>Range (ppm)</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
<th>Range (ppm)</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
<th>Range (ppm)</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cerium</td>
<td>4.4—46</td>
<td>14</td>
<td>12</td>
<td>11—42</td>
<td>25</td>
<td>23</td>
<td>2.8—30</td>
<td>11</td>
<td>9.1</td>
</tr>
<tr>
<td>Samarium</td>
<td>0.4—3.8</td>
<td>1.2</td>
<td>1.1</td>
<td>0.87—4.3</td>
<td>2.6</td>
<td>2.4</td>
<td>0.22—1.4</td>
<td>0.61</td>
<td>0.56</td>
</tr>
<tr>
<td>Europium</td>
<td>0.1—0.87</td>
<td>0.26</td>
<td>0.25</td>
<td>0.16—0.92</td>
<td>0.52</td>
<td>0.47</td>
<td>0.07—0.60</td>
<td>0.20</td>
<td>0.16</td>
</tr>
<tr>
<td>Terbium</td>
<td>0.04—0.65</td>
<td>0.22</td>
<td>0.18</td>
<td>0.06—0.63</td>
<td>0.34</td>
<td>0.28</td>
<td>0.06—0.58</td>
<td>0.21</td>
<td>0.17</td>
</tr>
<tr>
<td>Dysprosium</td>
<td>0.5—3.3</td>
<td>1.1</td>
<td>1.0</td>
<td>0.74—3.5</td>
<td>2.3</td>
<td>2.0</td>
<td>0.22—1.4</td>
<td>0.63</td>
<td>0.57</td>
</tr>
<tr>
<td>Ytterbium</td>
<td>0.27—0.15</td>
<td>0.56</td>
<td>0.53</td>
<td>0.18—1.4</td>
<td>0.83</td>
<td>0.73</td>
<td>0.13—0.78</td>
<td>0.38</td>
<td>0.34</td>
</tr>
<tr>
<td>Lutetium</td>
<td>0.02—0.44</td>
<td>0.09</td>
<td>0.08</td>
<td>0.04—0.40</td>
<td>0.22</td>
<td>0.18</td>
<td>0.01—0.43</td>
<td>0.07</td>
<td>0.05</td>
</tr>
</tbody>
</table>

### Table 9
CONCENTRATION OF RARE EARTH ELEMENTS IN COAL (PPM) [17], [18]

<table>
<thead>
<tr>
<th>Element</th>
<th>U.S. subbituminous</th>
<th>U.S. lignite</th>
<th>U.S. av</th>
<th>Worldwide av</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cerium (Ce)</td>
<td>5.5</td>
<td>12.3</td>
<td>7.7</td>
<td>11.5</td>
</tr>
<tr>
<td>Praseodymium (Pr)</td>
<td>6.1</td>
<td>2.7</td>
<td>2.7</td>
<td>2.2</td>
</tr>
<tr>
<td>Neodymium (Nd)</td>
<td>50</td>
<td>11</td>
<td>37</td>
<td>4.7</td>
</tr>
<tr>
<td>Samarium (Sm)</td>
<td>0.50</td>
<td>0.27</td>
<td>0.42</td>
<td>1.6</td>
</tr>
<tr>
<td>Europium (Eu)</td>
<td>0.61</td>
<td>0.13</td>
<td>0.45</td>
<td>0.7</td>
</tr>
<tr>
<td>Gadolinium (Gd)</td>
<td>0.13</td>
<td>0.21</td>
<td>0.17</td>
<td>1.6</td>
</tr>
<tr>
<td>Terbium (Tb)</td>
<td>0.1</td>
<td>0.1</td>
<td>0.1</td>
<td>0.3</td>
</tr>
<tr>
<td>Dysprosium (Dy)</td>
<td>2.7</td>
<td>1.4</td>
<td>2.2</td>
<td>—</td>
</tr>
<tr>
<td>Holmium (Ho)</td>
<td>0.13</td>
<td>0.06</td>
<td>0.11</td>
<td>0.3</td>
</tr>
<tr>
<td>Erbium (Er)</td>
<td>0.46</td>
<td>0.16</td>
<td>0.34</td>
<td>0.6</td>
</tr>
<tr>
<td>Thulium (Tm)</td>
<td>0.07</td>
<td>0.07</td>
<td>0.07</td>
<td>—</td>
</tr>
<tr>
<td>Ytterbium (Yb)</td>
<td>0.5</td>
<td>1.5</td>
<td>1</td>
<td>0.5</td>
</tr>
<tr>
<td>Lutetium (Lu)</td>
<td>0.09</td>
<td>0.05</td>
<td>0.08</td>
<td>0.07</td>
</tr>
</tbody>
</table>
## TABLE 10

**URANIUM–RADIIUM (4n+2) SERIES (17)**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Type of decay and particle energies (MeV) (%)</th>
<th>Gamma energies — internal conversion (IC)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>238U</strong></td>
<td>4.51 × 10⁹ years</td>
<td>α: 4.2 (100)</td>
<td>γ: 0.048 (0% + 23% IC)</td>
</tr>
<tr>
<td><strong>234Th</strong></td>
<td>24.1 days</td>
<td>β⁺: 0.19 (65), 0.10 (35)</td>
<td>γ: 0.091 (α, IC = 2.5), 0.063 (α, IC = 0.2), 0.029 (α, IC = 10)</td>
</tr>
<tr>
<td><strong>Protactinium-234</strong></td>
<td>1.18 min</td>
<td>β⁺: 2.31 (~90), 1.50 (~9), 0.58 (~1)</td>
<td>γ: 1.00, 0.75, others</td>
</tr>
<tr>
<td><strong>Protactinium-234</strong></td>
<td>6.66 hr</td>
<td>β⁺: 1.13 (13), 0.53 (27), 0.32 (32), 0.16 (28)</td>
<td>γ: 1.68, 1.43, 1.24, 0.924, 0.877, 0.803, 0.732, 0.603, 0.566, 0.368, 0.333, 0.293, 0.225, 0.153, 0.099, 0.043</td>
</tr>
<tr>
<td><strong>238U</strong></td>
<td>2.5 × 10⁴ years</td>
<td>α: 4.768 (72), 4.717 (28)</td>
<td>γ: 0.051 0% + 28% IC</td>
</tr>
<tr>
<td><strong>230Th</strong></td>
<td>8 × 10⁴ years</td>
<td>α: 4.682 (76)</td>
<td>γ: 0.067 (0% + 24% IC)</td>
</tr>
<tr>
<td><strong>236Ra</strong></td>
<td>1620 years</td>
<td>α: 4.777 (94.3)</td>
<td>γ: 0.188 (~4% + ~2% IC)</td>
</tr>
<tr>
<td><strong>222Rn</strong></td>
<td>3.825 days</td>
<td>α: 5.48 (~100)</td>
<td></td>
</tr>
<tr>
<td><strong>231Po</strong></td>
<td>3.05 min</td>
<td>α: 6.00 (~100)</td>
<td></td>
</tr>
<tr>
<td><strong>218At</strong></td>
<td>1.3 sec</td>
<td>α: 6.70 (~0.02), 6.65 (~0.001)</td>
<td></td>
</tr>
<tr>
<td><strong>218Rn</strong></td>
<td>1.9 × 10⁻² sec</td>
<td>α: 7.13 (very weak)</td>
<td>γ: 0.61 (very weak)</td>
</tr>
<tr>
<td><strong>214Pb</strong></td>
<td>26.8 min</td>
<td>β⁺: 0.65 (~44), 0.59 (~56)</td>
<td>γ: 0.35, 0.30, 0.24</td>
</tr>
<tr>
<td><strong>214Bi</strong></td>
<td>19.9 min</td>
<td>α: ~5.5 (0.04), β⁺: 3.26 (~19), 1.88 (9), 1.51 (40), 1.0 (23), 0.4 (9)</td>
<td>1.76⁺, 1.12⁺, 0.61⁺, others up to 2.43</td>
</tr>
<tr>
<td><strong>214Po</strong></td>
<td>1.6 × 10⁻⁴ sec</td>
<td>α: 7.68 (~100)</td>
<td></td>
</tr>
<tr>
<td><strong>210Ti</strong></td>
<td>1.3 min</td>
<td>β⁺: 0.061 (20), 0.015 (80)</td>
<td>γ: very weak</td>
</tr>
<tr>
<td><strong>210Pb</strong></td>
<td>22y</td>
<td>β⁺: 0.061 (20), 0.015 (80)</td>
<td>γ: 0.0465 (~5% + ~75% IC)</td>
</tr>
<tr>
<td><strong>210Bi</strong></td>
<td>5.0 d</td>
<td>α: 5.06 (1.7 × 10⁻⁴), β⁺: 1.17 (~100)</td>
<td>γ: 0.8 (very weak)</td>
</tr>
<tr>
<td><strong>210Po</strong></td>
<td>138.4 days</td>
<td>α: 5.305 (~100)</td>
<td></td>
</tr>
<tr>
<td><strong>208Ti</strong></td>
<td>4.2 min</td>
<td>β⁺: 1.51 (1.7 × 10⁻⁴)</td>
<td>γ:</td>
</tr>
<tr>
<td><strong>208Pb</strong></td>
<td>Stable</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Most abundant.
### TABLE 11

**THORIUM SERIES (4n) (17)**

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Type of decay and particle energies (MeV) (%)</th>
<th>Gamma energies — internal conversion (IC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{222}$Th</td>
<td>$1.4 \cdot 10^{10}$ years</td>
<td>$\alpha$: 4.007 (76), 3.948 (24), 3.948 (24)</td>
<td>$\gamma$: 0.059</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>6.7 years</td>
<td>$\beta^- \sim 0.04$ (100)</td>
<td></td>
</tr>
<tr>
<td>$^{228}$Ac</td>
<td>6.13 hr</td>
<td>$\beta^-$: 2.10 (~12), 1.76 (~12), 1.18 (~35), others of low energy (41)</td>
<td>$\gamma$: 0.057 to 1.46 (many lines)</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>1.91 years</td>
<td>$\alpha$: 5.421 (71), 5.338 (28), $\sim 5.2$ (0.4)</td>
<td>$\gamma$: 0.084 (2%), others (very weak)</td>
</tr>
<tr>
<td>$^{228}$Ra</td>
<td>3.64 days</td>
<td>$\alpha$: 5.681 (95), 5.445 (4.9)</td>
<td>$\gamma$: 0.241 (3.2% + 0.5% (IC))</td>
</tr>
<tr>
<td>$^{228}$Rn</td>
<td>51.5 sec</td>
<td>$\alpha$: 6.28 (~100)</td>
<td></td>
</tr>
<tr>
<td>$^{228}$Po</td>
<td>0.16 sec</td>
<td>$\alpha$: 6.775 (~100), $\beta$: 0.04</td>
<td></td>
</tr>
<tr>
<td>$^{226}$At</td>
<td>$3.10^{-4}$ sec</td>
<td>$\alpha$: 7.79 (0.04), $\beta$: 0.57 (~12), 0.33 (~80), others of low energy (8)</td>
<td>$\gamma$: 0.30 (~3% + 1% IC), 0.24 (~36% + 36% IC), 0.12 (~0% + 3% IC)</td>
</tr>
<tr>
<td>$^{214}$Pb</td>
<td>10.6 hr</td>
<td>$\alpha$: 6.08 (10), 6.04 (25), others (1)</td>
<td>$\gamma$: 1.62, 1.08, 0.79, 0.73 (together $\sim 10%$), 0.04 (0% + ~25% IC)</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>60.5 min</td>
<td>$\alpha$:</td>
<td></td>
</tr>
<tr>
<td>$^{214}$Po</td>
<td>$3 \cdot 10^{-7}$ sec</td>
<td>$\alpha$: 8.78 (~64), others of lower energy (5)</td>
<td></td>
</tr>
<tr>
<td>$^{212}$Po</td>
<td>39.8 sec</td>
<td>$\alpha$: 8.78 (~64), others of higher energy (very weak)</td>
<td></td>
</tr>
<tr>
<td>$^{208}$Tl</td>
<td>3.1 min</td>
<td>$\beta$: 1.79 (19), 1.52 (7), 1.25 (9), 1.03 (1)</td>
<td>$\gamma$: 2.62 (36%), 0.86 (4%), 0.58 (31%), 0.51 (~8% + ~1% IC), 0.28 (~3% + ~1% IC)</td>
</tr>
<tr>
<td>$^{208}$Pb</td>
<td>Stable</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
# TABLE 12

## URANIUM–ACTINIUM SERIES (4n+3) (17)

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>Type of decay and particle energies (MeV) (%)</th>
<th>Gamma energies — internal conversion (IC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U</td>
<td>$7.1 \cdot 10^8$ years</td>
<td>$\alpha$: 4.56 (7), 4.52 (4), 4.35 (84), 4.18 (6)</td>
<td>$\gamma$: ~0.2000 (&gt;4%), 0.185 (55%), ~0.165, (&gt;4%), 0.143 (12%), 0.110 (5%), 0.095 (9%)</td>
</tr>
<tr>
<td>$^{232}$Th</td>
<td>25.6 hr</td>
<td>$\beta^-$: 0.30, others $\alpha$: 5.046 (10), 5.017 (23), 5.001 (24), 4.938 (22), 4.722 (11), others</td>
<td>$\gamma$: 0.084, others $\gamma$: 0.29, 0.027, many others</td>
</tr>
<tr>
<td>Protactinium-231</td>
<td>$3.4 \cdot 10^8$ years</td>
<td>$\alpha$: 4.94 (1.2) $\beta^-$: 0.046 (99)</td>
<td></td>
</tr>
<tr>
<td>$^{232}$Ac</td>
<td>22 years</td>
<td>$\alpha$: 6.036 (23), 5.976 (24), 5.958 (3.5), 5.865 (3), 5.755 (21), 5.712 (5), 5.708 (8.7), 5.699 (4), others ($\sim 7.8$)</td>
<td>$\gamma$: 0.24 ($\sim 10%$), 0.05 ($\sim 7.5%$), many others of low energy (weak), all highly converted</td>
</tr>
<tr>
<td>$^{231}$Th</td>
<td>18.2 days</td>
<td>$\alpha$: 5.34 (very weak) $\beta^-$: 1.15 ($\sim 1%$)</td>
<td>$\gamma$: 0.31, 0.21, 0.08</td>
</tr>
<tr>
<td>$^{231}$Fr</td>
<td>22 min</td>
<td>$\alpha$: 7.360 ($\sim 100$)</td>
<td>$\gamma$: 0.34 (2.8%), 0.32 (2.3%), 0.27 (10%), 0.15 (5.5%), 0.14 (4%), 0.12 (2%), all highly converted</td>
</tr>
<tr>
<td>$^{229}$Ra</td>
<td>11.7 days</td>
<td>$\alpha$: 8.00 (very weak) $\beta^-$: (0.0005)</td>
<td>$\gamma$: 0.40 ($\sim 5%$), 0.27 ($\sim 9% + \sim 4%$ IC)</td>
</tr>
<tr>
<td>$^{228}$Rn</td>
<td>3.9 sec</td>
<td>$\alpha$: 6.818 (82), 6.547 (13), 6.419 (5)</td>
<td>$\gamma$: 0.43 (6%), 0.40 (6%)</td>
</tr>
<tr>
<td>$^{218}$Po</td>
<td>$1.8 \cdot 10^{-3}$ sec</td>
<td>$\alpha$: 6.617 ($\sim 83$)</td>
<td>$\gamma$: 0.35 (13% + 4% IC)</td>
</tr>
<tr>
<td>$^{218}$At</td>
<td>$\sim 1 \cdot 10^{-4}$ sec</td>
<td>$\alpha$: 1.39 ($\sim 80$), 0.5 ($\sim 20$)</td>
<td>$\gamma$: 0.83 (13%), 0.43 (6%), 0.40 (6%)</td>
</tr>
<tr>
<td>$^{218}$Pb</td>
<td>36 min</td>
<td>$\beta^-$: 0.3%</td>
<td>$\gamma$: 0.35 (13% + 4% IC)</td>
</tr>
<tr>
<td>$^{214}$Bi</td>
<td>2.16 min</td>
<td>$\alpha$: 6.617 ($\sim 83$)</td>
<td>$\gamma$: 0.35 (13% + 4% IC)</td>
</tr>
<tr>
<td>$^{214}$Po</td>
<td>0.52 sec</td>
<td>$\beta^-$: 0.3%</td>
<td>$\gamma$: 0.35 (13% + 4% IC)</td>
</tr>
<tr>
<td>$^{207}$Tl</td>
<td>4.79 min</td>
<td>$\beta^-$: 1.44 (100)</td>
<td>$\gamma$: 0.89 (weak), 0.57 (weak)</td>
</tr>
<tr>
<td>$^{207}$Pb</td>
<td>Stable</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
### TABLE 13

LEAD IN U.S. COALS (17)

<table>
<thead>
<tr>
<th>Region</th>
<th>Coal</th>
<th>Conc range (ppm)</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Appalachian</td>
<td>Bituminous</td>
<td>0.37—86</td>
<td>9.2</td>
<td>6.6</td>
</tr>
<tr>
<td>Interior province</td>
<td>Bituminous</td>
<td>0.78—590</td>
<td>44</td>
<td>18</td>
</tr>
<tr>
<td>N. Great Plains</td>
<td>Lignite</td>
<td>1.4—17</td>
<td>4.0</td>
<td>3.5</td>
</tr>
<tr>
<td>N. Great Plains</td>
<td>Subbituminous</td>
<td>0.72—58</td>
<td>5.5</td>
<td>4.1</td>
</tr>
<tr>
<td>Rocky Mountain</td>
<td>Subbituminous</td>
<td>0.95—76</td>
<td>8.0</td>
<td>6.0</td>
</tr>
<tr>
<td>Rocky Mountain</td>
<td>Bituminous</td>
<td>0.76—137</td>
<td>7.3</td>
<td>5.2</td>
</tr>
<tr>
<td>All U.S.</td>
<td>Different</td>
<td>0.06—1300</td>
<td>15</td>
<td>7.0</td>
</tr>
<tr>
<td>Illinois Basin</td>
<td>—</td>
<td>0.8—220</td>
<td>32</td>
<td>15</td>
</tr>
<tr>
<td>Eastern U.S.</td>
<td>—</td>
<td>1.0—18</td>
<td>5.9</td>
<td>4.7</td>
</tr>
<tr>
<td>Western U.S.</td>
<td>—</td>
<td>0.70—9.0</td>
<td>3.4</td>
<td>2.6</td>
</tr>
<tr>
<td>North Dakota</td>
<td>Lignite</td>
<td>1.5—8</td>
<td>5.44</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE 14

LEAD CONTENT OF SURFACE SOILS OF THE U.S. (PPM DW) (13)

<table>
<thead>
<tr>
<th>Soil</th>
<th>Range</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sandy soils and lithosols on sandstones</td>
<td>&lt;10—70</td>
<td>17</td>
</tr>
<tr>
<td>Light loamy soils</td>
<td>&lt;10—50</td>
<td>20</td>
</tr>
<tr>
<td>Loess and soils on silt deposits</td>
<td>10—30</td>
<td>19</td>
</tr>
<tr>
<td>Clay and clay loamy soils</td>
<td>10—70</td>
<td>22</td>
</tr>
<tr>
<td>Alluvial soils</td>
<td>10—30</td>
<td>18</td>
</tr>
<tr>
<td>Soils over granites and gneisses</td>
<td>10—50</td>
<td>21</td>
</tr>
<tr>
<td>Soils over volcanic rocks</td>
<td>10—70</td>
<td>20</td>
</tr>
<tr>
<td>Soils over limestones and calcareous rocks</td>
<td>10—50</td>
<td>22</td>
</tr>
<tr>
<td>Soils on glacial till and drift</td>
<td>10—30</td>
<td>17</td>
</tr>
<tr>
<td>Light desert soils</td>
<td>10—70</td>
<td>23</td>
</tr>
<tr>
<td>Silty prairie soils</td>
<td>10—30</td>
<td>21</td>
</tr>
<tr>
<td>Chernozems and dark prairie soils</td>
<td>10—70</td>
<td>19</td>
</tr>
<tr>
<td>Organic light soils</td>
<td>10—50</td>
<td>24</td>
</tr>
<tr>
<td>Forest soils</td>
<td>10—50</td>
<td>20</td>
</tr>
<tr>
<td>Various soils</td>
<td>&lt;10—70</td>
<td>26</td>
</tr>
</tbody>
</table>
### TABLE 15

**LEAD CONTENT OF SURFACE SOILS OF DIFFERENT COUNTRIES (PPM DW) (13)**

<table>
<thead>
<tr>
<th>Soil</th>
<th>Country</th>
<th>Range</th>
<th>Mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>Podzols and sandy soils</td>
<td>Australia</td>
<td>—</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td>Canada</td>
<td>2.3—47.5</td>
<td>10.4</td>
</tr>
<tr>
<td></td>
<td>Madagascar</td>
<td>—</td>
<td>37</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>8.5—23.5</td>
<td>16</td>
</tr>
<tr>
<td>Loess and silty soils</td>
<td>Poland</td>
<td>14—32</td>
<td>26</td>
</tr>
<tr>
<td>Loamy and clay soils</td>
<td>Canada</td>
<td>1.5—50.1</td>
<td>16.6</td>
</tr>
<tr>
<td></td>
<td>Chad</td>
<td>20—45</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Madagascar</td>
<td>—</td>
<td>48</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>12.5—52</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>U.S.S.R.</td>
<td>—</td>
<td>40</td>
</tr>
<tr>
<td>Soils on glacial till</td>
<td>Denmark</td>
<td>11.3—17.3</td>
<td>14.7</td>
</tr>
<tr>
<td>Fluvisols</td>
<td>Austria</td>
<td>16—22</td>
<td>19</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>24—96</td>
<td>63</td>
</tr>
<tr>
<td></td>
<td>Madagascar</td>
<td>19—47</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>12.5—48.5</td>
<td>39</td>
</tr>
<tr>
<td>Paddy soils</td>
<td>Japan</td>
<td>6—189</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>Chad</td>
<td>20—50</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>19.5—48.5</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>U.S.S.R.</td>
<td>—</td>
<td>67</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>17—63</td>
<td>40</td>
</tr>
<tr>
<td>Rendzinas</td>
<td>Ireland</td>
<td>25—45</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Madagascar</td>
<td>—</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>17—46</td>
<td>28.5</td>
</tr>
<tr>
<td>Kastanozems and brown soils</td>
<td>Austria</td>
<td>13—31</td>
<td>21</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>20—50</td>
<td>35</td>
</tr>
<tr>
<td>Ferralsols</td>
<td>Chad</td>
<td>10—30</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Sierra Leone</td>
<td>3—91</td>
<td>47</td>
</tr>
<tr>
<td>Chernozems</td>
<td>Poland</td>
<td>19—29</td>
<td>25</td>
</tr>
<tr>
<td></td>
<td>U.S.S.R.</td>
<td>—</td>
<td>61</td>
</tr>
<tr>
<td></td>
<td>West Germany</td>
<td>11.5—79.5</td>
<td>—</td>
</tr>
<tr>
<td>Histosols, other organic soils</td>
<td>Canada</td>
<td>1.5—50.0</td>
<td>12.6</td>
</tr>
<tr>
<td></td>
<td>Denmark</td>
<td>43—176</td>
<td>50.5</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>26—142</td>
<td>84</td>
</tr>
<tr>
<td></td>
<td>Ireland</td>
<td>120</td>
<td>—</td>
</tr>
<tr>
<td></td>
<td>Poland</td>
<td>18—85</td>
<td>—</td>
</tr>
<tr>
<td>Forest soils</td>
<td>China</td>
<td>—</td>
<td>26</td>
</tr>
<tr>
<td></td>
<td>U.S.S.R.</td>
<td>10—56</td>
<td>37</td>
</tr>
<tr>
<td>Various soils</td>
<td>Austria</td>
<td>21—33</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>Canada</td>
<td>—</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>Great Britain</td>
<td>15.5—41</td>
<td>29</td>
</tr>
<tr>
<td></td>
<td>Japan</td>
<td>5—189</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>West Germany</td>
<td>15—68</td>
<td>—</td>
</tr>
</tbody>
</table>

* Mean for whole profiles of arable soils.
<table>
<thead>
<tr>
<th>Element</th>
<th>Constituent of</th>
<th>Involved in</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al⁺</td>
<td>Phospholipid (in algae)</td>
<td>Controlling colloidal properties in the cell, possible activation of some dehydrogenases and oxidases</td>
</tr>
<tr>
<td>As⁺</td>
<td>Phosphogluconate</td>
<td>Metabolism of carbohydrates in algae and fungi</td>
</tr>
<tr>
<td>B</td>
<td>Bromophenols (in algae)</td>
<td>Metabolism and transport of carbohydrates, flavonoid synthesis, nucleic acid synthesis, phosphate utilization, and polyphenol production</td>
</tr>
<tr>
<td>Br⁻</td>
<td>Cobamide coenzyme</td>
<td>Symbiotic N₂ fixation, possibly also in non-nodulating plants, and valence changes. Stimulation synthesis of chlorophyll and proteins (2)</td>
</tr>
<tr>
<td>Co</td>
<td>Various oxidases, plastocyanins, and ceruloplasmin</td>
<td>Oxidation, photosynthesis, protein and carbohydrate metabolism, possibly involved in symbiotic N₂ fixation, and valence changes</td>
</tr>
<tr>
<td>Cu</td>
<td>Fluoracetate (in a few species)</td>
<td>Citrate conversions</td>
</tr>
<tr>
<td>Fe</td>
<td>Hemo-proteins and nonheme iron proteins, dehydrogenases, and ferredoxins</td>
<td>Photosynthesis, N₂ fixation, and valence changes</td>
</tr>
<tr>
<td>I</td>
<td>Tyrosine and its derivatives (in angiosperms and algae)</td>
<td>—</td>
</tr>
<tr>
<td>Li⁺</td>
<td>Many enzyme systems</td>
<td>Metabolism in halophytes</td>
</tr>
<tr>
<td>Mn</td>
<td>Enzyme systems</td>
<td>Photoproduction of oxygen in chloroplasts and, indirectly, in NO₃ reduction</td>
</tr>
<tr>
<td>Mo</td>
<td>Nitrate reductase, nitrogenase, oxidases, and molybdoferredoxin</td>
<td>N₂ fixation, NO₃⁻ reduction, and valence changes</td>
</tr>
<tr>
<td>Ni</td>
<td>Enzyme urease (in Camalilla seeds)</td>
<td>Possibly in action of hydrogenase and translocation of N</td>
</tr>
<tr>
<td>Rb⁺</td>
<td>—</td>
<td>Function similar to that of K in some plants</td>
</tr>
<tr>
<td>Se⁺</td>
<td>Glycine reductase (in Clostridium cells)</td>
<td>—</td>
</tr>
<tr>
<td>Si</td>
<td>Structural components</td>
<td>—</td>
</tr>
<tr>
<td>Sr⁺</td>
<td>—</td>
<td>Function similar to that of Ca in some plants</td>
</tr>
<tr>
<td>Ti⁺</td>
<td>—</td>
<td>Possibly photosynthesis and N₂ fixation</td>
</tr>
<tr>
<td>V⁺</td>
<td>Porphyrins, hemoproteins</td>
<td>Lipid metabolism, photosynthesis (in green algae), and, possibly, in N₂ fixation</td>
</tr>
<tr>
<td>Zn</td>
<td>Anhydrases, dehydrogenases, proteinases, and peptidases</td>
<td>Carbohydrate and protein metabolism</td>
</tr>
</tbody>
</table>

Elements known to be essential for some groups or species and whose general essentiality needs confirmation.
ADDITIONAL DATA
### TABLE 17

**METALS IN SURFACE SOILS AND EARTHWORMS (PPM) (13)**

<table>
<thead>
<tr>
<th>Metal</th>
<th>Soil</th>
<th>Earthworms</th>
<th>Ratio, worms/soil</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd</td>
<td>2</td>
<td>15</td>
<td>7.5</td>
</tr>
<tr>
<td></td>
<td>4</td>
<td>4</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>1.6</td>
<td>11.1</td>
<td>6.9</td>
</tr>
<tr>
<td></td>
<td>0.9</td>
<td>14.4</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>1.1</td>
<td>18</td>
<td>16</td>
</tr>
<tr>
<td></td>
<td>0.6</td>
<td>12</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>0.1</td>
<td>2.7</td>
<td>27</td>
</tr>
<tr>
<td></td>
<td>4.1</td>
<td>10.3*</td>
<td>27.6</td>
</tr>
<tr>
<td>Cu</td>
<td>20</td>
<td>13</td>
<td>0.65</td>
</tr>
<tr>
<td></td>
<td>252</td>
<td>11</td>
<td>0.04</td>
</tr>
<tr>
<td></td>
<td>335</td>
<td>11</td>
<td>0.03</td>
</tr>
<tr>
<td></td>
<td>52</td>
<td>28</td>
<td>0.53</td>
</tr>
<tr>
<td></td>
<td>26</td>
<td>18</td>
<td>0.69</td>
</tr>
<tr>
<td></td>
<td>9</td>
<td>5</td>
<td>0.55</td>
</tr>
<tr>
<td>Hg</td>
<td>3.8</td>
<td>1.29*</td>
<td>0.33</td>
</tr>
<tr>
<td></td>
<td>0.1</td>
<td>0.04*</td>
<td>0.40</td>
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<tr>
<td>Mn</td>
<td>1330</td>
<td>82</td>
<td>0.06</td>
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<td>226</td>
<td>28</td>
<td>0.12</td>
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<tr>
<td></td>
<td>164</td>
<td>27</td>
<td>0.16</td>
</tr>
<tr>
<td>Ni</td>
<td>26</td>
<td>31</td>
<td>1.19</td>
</tr>
<tr>
<td></td>
<td>18</td>
<td>29</td>
<td>1.61</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>32</td>
<td>2.66</td>
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<tr>
<td>Pb</td>
<td>1314</td>
<td>3592</td>
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<tr>
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<td>629</td>
<td>9</td>
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<tr>
<td></td>
<td>700</td>
<td>331</td>
<td>0.47</td>
</tr>
<tr>
<td></td>
<td>94</td>
<td>101</td>
<td>1.04</td>
</tr>
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<td></td>
<td>170</td>
<td>62</td>
<td>0.36</td>
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<tr>
<td></td>
<td>20</td>
<td>9</td>
<td>0.45</td>
</tr>
<tr>
<td></td>
<td>870</td>
<td>109*</td>
<td>0.12</td>
</tr>
<tr>
<td>Zn</td>
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<td>900</td>
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</tr>
<tr>
<td></td>
<td>81</td>
<td>662*</td>
<td>8.17</td>
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</table>

*Note:* Element concentrations expressed on dry weight basis. Organisms analyzed are *Lumbricus rubellus* or *L. terrestris*, except as indicated.
TABLE 18
THORIUM AND URANIUM IN MAJOR ROCK TYPES (PPM) (VALUES COMMONLY FOUND, BASED ON VARIOUS SOURCES) (13)

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<thead>
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<th>Rock type</th>
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<td></td>
</tr>
<tr>
<td>Ultramafic rocks</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dunites, peridotites, pyroxenites</td>
<td>0.004—0.005</td>
<td>0.003—0.010</td>
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<tr>
<td>Mafic rocks</td>
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<td>0.3—1.0</td>
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<tr>
<td>Basalts, gabbros</td>
<td>7—14</td>
<td>1.4—3.0</td>
</tr>
<tr>
<td>Intermediate rocks</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Diorites, syenites</td>
<td>10—23</td>
<td>2.5—6.0</td>
</tr>
<tr>
<td>Acid rocks (igneous)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Granites, gneisses</td>
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<td></td>
</tr>
<tr>
<td>Acid rocks (volcanic)</td>
<td>15</td>
<td>5</td>
</tr>
<tr>
<td>Rhyolites, trachytes, dacites</td>
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<tr>
<td><strong>Sedimentary rocks</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Argillaceous sediments</td>
<td>9.6—12.0</td>
<td>3—4</td>
</tr>
<tr>
<td>Shales</td>
<td>12</td>
<td>3.0—4.1</td>
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<tr>
<td>Sandstones</td>
<td>1.7—3.8</td>
<td>0.45—0.59</td>
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<tr>
<td>Limestones, dolomites</td>
<td>1.7—2.9</td>
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TABLE 19
THORIUM AND URANIUM CONTENTS OF SURFACE SOILS OF DIFFERENT COUNTRIES (PPM DW) (13)

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<tr>
<th>Country</th>
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<tr>
<td></td>
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<td>Mean</td>
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<td>3.6—17.8</td>
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<tr>
<td>Canada</td>
<td>4.2—14.1</td>
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</tr>
<tr>
<td>Great Britain</td>
<td>—</td>
<td>10.5</td>
</tr>
<tr>
<td>West</td>
<td>0.4—15.0</td>
<td>8.0</td>
</tr>
<tr>
<td>Germany</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>India</td>
<td>1.4—7.2</td>
<td>3.4</td>
</tr>
<tr>
<td>Poland</td>
<td>—</td>
<td>—</td>
</tr>
<tr>
<td>U.S.</td>
<td>2.2—21.0</td>
<td>7.6</td>
</tr>
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### TABLE 20

**URANIUM CONCENTRATIONS IN SOME U.S. COALS (17)**

<table>
<thead>
<tr>
<th>Region</th>
<th>Coal</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
<th>Conc range (ppm)</th>
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<tbody>
<tr>
<td>Appalachian</td>
<td>Bituminous</td>
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<td>1.2</td>
<td>0.10—19</td>
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<td>Interior province</td>
<td>Bituminous</td>
<td>3.2</td>
<td>1.7</td>
<td>0.20—59</td>
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<tr>
<td>N. Great Plains</td>
<td>Lignite</td>
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<td>1.2</td>
<td>0.21—13</td>
</tr>
<tr>
<td>N. Great Plains</td>
<td>Subbituminous</td>
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<td>0.98</td>
<td>0.9—16</td>
</tr>
<tr>
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<td>Subbituminous</td>
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<td>1.9</td>
<td>0.06—76</td>
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<td>Rocky Mountain</td>
<td>Bituminous</td>
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<td>0.13—42</td>
</tr>
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<td>1.3</td>
<td>0.06—2700</td>
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<td>0.31—4.6</td>
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### TABLE 21

**THORIUM CONCENTRATION IN SOME U.S. COALS (17)**

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<th>Area</th>
<th>Conc range (ppm)</th>
<th>Arithmetic mean (ppm)</th>
<th>Geometric mean (ppm)</th>
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<td>Western U.S.</td>
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<td>1.8</td>
</tr>
<tr>
<td>Eastern U.S.</td>
<td>1.8—9.0</td>
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<td>4.0</td>
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<td>---------</td>
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</tr>
<tr>
<td>Pb\textsuperscript{206}(s)</td>
<td>Pb\textsuperscript{207}(s)</td>
<td>Pb\textsuperscript{208}(s)</td>
<td>( \frac{Y\textsuperscript{238}(s)}{Y\textsuperscript{235}(s)} )</td>
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<td>1</td>
</tr>
</tbody>
</table>
\[ U^{238} \rightarrow Pb^{206} \]

\[ \begin{array}{c}
25.1 \% \\
25.2 \% \\
23.6 \%
\end{array} \]

\[ T_{1/2} = 4.51 \times 10^9 \text{ yr} \]

\[ \lambda_{238} = 1.5 \times 366 \times 10^9 \text{ yr}^{-1} \]

\[ U^{235} \rightarrow Pb^{207} \]

\[ \begin{array}{c}
21.7 \% \\
21.3 \% \\
22.6 \%
\end{array} \]

\[ T_{1/2} = 7.13 \times 10^8 \text{ yr} \]

\[ \lambda_{235} = 9.7 \times 35 \times 10^8 \text{ yr}^{-1} \]

\[ Th^{232} \rightarrow Pb^{208} \]

\[ \begin{array}{c}
52.3 \% \\
52.1 \% \\
51.7 \%
\end{array} \]

\[ T_{1/2} = 1.41 \times 10^{10} \text{ yr} \]

\[ \lambda_{232} = 4.9 \times 149 \times 10^9 \text{ yr}^{-1} \]

\[ Pb^{206} \rightarrow Pb^{204} \]

\[ \begin{array}{c}
1.4 \% \\
1.36 \% \\
\end{array} \]

\[ \lambda_{238} = 1.41 \text{ yr}^{-1} \]

\[ [1] : \quad \frac{Pb^{206}(t) - Pb^{206}(0)}{Pb^{207}(t) - Pb^{207}(0)} = \frac{U^{238}(0)}{U^{235}(0)} \frac{(1-e^{-\lambda_{238} t})}{(1-e^{-\lambda_{235} t})} \]

\[ [2] : \quad \frac{Pb^{206}(t) - Pb^{206}(0)}{Pb^{207}(t) - Pb^{207}(0)} = \frac{U^{238}(0)}{Th^{232}(0)} \frac{(1-e^{-\lambda_{238} t})}{(1-e^{-\lambda_{232} t})} \]

\[ [3] : \quad \frac{Pb^{207}(t) - Pb^{207}(0)}{Pb^{208}(t) - Pb^{208}(0)} = \frac{U^{235}(0)}{Th^{232}(0)} \frac{(1-e^{-\lambda_{235} t})}{(1-e^{-\lambda_{232} t})} \]
REFERENCES TO APPENDICES


