DOE/EML-385

REVIEW OF ^{222}Rn IN NATURAL GAS PRODUCED FROM UNCONVENTIONAL SOURCES

By
Carl V. Gogolak

November 1980

U. S. Department of Energy
Environmental Measurements Laboratory
New York, New York

TECHNICAL INFORMATION CENTER
UNITED STATES DEPARTMENT OF ENERGY

DISCLAIMER

DISCLAIMER

Available from the National Technical Information Service, U. S. Department of Commerce, Springfield, Virginia 22161.

Price: Printed Copy A03
Microfiche A01

Printed in the United States of America
USDOE Technical Information Center, Oak Ridge, Tennessee

DOE/EML-385
Distribution Category UC-92a

REVIEW OF ^{222}Rn IN NATURAL GAS PRODUCED FROM UNCONVENTIONAL SOURCES

Carl V. Gogolak

Environmental Measurements Laboratory
U. S. Department of Energy
New York, New York 10014

November 1980

ABSTRACT

A review of the literature on trace radioactivity in natural gas and natural gas products has been performed and the consequent radioactivity concentrations and dose rates due to natural radioactive elements in natural gas produced from Devonian shale wells, western tight gas sands, geo-pressurized aquifiers and coal beds have been studied. Preliminary data on ^{222}Rn concentrations from these energy sources fall within the range observed for more conventional sources. Gas produced from reservoirs with higher than average natural ^{238}U contain higher than average levels of ^{222}Rn. Massive fracturing techniques do not appear to raise the relative concentration of radon in natural gas.

TABLE OF CONTENTS

INTRODUCTION

Natural gas currently supplies about one-fourth of the total energy used in the United States, while it constitutes less than 5% of the total domestic conventional energy reserves (ASET, 1979). Proven producible reserves of gas (209 Tcf)[a] are about a 10 year supply at the present consumption rate. Over the last seven years, new discoveries and extensions of known fields have replaced only 1 Tcf for every 2.5 Tcf consumed (ASET, 1979).

The main objective of the Department of Energy's (DOE) Unconventional Gas Recovery program is to increase recoverable natural gas reserves by developing and demonstrating new recovery methods that will make it economically possible to recover gas resources that are currently considered uneconomic. The four gas resource targets of this program and estimates of the potentially recoverable gas (ASET, 1979) are the gas-bearing Devonian shales of the eastern United States (10-520 Tcf), the low permeability (tight) gas sandstones of the Rocky Mountain region (50-320 Tcf), the free methane present within coal seams (16-500 Tcf), and the high-pressure, methane-saturated saltwater aquifers of the Gulf Coast region (150-2000 Tcf).

Radon-222 is formed from the decay of ^{226}Ra in the natural ^{238}U decay series. Uranium-238 is found in varying amounts throughout the earth's crust (NCRP, 1976). Gaseous ^{222}Rn escapes mineral crystals and collects in porous geological formations and consequently in natural gas production wells. The use of natural gas and natural gas products thus entails exposure to trace radioactivity. This report is a review of previous studies on the concentrations and radiological significance of radon in natural gas from the well-head to the end-point user and an assessment of the differences which may result from the

---

[a] 1 Tcf = 10^{12} cubic feet.

use of natural gas produced from unconventional sources. This report is part of an on-going study of the radiological implications of trace radioactivity which may be released in the development of new energy sources and technologies.

^{222}Rn CONCENTRATION IN NATURAL GAS

Several studies have been performed on the ^{222}Rn content of natural gas products (Johnson et al., 1973; Barton et al., 1973; Gesell, 1975; Gesell et al., 1977; Wardaszko, 1976; van der Heijde et al., 1977). Table 1 lists the concentrations of ^{222}Rn in natural gas at the well head from these various studies as summarized in the 1977 UNSCEAR report (UNSCEAR, 1977). The overall average for the United States is 37 pCi/l on a well by well basis; this value is not weighted for the variation in production between wells (Johnson et al., 1973).

Natural gas is generally processed to remove impurities and the hydrocarbons heavier than methane before being distributed. Studies have shown (Gesell et al., 1977; Gesell, 1975; van der Heijde, 1977; Fries and Kilgren, 1972) that between 30 and 75% of the radon in the well head gas can be removed in processing natural gas for the production of liquified petroleum gas (LPG) which is primarily propane. This occurs because of the thermal separation process used. The boiling point of radon (-61.8°C) falls between that of ethane (-88.3°C) and propane (-42.2°C) and is considerably higher than that of methane (-161.5°C). As a result, the concentration of radon in LPG has been found to be 10±5 times higher at one standard deviation than in the inlet gas (Gesell et al., 1977).

After processing, the natural gas enters a pipeline distribution system. Typically the gas from many different wells become mixed. The gas is transported at speeds of 16-20 km/h, so there is decay of the 3.8 day half-life ^{222}Rn. For transmission lines between the production areas of the Gulf Coast to distribution areas in the northeast, the distance is about 2400 km and thus the transit time equals one to two half lifes. Natural gas has been sampled at various points in the United States distribution systems and the results are summarized

in Table 2 (UNSCEAR, 1977). The average value for the United States is about 23 pCi/l (Johnson et al., 1973). The short lived daughter products of ^{222}Rn have not been found in pipeline gas because they tend to plate out on the sides of the pipes shortly after they are formed. The main pipelines are designed to be operated continuously at full capacity, whereas demand varies seasonally. Therefore, gas is often stored near the distribution centers during times of low demand and released during times of high demand. This storage provides an opportunity for further decay of ^{222}Rn. Seasonal variation in demand for natural gas may result in maximum concentrations during the winter (Barton et al., 1973).

There have been few attempts to measure the increase of ^{222}Rn concentrations in homes resulting from the use of natural gas for cooking and from unventilated space heaters, primarily because of the extensive survey that would be required in order to characterize the "typical" home. Measurements by van der Heijde et al. (1977), however, indicated that heat buildup in the kitchen would often prompt the user to increase the ventilation rate. This may result in a decrease in ^{222}Rn concentration and tend to mask any increase due to ^{222}Rn in the cooking gas.

The distribution system for liquified petroleum gas (LPG) is more complicated than that for natural gas (Gesell et al., 1977). It is shipped from the processing plant by truck, rail or pipeline and then distributed to consumers through retail delivery trucks or central "bottle stations". There may be several intermediate regional distribution and storage centers. Data on the concentrations of ^{222}Rn in LPG at the retail level in various parts of the country are listed in Table 3. The average of 157 samples was 76 pCi/l.

As one would expect, the concentration of ^{222}Rn in LPG decreases both with distance from major gas production and processing areas and also with distance down the retail distribution chain from large storage tanks to delivery trucks to small residential-size storage tanks. Seasonal patterns of ^{222}Rn concentrations in LPG in the Houston area show a minimum during the winter when previously stored gas is brought on to the market to meet the higher

demand, since LPG, like natural gas, is produced at a fairly constant rate throughout the year. The precise seasonal pattern in a given locale for ^{222}Rn concentrations in both natural gas and LPG will depend on both geographical distance from production areas and the distribution system used. Gesell et al. (1977) have assigned average ^{222}Rn concentrations to LPG at the consumer level by state:

150 pCi/l - California
100 pCi/l - Texas
50 pCi/l - Nevada, Utah, Colorado, Arizona, New Mexico
10 pCi/l - all other states.

A summary of the various sources of ^{222}Rn in the United States, including natural gas and LPG, has recently been compiled by Travis et al. (1979). Their estimates of the average ^{222}Rn air concentration (indoor or outdoor as appropriate) due to each source are shown in Table 4.

^{222}Rn CONCENTRATION IN NEW OR UNCONVENTIONAL SOURCES OF NATURAL GAS

Many factors can affect the ^{222}Rn concentrations in natural gas and natural gas products at the consumer level, but these concentrations ultimately depend on the original ^{222}Rn content of the gas at the well head. Each of the four target areas of the Unconventional Gas Recovery program were examined in order to determine how the ^{222}Rn concentrations in gas from these sources might compare with the range (0.2- 1450 pCi/l) and average (37 pCi/l) observed in gas from the conventional sources.

Eastern Gas Shales Project

Sedimentary rock generally contains between 1 and 3 ppm ^{238}U and shales average about 3.7 ppm ^{238}U (NCRP, 1975; UNSCEAR, 1977). Eastern gas bearing Devonian shales, however, can contain higher levels of ^{238}U. Measurements of ^{238}U in Devonian shales and in middle and upper Devonian clastic rocks have

been collected from the literature and summarized in Table 5. The data indicate that ^{238}U concentrations in samples from Devonian shale formations can average up to 40 ppm. Organic rich black shales usually contain twice as much ^{238}U as lighter-colored organic poor shale (Fulton, 1977). Core samples show that ^{238}U concentrations can change by over an order of magnitude over depth intervals of less than one foot in these formations (Leventhal and Goldhaber, 1978). These changes are usually associated with marked observable lithological transitions, and correlate well with the percentage of organic carbon in the rock.

Models have been developed that relate the concentrations of ^{222}Rn in natural gas to the concentrations of ^{238}U in the reservoir rock (van der Heidje et al., 1977; Sakakura et al., 1959). However, these models assume homogeneity in the reservoir and require a knowledge of reservoir parameters which are often not available. These models are discussed in Appendix A.

To determine the effect of these higher ^{238}U concentrations in Devonian shale formations on the ^{222}Rn concentrations of natural gas we obtained samples of gas from existing wells. These were analyzed for ^{222}Rn content at EML. In Table 6 are listed the well head concentrations of ^{222}Rn we observed in natural gas samples from eight wells in Devonian shale formations in Kentucky and West Virginia. These concentrations range over approximately an order of magnitude (26-247 pCi/l). In half of these wells some form of stimulation was used to increase production. These wells had slightly lower production weighted average ^{222}Rn concentrations than those which received no stimulation. In such a small sample, however, this difference is not significant. Nor does there appear to be any significant correlation between well head ^{222}Rn concentration and either flow rate or reservoir pressure. The flow rates from the stimulated wells were generally much lower than from the unstimulated wells. The overall average ^{222}Rn concentration weighted for differences in production among these eight wells was 151 pCi/l.

The data in Table 1 indicate that the average and range observed in these wells is not greatly different from what is observed in wells located

in other parts of the country such as the Texas Panhandle, Oklahoma and Kansas. If one were to assume that the United States average concentration of 37 pCi/l of ^{222}Rn in natural gas were due to approximately 3 ppm ^{238}U in the reservoir rock, the 151 pCi/l of ^{222}Rn observed in the gas from Devonian shale wells is roughly in proportion to the overall average 16 ppm ^{238}U observed in samples of Devonian shale.

Western Tight Gas Sands Project

The recovery of natural gas from the low permeability, western tight gas sands requires massive fracturing of the reservoir formations. Table 1 shows that natural gas from wells in New Mexico, Colorado and Wyoming generally contain 10-50 pCi/l of ^{222}Rn. To investigate the effect of massive fracturing on radon concentrations in natural gas, we have examined data accumulated during project PLOWSHARE in which nuclear explosives were used to stimulate the production of natural gas.

Studies following project GASBUGGY (December 10, 1967 in New Mexico) and project RULISON (September 10, 1979 in Colorado) showed that these detonations caused no significant changes in the ^{222}Rn concentration in natural gas from nearby wells (Bunce and Sattler, 1966; McBride and Hill, 1969; SWRHL, 1970). Table 7 contains data on samples obtained from the test wells following Project RULISON and Project RIO BLANCO (May 17, 1973 in Colorado). The ^{222}Rn concentrations in the natural gas from these wells (10-40 pCi/l) is within the range expected for that area. Thus, it appears that the massive fracturing of these wells did not cause a great increase in ^{222}Rn concentrations in the gas.

Geopressurized Aquifers

The geopressurized geothermal brines of the Gulf Coast region of Louisiana are capable of providing kinetic energy from the high pressures of the reservoir, thermal energy from the hot water and energy from the natural gas which is dissolved in the water (Newchurch et al., 1978). Measurements at one well have shown that the waters were substantially saturated with methane (Karkalits and Hankins, 1979). The geopressurized brines and their

dissolved methane gas have been analyzed for radioactivity (Hankins et al., 1978; Kraemer, 1980) and data are shown in Table 8. Kraemer (1980) has found that nearly all of the ^{222}Rn in the water is separated along with the methane and that nearly all of the ^{222}Rn in the methane can be traced to the ^{226}Ra in the water. There is very little, if any, unsupported ^{222}Rn in the water. The range of ^{222}Rn concentrations found in individual gas samples was 24-238 pCi/l. The unweighted average for the four wells that have been studied is 110 pCi/l. Further data on ^{222}Rn concentrations in methane from this resource should be obtained as more wells are tested.

Free Methane from Coal Beds

During the natural process of coal formation, methane is generated and trapped in the coal seam. For safety reasons, the methane in coal mines is vented to the atmosphere and is thus wasted. Part of the methane from coal project is directed towards the economic recovery and use of this gas.

There are as yet no direct measurements of the ^{222}Rn content of methane recovered from coal mines but the ^{222}Rn content of the air in coal mines has been studied. These data are given in Table 9.

Since the concentration of methane in these air samples was not measured, these results cannot be directly applied to the question of ^{222}Rn concentrations in any natural gas that would be recovered from ventilating coal mines. It is likely, however, that dilute mixtures of methane in ventilating air would be used primarily on site, rather than being processed for the pipeline. The ^{222}Rn in this gas would then be vented to the atmosphere along with the combustion products.

Other portions of the methane are concerned with the recovery of methane from unminable coal seams. This is a situation equivalent to that of a natural gas well in which the reservoir rock is coal. There have been as yet no direct measurements of ^{222}Rn in gas produced this way, but the ^{238}U content of coal was recently studied by Beck et al. (1980). These data, summarized in Table 10, show that coal on the average contains about the same amount of uranium as is

usually found in soil, and about 65% of that usually found in rocks. The emanation fraction of radon from coal is similar to that from typical soil, approximately 13-14%. On this basis, one would not expect particularly high ^{222}Rn concentrations in the gas produced from coal beds.

This review of unconventional sources of natural gas indicates that the ^{222}Rn concentrations of the gas produced from these sources should fall within the overall range that is observed in gas from conventional sources. Gas from Devonian shale wells appears to be higher in ^{222}Rn content than the United States average and is roughly in proportion to the higher average ^{238}U concentrations found in the reservoir rock. Because these unconventional technologies are new, it has not been possible in many cases to obtain samples which could be considered typical of future production, and a precise estimate of the average ^{222}Rn concentrations in gas as it would be delivered to the consumer cannot be made at this time. It is possible, nevertheless, to estimate the well head concentration of ^{222}Rn in natural gas necessary to produce indoor air concentrations in homes which might be considered unacceptable.

^{222}Rn CONCENTRATIONS IN HOMES FROM NATURAL GAS AND NATURAL GAS PRODUCTS

The major potential radiological hazard of ^{222}Rn in natural gas and gas products lies in the combustion of these fuels in unvented appliances. We have used the model home parameters developed by Johnson et al. (1973) and Gesell et al. (1977) to calculate the annual average increase in indoor ^{222}Rn concentrations that would be expected in an average home per unit of ^{222}Rn in fuel as delivered:

$$\text{Average Indoor } ^{222}Rn \text{ Concentration} = \frac{(^{222}Rn \text{ Concentration in Fuel})(\text{Amount of Fuel Used})(\text{Storage decay factor})}{(\text{Home Volume})(\text{Ventilation Rate})} .$$

The results are given in Table 11, and may be adjusted for different house

sizes, ventilation rates and fuel consumptions.

In Table 12 are given the concentrations of ^{222}Rn in fuel as delivered that would be necessary to increase indoor ^{222}Rn air concentrations to given levels above background and the corresponding working level (WL) at an equilibrium factor of 0.5.[b]

UNSCEAR (1977) has compiled background indoor radon concentrations and working levels measurements and estimated a worldwide average of 1 pCi/l of ^{222}Rn with an equilibrium factor of 0.5. This is consistent with the NCRP data for the U.S. (1975) and the more recent data of Breslin and George (1979) and George and Breslin (1980).

The concentrations listed in Table 12 correspond in several cases to levels that have been recommended to limit the exposure of the general public to radon and its daughter products (ICRP, 1960; CFR, 1978; EPA, 1973).

The concentration of ^{222}Rn in fuel as delivered is not equal to the well-head concentration of ^{222}Rn in natural gas, since there is decay during shipping and fractionation of ^{222}Rn between natural gas and LPG during processing. However, by comparing the overall average ^{222}Rn concentrations in natural gas at the well head from Table 1 (37 pCi/l) with the average concentration in natural gas distribution lines from Table 2 (23 pCi/l), it can be conservatively estimated that on average the ^{222}Rn concentrations in natural gas at the use point are generally about 60% of the well-head concentrations.

---

[b]One working level is equivalent to any combination of the short lived daughter products of ^{222}Rn leading to the potential emission of 1.3×10^5 MeV of alpha activity per liter of air. For ^{222}Rn in equilibrium with its daughters, 100 pCi/l of ^{222}Rn results in one working level. However, for the more realistic ratios for indoor air of ^{222}Rn: ^{218}Po: ^{214}Pb: ^{214}Bi: ^{214}Po of 1.0: 0.9: 0.5: 0.35: 0.35, approximately 200 pCi/l of ^{222}Rn would result in one working level. The ratio of the actual working level to that which would occur if the ^{222}Rn present were in equilibrium with its daughters is called the equilibrium factor.

Using the average concentration of ^{222}Rn in LPG at the retail level from Table 3 (76 pCi/l), the ^{222}Rn concentrations in LPG are similarly estimated to be about a factor of two higher than the well-head concentration of the gas from which it is produced. On this basis, the well-head concentrations of ^{222}Rn in natural gas necessary to produce a given increase in indoor ^{222}Rn concentrations from the use of either natural gas or LPG for unvented cooking and space heating are also given in Table 12.

In most cases, the concentrations of ^{222}Rn in well-head gas that would be required to produce unacceptably high indoor ^{222}Rn concentrations are far in excess of those that have been observed. The resultant increases in indoor ^{222}Rn concentrations in the model home for the maximum values of ^{222}Rn found in fuel (from Tables 2 and 3) and for the maximum well-head concentration from an individual well (from Table 1) are shown in Table 12 for comparison. Concentrations of ^{222}Rn in natural gas approaching 1000 pCi/l would be necessary to increase indoor concentrations by more than an annual average of 0.33 pCi/l. On the basis of present information it seems unlikely that ^{222}Rn in natural gas would pose a radiological hazard to domestic users except perhaps in specific local uses near wells with extraordinarily high concentrations.

OCCUPATIONAL EXPOSURE TO ^{222}Rn IN NATURAL GAS AND NATURAL GAS PRODUCTS

Surveys of the potential occupational exposure due to ^{222}Rn and its daughters in natural gas processing plants have been made in the United States (Gesell, 1975) and in the Netherlands (van der Heijde et al., 1977).

In nine United States plants, the external gamma-ray exposure from short-lived radon daughters which had plated out on the internal surfaces of equipment was less than 8 mR/h. The highest levels were found near propane reflux pumps which were usually located in low-occupancy areas. The external gamma-ray exposure levels in the Netherlands plants were all lower

than .01 mR/h, which presumably reflects the lower concentrations of ^{222}Rn in the well head gas which is processed there (see Table 1).

The inhalation of the long-lived ^{222}Rn daughters ^{210}Pb and ^{210}Po may be a hazard in the repairing and cleaning of pumps (Gesell, 1975; van der Heijde, 1977; Fries and Kilgren, 1972), but not during the normal operation of the plant. The pumps in question generally require service at infrequent intervals (0.5-5 years), and much of the inhalation hazard may be avoided by wearing masks when cleaning the internal surfaces.

Gesell (1975) has pointed out that occupational inhalation exposures to ^{222}Rn in natural gas and natural gas products in the air will be controlled by the existing threshold limit values (TLV) for hydrocarbon gases and CO_2. The ICRP guideline (1960) for continuous occupational exposure to ^{222}Rn is 10 pCi/1. The TLV for hydrocarbon gases is 0.1% and, therefore, ^{222}Rn concentrations in these gases of up to 10,000 pCi/1 could occur before the ICRP guideline would be exceeded in a gas processing plant. In the industrial combustion of hydrocarbon gases, the CO_2 TLV of 0.5% would apply. Since a unit volume of CH_4 (methane, the primary constituent of natural gas) produces a unit volume of CO_2 on combustion and C_3H_8 (propane, the primary constituent of LPG) produces 3 volumes of CO_2 on combustion, a minimum dilution of 200 fold in the case of natural gas and 600 fold in the case of LPG is guaranteed by the TLV for CO_2. Thus, natural gas could contain 2000 pCi/1 of ^{222}Rn and LPG could contain 6000 pCi/1 of ^{222}Rn before the ICRP occupational guidelines would be exceeded.

SUMMARY

Studies of the radiological consequences of ^{222}Rn in natural gas and LPG used in unvented ranges and space heaters have been reviewed. On the basis of some simple model calculations it would appear that well-head concentrations of ^{222}Rn in natural gas approaching 1000 pCi/1 would be required to produce increases in indoor ^{222}Rn concentrations in excess of current guidelines in the average American home using these appliances. Radon-222 concentrations at the well head of individual conventional wells as high as 1450 pCi/1

have been observed, but the average is closer to 37 pCi/l. Preliminary measurements of ^{222}Rn concentrations in natural gas from new or unconventional sources fall within the range observed for conventional wells.

The average ^{222}Rn concentration in natural gas from 8 Devonian shale wells (151 pCi/l) is higher than the United States average (37 pCi/l) because of the higher ^{238}U concentrations in this shale. The maximum concentration found was 249 pCi/l.

Many of the unconventional natural gas recovery projects involve the use of massive fracturing techniques to stimulate production from wells. The production weighted average ^{222}Rn concentrations in natural gas from stimulated and natural flow wells in Devonian shale formations showed no significant difference. In fact, the concentrations in the stimulated wells were slightly lower. A review of the data obtained in the massive fracturing of western tight gas sandstone deposits during project PLOWSHARE also indicates that the relative concentration of ^{222}Rn in natural gas from these wells was probably not effected substantially by the fracturing. Because reservoir fracturing is involved in most new gas recovery projects, however, a few careful studies of the effects of fracturing on specific wells would be useful. This would include analysis for ^{222}Rn of samples taken before and after stimulation and either a calibrated well log or analysis of core samples for ^{238}U with depth. If any effects of fracturing on ^{222}Rn concentrations are observed, they may prove useful in interpreting the efficiency of various fracturing techniques. Radon-222 could thus be used as an internal reservoir tracer.

Preliminary data from several wells in southern Louisiana show ^{222}Rn concentrations in methane dissolved in geopressurized brines averaging about 110 pCi/l. Further data should be obtained as this resource is developed. There is currently no data available on ^{222}Rn concentrations in free methane recovered from coal beds. Nevertheless, ^{238}U concentrations in coal are such that one would expect ^{222}Rn concentrations no higher than are observed in wells drilled in other reservoir formations.

Because of the experimental nature of the unconventional gas recovery program it has not been possible in every case to directly obtain information on ^{222}Rn concentrations in natural gas that could be considered typical of future production. However, this review of the new gas recovery technologies was not able to identify any circumstances which would result in a significantly higher average radiological impact on the United States population than exists from conventional recovery technologies.

REFERENCES

ASET (1979).
Assistant Secretary for Energy Technology, Office of Program Control & Support
Fossil Energy Program Summary Document
USDOE Report ET-0087

ACGIH (1973).
Threshold Limit Values for Chemical Substances and Physical Agents in the Workroom Environment with Intended Changes for 1973
American Conference of Governmental Industrial Hygienists, Cincinnati, Ohio

J. A. S. Adams (1962).
Radioactivity in the Lithosphere, p. 1
in: Nuclear Radiation in Geophysics, H. Israel and A. Krebs (Editors)
Springer-Verlag, Berlin

C. J. Barton, R. E. Moore and P. S. Rohwer (1973).
Contribution of Radon in Natural Gas to the Dose from Airborne Radon-Daughters in Homes, p. 134
in: Noble Gases, R. E. Stanley and A. A. Moghissi (Editors)
Proceedings of Symposium held in Las Vegas, September 24-28, 1973
USERDA Report CONF-730915

H. L. Beck, C. V. Gogolak, K. M. Miller and W. M. Lowder (1980).
Perturbations on the Natural Radiation Environment due to the Utilization of Coal as an Energy Source
in: Proceedings of the Third International Symposium on the Natural Radiation Environment, Houston, Texas, April 23-28, 1978
USDOE Report CONF-780422

G. A. Boysen (1976).
Offsite Radiological Safety Program for Project Rulison Flaring, Phase III
USEPA Report EMSL-LV-539-8

A. J. Breslin and A. C. George (1979).
Radon Sources, Distributions and Exposures in Residential Buildings
Trans. Am. Nuc. Soc., 33, 150

L. A. Bunce and F. W. Sattler (1966).
Radon-222 in Natural Gas
Radiat. Health Data and Reports, 7, 441-444

M. W. Carter and A. A. Moghissi (1974).
Radiation Dose from Natural Gas vs that from Nuclear Stimulated Natural Gas, p. 355
Proceedings of the 8th Topical Midyear Symposium of the Health Physics Society, Knoxville, TN, Oct. 21-24, 1974
USAEC Report CONF-741018

CFR (1978).
Code of Federal Regulations, Title 10, Part 20, Appendix B, Amended 40 FR 50704

EPA (1977).
Radiological Quality of the Environment in the United States
USEPA Report 520/1-77-009, p. 62

B. A. Fries and K. H. Kilgren (1972).
Radon in Natural Gas and Natural Gas Liquids
Chevron Research Co., Richmond, CA

L. P. Fulton (1977).
Uranium in Devonian Shales, p. 111
USDOE Report COO-1650-1

A. G. George and A. J. Breslin (1978).
Distribution of Ambient Radon and Radon Daughters in Residential Buildings in the New Jersey - New York Area, p. 127
Proceedings of the Third International Symposium Natural Radiation Environment, Houston, Texas, April 23-28, 1978
USDOE Report CONF-780422

T. F. Gesell, R. H. Johnson, Jr., and D. E. Bernhardt (1977).
Assessment of Potential Radiological Health Effects from Radon in Liquified Petroleum Gas
USEPA Report 520/1-75-002

T. F. Gesell (1975).
Occupational Radiation Exposure Due to ^{222}Rn in Natural Gas and Natural Gas Products
Health Physics, 29, 681-687

R. L. Gotchy (1972).
Project Rulison Final Operational Radioactivity Report: Production Tests
USAEC Report NVO-112

B. E. Hankins, R. E. Chavanne, R. A. Han, O. C. Karkalits, and J. I. Palermo (1978).
Chemical Analyses of Geothermal Waters from a South Louisiana Well
Proceedings of the 3rd Geopressurized Geothermal Energy Conference, University of Southwestern Louisiana, Lafayette, Louisiana, November 16-18, 1977

J. H. Harley (1973).
Environmental Radon, p. 109
in: Noble Gases, R. E. Stanley and A. A. Moghissi (Editors)
Proceedings of a Symposium held in Las Vegas, September 24-28, 1973
USERDA Report CONF-730915

Hennington, W. M. (1980)
Mitchell Energy Corp., private communication

M. Humphreys and G. M. Friedman (1972).
Radioactive Trace Elements in Upper Devonian Clastic Rocks, North Central Pennsylvania
USAEC Report GJO-7407

ICRP, International Commission on Radiological Protection (1960).
Recommendations of the ICRP on Permissible Dose for Internal Radiation, Report No. 2
Pergamon Press, N.Y.

P. W. Jacoe (1953).
Occurrence of Radon in Nonuranium Mines in Colorado
AMA Arch. Ind. Hyg. Occup. Med., 8, 118

R. H. Johnson, Jr., D. E. Bern, N. S. Nelson and H. W. Calley, Jr. (1973).
Assessment of Potential Radiological Health Effects from Radon in Natural Gas
USEPA Report 520/1-73-004

O. C. Karkalits and B. E. Hankins (1979).
Chemical Analysis of Gas Dissolved in Geothermal Waters in a South Louisiana Well
Proceedings of the 3rd Geopressurized Geothermal Energy Conference, University of Southwestern Louisiana, Lafayette, Louisiana, November 16-18, 1977

T. F. Kraemer (1980).
U. S. Geological Survey, Gulf Coast Hydroscience Center, National Space Technology Laboratories, NSTL Station, Mississippi, private communication

P. Kruger, G. Cederberg, and L. Semprini (1978).
Radon Data - Phase I Test LASL Hot Dry Rock Project January 28 - April 27, 1978
Stanford University Geothermal Project Report SGP-TR-27

P. Kruger, A. Stoher and A. Umona (1977).
Radon in Geothermal Reservoir Engineering
Geothermics, 5, 13-19

P. Kruger, G. Warren and B. D. Honeyman (1977).
Radon as an Internal Tracer in Geothermal Reservoirs
Nuclear Methods in Environmental and Energy Research, Proceedings of the 3rd Conference October 10-13, 1977, University of Missouri, Columbia
James R. Vogt (Editor)
USDOE Report CONF-771072

J. S. Leventhal and M. B. Goldhaber (1978).
New Data for Uranium, Thorium, Carbon and Sulfur in Devonian Black Shale from West Virginia, Kentucky and New York, p. 215
Proceedings of the 1st Eastern Gas Shales Symposium, October 17-19, 1977
USDOE Report MERC/SP-TI/5

H. F. Lucas, Jr. (1967).
Radon in Coal Mine Air, Part II
Radiological Physics Division Annual Report, Argonne National Laboratory, July 1966 - June 1967
USAEC Report ANL-7360, pp. 104-105

H. F. Lucas, Jr. and A. F. Gabrysh (1966).
Radon in Coal Mines, p. 58
Radiological Physics Division Annual Report, Argonne National Laboratory, July 1965 - June 1966
USAEC Report ANL-7220

J. R. McBride and D. Hill (1969).
Offsite Radiological Surveillance for Project Gasbuggy June 1967 - July 1968
Radiation Health Data and Reports, 10, 535-546

NCRP, National Council on Radiation Protection and Measurements (1975).
National Background Radiation in the United States
NCRP Report No. 45

NCRP, National Council on Radiation Protection and Measurements (1976).
Environmental Radiation Measurements
NCRP Report No. 50

E. J. Newchurch, C. F. Bryan, D. P. Harrison, R. A. Muller, R. E. Wilcox, A. L. Bachman, J. P. Newman, K. J. Cunningham, R. K. Hilding and J. A. Rehage (1978).
A Plan for the Long Term Environmental Assessment of Geopressurized Resource Development in the Louisiana Gulf Coast Region
Order No. 8316603, Institute for Environmental Studies, Louisiana State University, Baton Rouge, LA

A. P. Pierce, G. B. Gott and J. W. Mytton (1964).
Uranium and Helium in the Panhandle Gas Field, Texas, and Adjacent Areas
U. S. Geological Survey Professional Paper 454-G

L. J. Provo (1977)
Stratigraphy and Sedimentology of Radioactive Devonian - Mississippian Shales of the Central Appalachian Basin
USERDA Report COO-1650-1

R. L. Rock, G. Suilar, R. T. Beckman, and D. D. Rapp (1975).
Evaluation of Radioactive Aerosols in United States Underground Coal Mines
Mining Enforcement and Safety Administration Informational Report No. 1025. U. S. Department of Interior, Washington, D. C.

A. Y. Sakakura, C. Lindberg and H. Faul (1959).
Equation of Continuity in Geology with Applications to the Transport of Radioactive Gas
U. S. Geological Survey Bulletin 1052-I

C. F. Smith and R. W. Taylor (1973).
Rio Blanco Gas Quality Analysis and Evaluation Program Status Report
USAEC Report UCRL-74868

SWRHL, Southwestern Radiological Health Laboratory (1970).
Offsite Radiological Safety and Resident Evacuation Program for Project Rulison (Detonation Phase)
USDHEW Report SWRHL-94r

C. C. Travis, S. J. Cotter, A. P. Watson, M. L. Randolph, L. M. McDowell-Boyer, and D. E. Fields (19).
A Radiological Assessment of Radon-222 Released from Uranium Mills and Other Natural and Technologically Enhanced Sources
USNRC Report NUREG/CR-0573, ORNL/NUREG-55

C. C. Travis (1979)
Environmental Sources of Radon
Trans. Am. Nuclear Soc., 33, 144

UNSCEAR, United Nations Scientific Committee on the Effects of Atomic Radiation (1977).
Report to the General Assembly, Annex B, Paragraph 262-266

H. B. van der Heijde, H. Beens and A. R. de Monchy (1977).
The Occurrence of Radioactive Elements in Natural Gas
Ecotoxicology and Environmental Safety, 1, 49-87

T. Wardaszko (1976).
Radiation Hazard to Population Due to Radon-222 in Natural Gas
Postepy Fiz. Med. XI, 4

J. H. Way, Jr., and G. M. Friedman (1972).
Radioactive Trace Elements in Middle and Upper Devonian Clastic Rocks, Catskill Mountain Area, N.Y.
USAEC Report GJO-7406

TABLE 1

RADON-222 CONCENTRATIONS IN NATURAL GAS AT THE WELL HEAD*

LOCATION	AVERAGE (pCi/l)	RANGE (pCi/l)
United States		
Colorado, New Mexico	25	0.2 - 160
Texas, Kansas, Oklahoma	<100	5 - 1450
Texas Panhandle	-	10 - 520
Colorado	25.4	11 - 45
NW New Mexico, SW Colorado	15.8 - 19.4	-
NW New Mexico, SW Colorado	29.4	12 - 59
California	-	1 - 100
Gulf Coast (Louisiana, Texas)	5	-
Kansas	100	-
Wyoming	10	-
Overall Average	37	
Canada		
Alberta	62	10 - 205
British Columbia	473	390 - 540
Ontario	169	4 - 800
Federal Republic of Germany	-	1.0 - 9.6
Netherlands		
Slochteren	-	1.1 - 2.8
Other Fields	-	3.7 - 44.7
Nigeria		
Niger Delta	-	0.9 - 2.9
North Sea		
Lemon Field	-	2.0 - 3.8
Indefatigable Field	1.8	-
Borneo		
Ampa Field	-	1.5 - 3.2

*UNSCEAR, 1977

TABLE 2

RADON-222 CONCENTRATIONS IN NATURAL GAS IN DISTRIBUTION LINES*

LOCATION	AVERAGE (pCi/l)	RANGE (pCi/l)
United States		
Chicago	14.4	2.3 - 31.3
New York City	1.5	0.5 - 3.8
Denver	50.5	1.2 - 119
West Coast	15	1 - 100
Colorado	25	6.5 - 43
Nevada	8	5.8 - 10.4
New Mexico	45	10 - 53
Houston	8	1.4 - 14.3
Overall Average	23	

*UNSCEAR, 1977

TABLE 3

RADON-222 CONCENTRATION IN RETAIL LIQUIFIED PETROLEUM GAS*

LOCATION	AVERAGE (pCi/l of gas)	RANGE (pCi/l of gas)
Alabama, Mississippi, Georgia, Florida	1.1	0.5 - 3.8
Arkansas, Louisiana	3.8	0.5 - 13.4
California	151	2 - 1049
Arizona - New Mexico	37	0.5 - 120
Texas:		
Panhandle & West Texas	95	3 - 292
Gulf Coast & South Texas	151	0.5 - 1240
East Texas	34	0.7 - 287
Oklahoma	4.5	0.9 - 10.1
Kentucky, Tennessee, North Carolina, South Carolina	1.9	0.5 - 10.3
Overall Average of 157 samples	76	

*Gesell et al., 1977

TABLE 4

SOURCES AND QUANTITIES OF RADON-222 RELEASED IN THE UNITED STATES*

SOURCE	ESTIMATED ANNUAL RELEASE (Ci/y)	ESTIMATED AIR CONCENTRATION (pCi/m^3)
Building Interiors	28,000	84**
Natural Soil	120,000,000	120
Evapotranspiration	8,800,000	8.8
Tillage of Soil	3,100,000	3.1
Natural Gas		
Domestic Ranges	200	3**
Domestic Heaters	80	10**
Industry	11,000	.012
Uranium Industry		
Mining	200,000	.22
Milling (active)	150,000	.078
Milling (inactive)	51,000	.027
Non-Uranium Mines		
Phosphate	53,000	.053
Coal	14,000	.014
Phosphate Fertilizer	48,000	.048
Liquified Petroleum Gas		
Domestic Ranges	1.8	.16**
Domestic Heaters	1.3	.92**
Geothermal Power	580	.00058
Coal Fired Power Plants	500	.0005
Gas and Oil Wells	230	.00023

*Travis, 1979

**Concentration in interior spaces.

TABLE 5

URANIUM-238 CONCENTRATIONS IN DEVONIAN ROCK

	MIN. (μg/g)	MAX. (μg/g)	AVERAGE (μg/g)	NO. OF SAMPLES	REFERENCE
Middle and Upper Devonian Clastic Rock					
North Central PA	0.6	165	3.9	300	Humphreys and Friedman (1972)
North-East PA	2.1	244	40.9	16	Way and Friedman (1972)
Catskill Mountains, NY	0	440	2.7	884	" "
Devonian Shale					
Cattarangus County, NY	1.5	15.8	6.7	48 (1 core)	Leventhal and Goldhaber (1978)
Perry County, KY	3.2	38.0	16.6	21	" "
Lincoln County, W. VA	3.4	36.6	11.7	38	" "
Jackson County, W. VA	3.4	21.0	9.1	25	" "
Kentucky	6	74	27.7±3.2		Fulton (1977)
Ohio, Kentucky, Tennessee, AL	1	106	32.9±2.9		" "
Gallia Co., OH	0	25	10	(Well log)	Hennington (1980)
Overall Average			16		

TABLE 6

RADON-222 CONCENTRATION IN NATURAL GAS FROM DEVONIAN SHALE WELLS

Location	Flow Rate (MCFD)*	Reservoir			Stimulation	222 Rn Conc. (pCi/l)
		Temp. (° F)	Pressure (psi)	Porosity (%)		
Letcher Co., KY	25	86	250	6	Yes	104
Johnson Co., KY	20	85	180	8	Yes	247
Floyd Co., KY	60	80	-	-	No	231
" "	100	85	-	-	No	249
Martin Co., KY	111	95	400	-	No	177
" "	12.5	95	213	-	Yes	26
Mingo Co., W. VA.	442	95	470	-	No	116
" "	8.5	95	168	-	Yes	43

	Average Radon-222 Concentration production weighted (pCi/l)
Stimulated Wells	125
Unstimulated Wells	154
All Wells	151

*Millions of cubic feet per day.

TABLE 7

RADON-222 CONCENTRATIONS IN NATURAL GAS FROM NUCLEAR STIMULATED WELLS

	^{222}Rn Concentration (pCi/l)	Reference
Rulison (9/10/69)		
Calibration Flaring: 10/5/70	12	Boysen (1976)
2nd Production Test: 12/2/70	35	Gotchy (1972)
Intermediate Rate Flaring: 12/5/70	11	" "
12/13/70	18	" "
Rio Blanco (5/17/73)		
Following Chimney Re-entry		
10/13/73	10	Smith and Taylor (1973)
10/17/73	40	" " "
10/25/73	40	" " "
		" " "

TABLE 8

RADON-222 IN METHANE FROM GEOPRESSURIZED AQUIFERS

Well	^{226}Ra in Water* (pCi/l)	^{222}Rn in Gas* (pCi/l)	Reference
#1 Edna Delcanbre Vermilion Parish, LA			
Sand #1	238 (6)	43 (6)	Hankins et al (1978)
Sand #3	393 (4)	60 (4)	" "
#2 Southport Beulah Simon Vermilion Parish, LA	277 (8)	97 (4)	Kraemer (1980)
#2 Fairfax Foster Sutter St. Mary Parish, LA	796 (13)	238 (1)	" "
#1 SL6701, Block 22 East Cameron Area Offshore, LA	157 (10)	-	" "
#3 Alma Plantation East Baton Rouge Parish, LA	88 (1)	-	" "
#1 Lorio East Baton Rouge Parish, LA	126 (1)	-	" "
#2 Pleasant Bayou Brazoria County, TX	727 (8)	-	" "
Average	350	110	

*Figures in parentheses indicate number of samples analyzed.

TABLE 9

RADON-222 CONCENTRATIONS IN U.S. COAL MINE AIR

Location	No. of Mines	Average (pCi/l)	Range (pCi/l)	Reference
Pennsylvania	18	13.2	0 - 147	Lucas and Gabrysh (1966)
West Virginia, Kentucky and Tennessee:	9	142	0.3 - 1108.7	Lucas (1967)
Still Air	5	422	11.7 - 1108.7	" "
Moving Air	9	18	0.3 - 79.1	" "
Colorado	-	-	10 - 130	Jacoe (1953)
Various U.S. Mines	213		0 - 58*	Rock et al. (1975)

*Inferred from Working Level Assuming an Equilibrium Factor of 0.5.

TABLE 10

URANIUM, THORIUM, AND POTASSIUM CONCENTRATIONS IN COAL AS MINED
(FRACTION OF DRY WEIGHT)

Region (Type)	No. of Samples	U (μg/g) Range	U Geo. Mean	U Mean	Th (μg/g) Range	Th Geo. Mean	Th Mean	K (g/100 g) Range	K Geo. Mean	K Mean
PA. Anthracite	53	0.3-25.2	1.2	1.5	2.8-14.4	4.7	5.4	.019-1.5	.16	.24
Appalachian (b)	331	<0.2-10.5	1.0	1.4	2.2-47.8	2.8	4.9	.008-2.4	.13	.23
Midwest (b)	143	0.2-43	1.4	3.3	<3 -79	1.6	5.2	.011-.53	.11	.16
N. Gt. Plains (sb, b)	93	<0.2-2.9	0.7	0.9	<2 -8.0	2.4	2.7	.006-.37	.03	.04
Gulf Coast, Lignite	34	0.5-16.7	2.4	3.2	<3 -28.4	3.0	8.3	.012-1.3	.15	.30
Rocky Mt. (b, sb)	134	<0.2-23.8	0.8	1.6	<3 -34.8	2.0	3.6	.003-1.7	.03	.08
Alaska (sb)	18	0.4-5.2	1.0	1.2	<3 -18	3.1	4.4	.016-87	.08	.12
IL Basin	56 (113 K)	0.31-4.6	1.3	1.5	0.71-5.1	1.9	2.1	.04-0.56	.16	.17
Appalachian	14 (23 K)	0.4-2.9	1.3	1.5	1.8-9.0	4.0	4.5	.06-68	.21	.25
Western	22 (29 K)	0.3-2.5	1.0	1.2	0.62-5.7	1.8	2.3	.01-.32	.03	.05
Western	19	.11-3.5	0.85	0.9	-			-		
All Samples	9 (983 K)		1.04	1.74		2.40	4.47		.13	.17

	U Typical Range	U Avg.	Th Typical Range	Th Avg.	K Typical Range	K Avg.
Soil	0.9-4.0	1.8	2-12	6	.12-2.5	1.3
Rocks	0.5-5	2.7	1.6-20	9.6	0.3-5	2.1

sb = Subbituminous b = Bituminous
From Beck et al, 1980.

TABLE 11

EXPOSURE CONDITIONS FOR RADON-222 IN NATURAL GAS AND LPG*

Parameter	Average	Possible Range	Annual Average Increase in Interior Radon-222 Concentration**
House size (m^3)	226.6	(142 - 425)	
Ventilation rate (air change/hr)	1	(.25 - 5)	
Heating Requirements (degree days/day)	7.77	(2 - 33)	
Decay factor for LPG home tank storage	.183	(.363- .0613)	
Unvented Fuel Use:			
Natural Gas Ranges (m^3/day)	.765	(0 - 1.19)	.00014
Natural Gas Space Heaters (m^3/ degree day)	.354	(.28 - .42)	.00051
LPG Ranges (m^3/day)	.306	(0 - .476)	.00001
LPG Space Heaters (m^3/degree day)	.142	(.112- .168)	.00004

*Adapted from Johnson et al (1973) and Gesell et al, 1977.

**Per pCi/l in fuel as delivered.

TABLE 12

CONCENTRATIONS OF RADON-222 IN NATURAL GAS OR LPG NECESSARY TO PRODUCE A GIVEN INDOOR CONCENTRATION OF RADON-222

Average Indoor ^{222}Rn Concentration Above Background (pCi/l)	Working Level (0.5 Equilibrium Factor)	Fuel Concentration Cooking plus space heating (pCi/l)		Well-head Concentration necessary to produce a given fuel concentration (pCi/l)	
		Natural Gas	LPG	Natural Gas	LPG
.062	.00031	-	1240+		620
.077*	.00039	119+	-	191	
.15	.00073		2900		1450++
.33	.00165	507	6600	816	3300
.57	.00283	870		1450++	
1	.0050***	1538	20000	2474	10000
2	.010	3077	40000	4950	20000
3**	.015	4615	60000	7424	30000
4	.020	6154	80000	9900	40000
5	.025***	7692	100000	12374	50000
10	.050	15385	200000	24750	100000

* ICRP (1960) recommended maximum concentration above background.
** 10 CFR 20 (1978) maximum concentration above background in unrestricted areas.
***Surgeon General's Guidance to the State of Colorado regarding homes contaminated by uranium mill tailings (see for example EPA, 1977): Total (including background) below .01 WL - no remedial action necessary
Total (including background) above .05 WL - remedial action recommended
Background is estimated as 1 pCi/l Radon-222 with a 50% equilibrium factor.
\+ Maximum found in U.S. fuels from Tables 2 and 3.
++ Maximum found in U.S. wells from Table 1.

APPENDIX A

ESTIMATION OF ^{222}Rn CONCENTRATIONS IN NATURAL GAS FROM THE ^{238}U CONCENTRATIONS IN RESERVOIR ROCK

Computational models have been developed that relate the concentrations of ^{222}Rn in natural gas to the concentrations of ^{238}U in the reservoir rock. The simple model of van der Heijde et al. (1977) is

$$C = 3.3 \times 10^5 \frac{P_s}{P_r} \frac{T_r}{T_s} \frac{\epsilon \chi}{\Phi} \rho Z, \quad (1)$$

where

C is the ^{222}Rn concentration in pCi/l in natural gas at STP (15°C , 1 bar),

P_s is standard pressure,

T_s is standard temperature,

p_r is reservoir pressure,

T_r is reservoir temperature,

ϵ is the emanating power of ^{222}Rn in the rock, i.e. that fraction of the ^{222}Rn produced which escapes into interstitial spaces,

χ is the concentration (g/g) of ^{238}U in the reservoir rock,

Φ is the porosity of the reservoir,

ρ is the density of the reservoir rock (g/l), and

Z is the gas deviation factor from ideal gas behavior.

The specific activity of ^{238}U is 3.3×10^5 pCi/g. Using $\Phi = 0.25$, $\epsilon = 0.15$ and $\rho = 2.5 \times 10^3$ g/l, van der Heijde et al. (1977) estimated the frequency distribution of ^{238}U in the lithosphere from the ^{222}Rn concentrations in natural gas from 125 wells. This estimate was in reasonable agreement with the frequency distribution found by Adams (1962), based on a survey of rock sample analyses.

A more detailed analysis by Sakakura et al. (1959) resulted in a similar relation:

$$C = 3.3 \times 10^5 \frac{P_s}{P_w} \frac{T_w}{T_s} \frac{\epsilon\chi}{\Phi} \rho \left[1 + \frac{\Delta p^2}{2\, p_w^2 \log \frac{R}{r_w}}\right]^{-1} , \quad (2)$$

where:

P_w is well head pressure,
T_w is well head temperature,
$\Delta p^2 = p_r^2 - p_w^2$,
R is the reservoir radius, and
r_w is the well radius.

The term in brackets arises from a power series expansion to first order of the pressure ratio as a function of distance from the well

$$\frac{p(r)}{P_w} = \left[\frac{\Delta p^2}{p_w^2 \log \frac{R}{r_w}} \log \frac{r}{r_w} + 1\right]^{1/2} .$$

For $\Phi = 0.10$, $\epsilon = 0.10$, $\rho = 3 \times 10^3$ g/l, R = 500 ft and r_w = 3/8 ft, Sakakura et al. (1959) used ^{222}Rn concentrations measured in natural gas from wells in the Panhandle Field in Texas to estimate average ^{238}U concentrations in the reservoir rock of 0.4-9 ppm. However, ^{238}U in the Panhandle Field is generally concentrated in asphaltite nodules which average 0.4% uranium, whereas the reservoir rock generally contains 1-5 ppm uranium (Pierce, et al., 1964). With such inhomogeneity, the significance of the model results are not clear. Unfortunately, neither model has been tested rigorously on individual wells.

The difficulty in applying these models is often rooted in a lack of knowledge of the reservoir parameters of temperature, pressure, porosity, ^{238}U concentration and ^{222}Rn exhalation rate. Inhomogeneity in source and also in the fracture system further complicates the problem. However, these models may be useful for studying reservoir dynamics in certain special

circumstances. Further work is being performed on the behavior of ^{222}Rn in fractured reservoir systems as part of the Stanford University Geothermal Program (Kruger 1977a; 1977b; 1978).