

**ASSESSMENT OF POTENTIAL RADIOLOGICAL  
POPULATION HEALTH EFFECTS FROM RADON IN  
LIQUEFIED PETROLEUM GAS**



**U.S. ENVIRONMENTAL PROTECTION AGENCY**

**Office of Radiation Programs**

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**U. S. ENVIRONMENTAL PROTECTION AGENCY  
Office of Radiation Programs  
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## FOREWORD

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## ABSTRACT

Liquefied petroleum gas (LPG) contains varying amounts of radon-222 which becomes dispersed within homes when LPG is used in unvented appliances. Radon-222 decays to alpha-emitting daughter products which are associated with increased lung cancer when inhaled and deposited in the respiratory system. The average dose equivalents to the bronchial epithelium from the use of LPG in unvented kitchen ranges and space heaters are estimated to be about 0.9 and 4.0 mrem/year, respectively. When extrapolated to the United States population at risk, the estimated tracheobronchial dose equivalents are about 20,000 and 10,000 person-rems/year for these appliances, or a total of about 30,000 person-rems/year. These doses are very small compared to other natural and man-made sources of ionizing radiation. It is estimated that these low doses would result in less than one lung cancer a year for the total U. S. population. Consequently, the use of LPG containing radon-222 does not contribute significantly to the incidence of lung cancer in the United States. Furthermore, the cost for control of radon levels in LPG would be over \$50 million for reduction of one health effect, therefore it is concluded that a requirement for such controls would not be cost effective on a national basis. This study did indicate that individual dose equivalents could possibly exceed 500 mrem/year. However, existing data are not sufficient to determine the significance of such potentially high individual doses. ORP will be evaluating this matter further.

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# ASSESSMENT OF POTENTIAL RADIOLOGICAL HEALTH EFFECTS FROM RADON IN LIQUEFIED PETROLEUM GAS

## INTRODUCTION

### Radon in Liquefied Petroleum Gas (LPG)

Most natural gas is not used directly from production wells but undergoes routine processing for removal of impurities and the heavier, more valuable hydrocarbons. Part of these heavier hydrocarbons, consisting principally of propane, are bottled under pressure for sale as a fuel known as liquefied petroleum gas (LPG). The process for obtaining LPG is of particular interest because the radioactive noble gas, radon-222 a normal component of natural gas, is separated along with LPG. When this LPG is burned in unvented appliances, such as kitchen ranges and space heaters, the radon is released within the home. Here radon-222 decays to alpha-emitting daughter products which can contribute to lung dose when inhaled and deposited in the respiratory system. At high exposure levels, which occurred in some early underground uranium mines, radon-222 has been associated with the induction of lung cancer. This report assesses the potential for such lung cancers in the general population exposed to radon from LPG. The need for this assessment was determined from earlier studies on the health significance of radon in natural gas by Barton, et al. (1,2), Gesell (3,4), and Johnson, et al. (5,6).

### Approach

Potential health effects from radon in LPG are estimated according to the model outlined in figure 1. The overall analysis involves determining the radon-222 concentration in LPG at the points of home use, the definition of exposure conditions for calculating individual and population dose equivalents, and finally the calculation of potential health effects. Figure 1 also shows many of the factors to be considered in each step of this analysis.

### Scope and Objectives

This assessment is concerned with potential health effects which may result from inhalation of radon daughter products in homes where LPG containing radon-222 is used in unvented cooking ranges or space heaters. The use of LPG in other home appliances is of much less importance (5) and will not be included here.

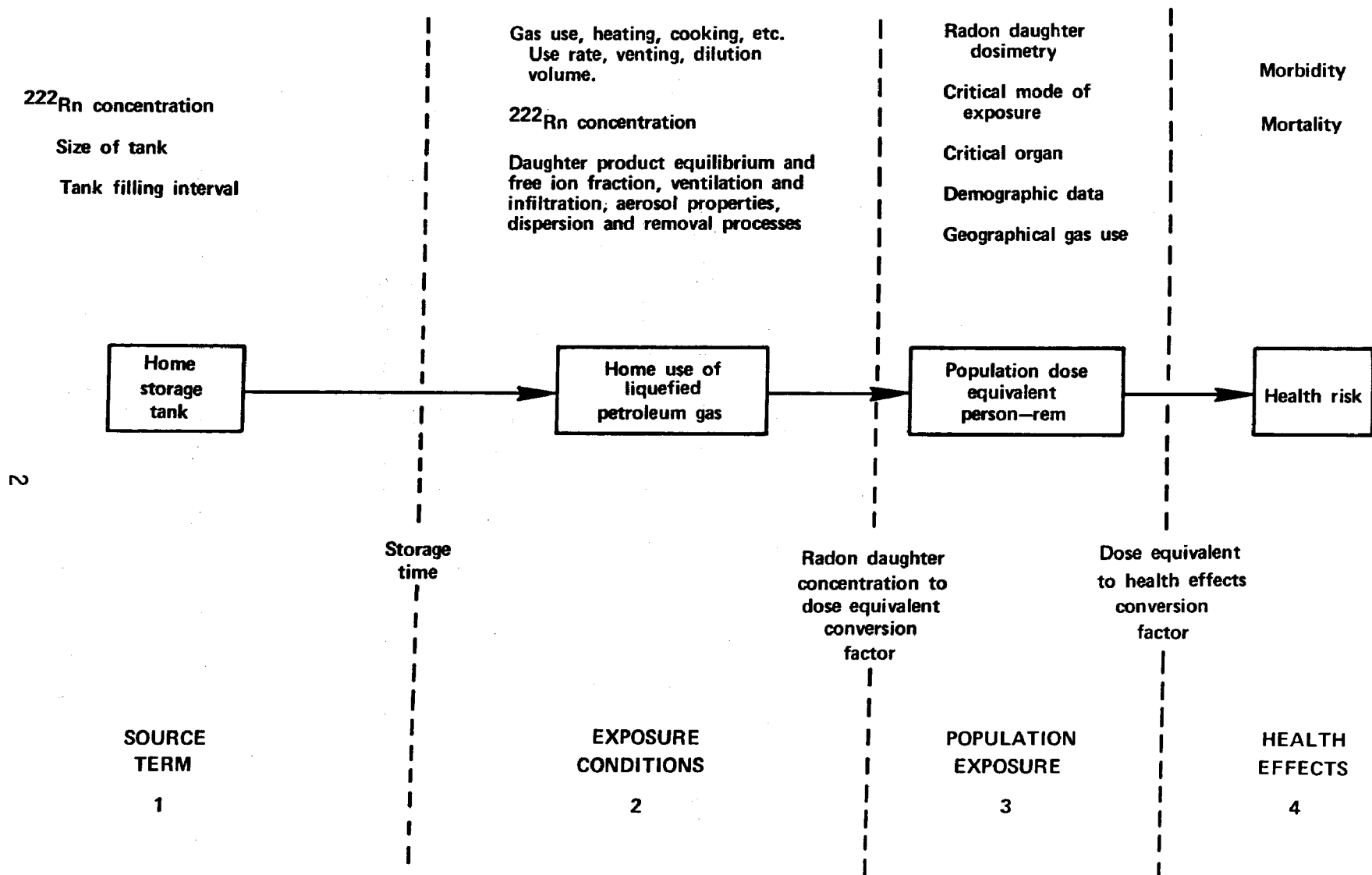


Figure 1. Model for estimating potential health effects from radon in liquefied natural gas

Information on the following items is provided in this report:

- (a) measured radon-222 concentrations in natural gas and in LPG at processing plants and retail outlets,
- (b) estimated radon-222 concentrations at points of consumer use,
- (c) critical mode of exposure and radon dosimetry,
- (d) individual and population dose equivalents, and
- (e) estimation of potential health effects and interpretation of their significance in relation to costs for control of radon in LPG.

### RADON CONCENTRATIONS IN NATURAL GAS

Since LPG and its associated radon are derived primarily from natural gas, it is useful to briefly review the source of radon in natural gas. Radon-222 is the gaseous daughter product of radium-226 which is part of the naturally occurring uranium-238 decay series. According to Bell (7) uranium minerals are distributed throughout the earth's crust, often in association with carbonaceous materials. Sedimentary formations that are good hosts for uranium deposits are generally good for petroleum as well. The genesis of uranium deposits is not certain, but it is believed that soluble uranium is deposited from transporting ground water by chemical reducing conditions resulting from decaying organic material (8). Adsorption and the formation of uranium metallicorganic compounds may also be involved. Bell (7) noted that there is no evidence that petroleum acts as an ore forming fluid for uranium but the average uranium concentrations in rock formations do become elevated by the above processes.

The deposits of uranium associated with many petroleum or natural gas bearing formations result in greater amounts of radium-226 available for decay to radon-222. In addition, it is common for radium to leach from the adjacent uranium minerals so that more of the gaseous radon-222 is able to diffuse into the natural gas, i.e. it is not retained within the solid minerals<sup>1</sup> (9). Since radon is an inert gas, it permeates porous geological formations along with natural gas and is collected in production wells with methane, the primary component of natural gas.

Radioactivity in natural gases was first reported for Canadian gas in 1904 by Satterly and McLennan (10). At that time, it was suggested that helium, a known product of radioactive decay, might be associated with radioactivity. Somewhat later (1918), a systematic survey was made of Canadian natural gases (10) in an effort to find a relation between

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Tanner, A.B., U.S. Geological Survey, Reston, Virginia, personal communications to D. E. Bernhardt, 1973 and 1974.

helium concentrations and radioactivity. The radioactivity was reported as due to "radium emanation," now known to be radon-222, which decays by alpha particle emissions, as do its daughter products, polonium-214 and 218. Each alpha particle consists of two protons and two neutrons which is identical with the structure of a helium nucleus without its electrons,  $\text{He}^{++}$ . When this alpha particle or helium nucleus loses its decay energy and comes to rest, it picks up electrons and becomes a neutral helium atom.

Although no strong correlation between radon-222 and helium concentrations was found in the 1904-1918 studies, interest in a possible correlation was revived in the 1940's and early 1950's resulting in a series of papers dealing with radon and helium concentrations in U.S. natural gases (11,12). A comprehensive survey of radon-222 and helium concentrations in the Texas Panhandle Field gases was issued by the U.S. Geological Survey in 1964 (13). The hypothesis that most of the helium found in natural gases is of radiogenic origin is still widely accepted. Nevertheless, no correlation has been found between radon-222 and helium concentrations in individual wells. This is partly the result of rapid equilibration of radon with radium deposits which may be moved in time by ground water, whereas the helium has been collecting over geologic time.

Subsequent to the reports dealing with possible radon-222 and helium concentration correlations, reports were made on radon-222 concentrations in wellhead natural gas in connection with nuclear gas well stimulation experiments (14,15). These reports dealt with radon concentrations in natural gas in San Juan Basin, located in southwestern Colorado and northwestern New Mexico. These papers are significant because the potential for exposure of population groups to radon-222 via the natural gas pathway was explicitly recognized. These investigations determined that the Project Gasbuggy nuclear stimulation experiment did not raise the radon-222 concentrations in the neighboring wells above the naturally occurring levels.

In addition to the primary sources of data on radon in natural gas mentioned above, there are other data available in published and unpublished report form <sup>2,3</sup> (18-22) which have been reviewed by Gesell (3,4,16,17) and by Johnson et al. (5,6). A summary of the presently available information on radon-222 at the wellhead is given in table 1. The radon concentrations are seen to range from nearly zero to 1450 pCi/liter with averages for groups of wells up to about 100 pCi/liter in the U.S. and about 169 pCi/liter in Canada. The differences in radon concentrations are not due just to the amount of radium-226 present in the rock formations, but also to the pressure of the gases in the well. The production of radon in a given gas reservoir volume will be fairly stable with time, but as the reservoir pressure is reduced by removal of natural gas, less gas remains to dilute the radon. Thus radon concentrations will increase as gas reservoirs near depletion.<sup>2</sup> Radon concentrations are also influenced by gas flow rates which are functions of geological formation permeabilities as well as pressure.

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<sup>2</sup>Tanner, A.B., op. cit.

<sup>3</sup>Skipp, B., U.S. Geological Survey, unpublished data reports from work in the 1950's in Kansas, Oklahoma, and Texas.

Table 1.  $^{222}\text{Rn}$  Concentrations in Natural Gas at Production Wells

Area	$^{222}\text{Rn}$ level (pCi/l)		Reference
	Average	Range	
Ontario, Canada	169	4-800	10,18
Alberta, Canada	62	10-205	10,18
British Columbia	473	390-540	10,18
Colorado, New Mexico	25	0.2-160	14
Texas, Kansas, Oklahoma	<100	5-1450	13,18
Texas Panhandle	---	10-520	11
Colorado	25.4	11-45	15,19,20
Project Gasbuggy Area	15.8-19.4	---	15
Project Gasbuggy Area	29.4	12-59	21
California	---	1-100	22
Gulf Coast (Louisiana, Texas)	5	---	4*
Kansas	100	---	5*
Wyoming	10	---	5*

\*<sup>4</sup>Kaye, S. V., Oak Ridge National Laboratory, Memorandum to distribution. Progress Report for October 1 - December 31, 1972. January 17, 1973.

\*<sup>5</sup>Bernhardt, D. E., Memorandum to D. W. Hendricks, Radon-222 in natural gas (May 24, 1973).

## RADON CONCENTRATIONS IN LPG

### At the Processing Plant

A generalized flow sheet for the production and distribution of natural gas and LPG is shown in figure 2. Some of the factors affecting the concentration of radon-222 are also given in this figure. In particular, the behavior of radon in the processing, storage, and sale of LPG will be considered in detail.

Natural gas from the wellhead is usually processed by thermal fractionation and other means to remove the heavier, more valuable hydrocarbons and impurities (23). Gas processing plants are usually located near the gas fields which supply them. After processing, the "residue gas" as the processed natural gas is called, may be sold for use as a fuel or chemical feedstock or it may be pumped back underground to sweep the oil fields. The latter process is called a cycle operation and produces little or no methane for sale. After the field has been swept and the cycling operation is no longer profitable, residue gas is sold and the cycling operation curtailed. The typical products of a gas processing plant together with the average production for 1971 are given in table 2.

While the details of the processing method may vary, the physical principle behind the separation of the heavier hydrocarbons from the inlet stream mixture is thermal fractionation. Table 3 gives the boiling points for several of the products of gas processing together with that of radon. It is clear that the boiling point of radon is bracketed by those of ethane and propane, and radon will therefore tend to separate with these fractions.

The makeup of the LPG sold for domestic fuel may be principally propane with small amounts of the other hydrocarbons, or it may be principally butane with small amounts of the other hydrocarbons, or mixtures of propane and butane. The use of butane is limited to regions or seasons where the temperature is not likely to go below 0°C (32°F) because below this temperature the butane will not produce sufficient vapor pressure to be useful. This disadvantage coupled with the increasing value of butane as a chemical feedstock has curtailed its use as a fuel. Thus most of the LPG used as a domestic fuel is propane-based. Gesell (4) analyzed the composition of six samples of LPG collected from different retail dealers in the Houston, Texas area; the results are shown in table 3. These measurements indicate the predominant use of propane with some ethane in these LPG samples.

Liquefied petroleum gas is produced principally in the same regions of the country where natural gas is produced. Table 4 gives a listing of total gallons of propane and LPG mixture produced by region and by state together with estimates of domestic consumption in ranges and space heaters. These data were developed from Cannon (23) and from U.S. Census Bureau statistics (24). Just as with natural gas (5), the overwhelming majority of LPG is produced in the west south central region (Texas, Oklahoma, Louisiana, Arkansas).

Radon Concentration,  
Well Location, Depth  
and Pressure, Seasonal  
Variations

See Ref. (1-6)  
for Radon in  
Natural Gas

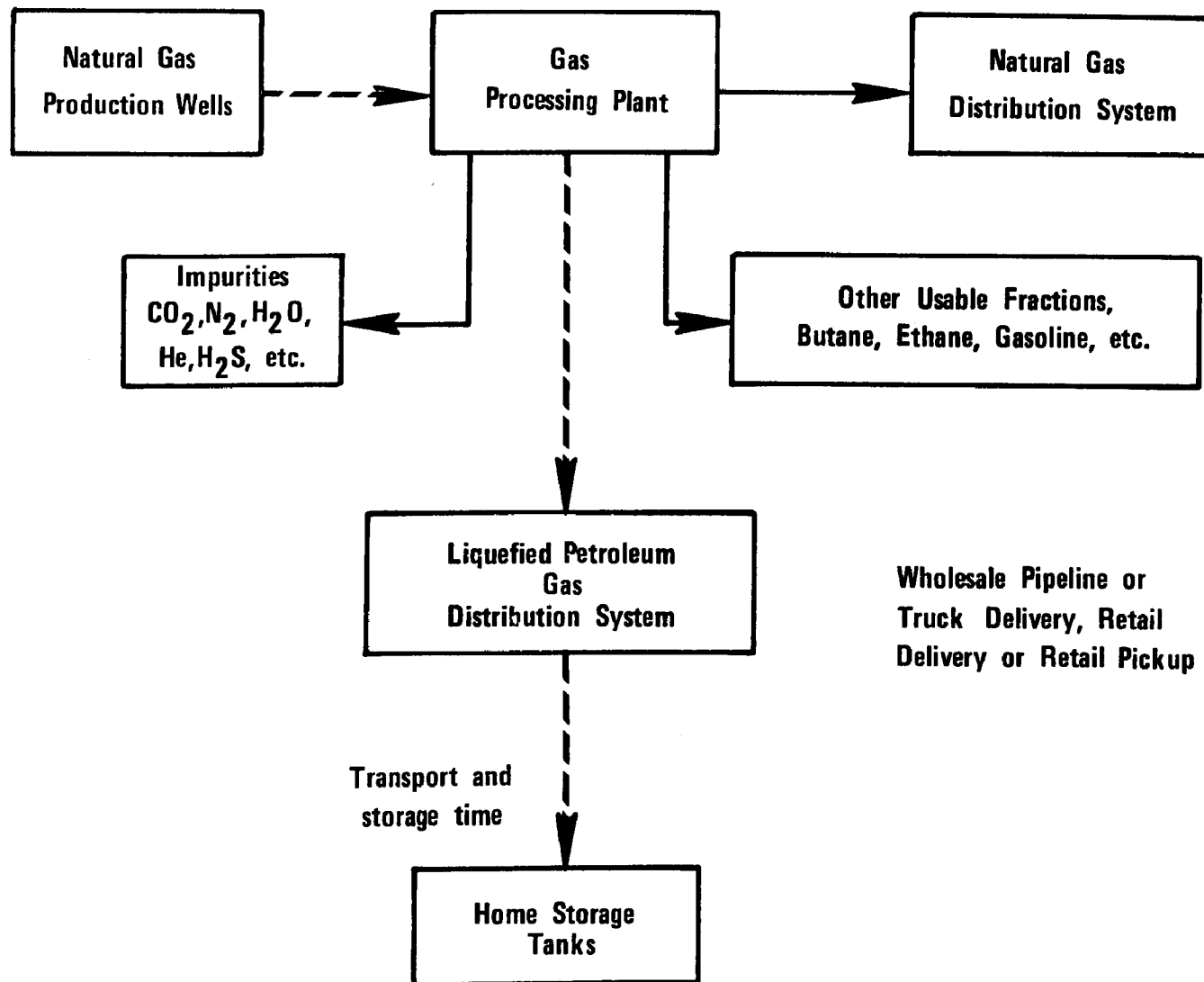


Figure 2. GENERAL FLOW DIAGRAM FOR PRODUCTION AND DISTRIBUTION OF NATURAL GAS AND LPG.

Table 2  
U.S. Production of Selected Gas Liquids from Gas Processing  
Plants and Central Fractionators (23)

Product	Production - 1971 (10 <sup>9</sup> gal/yr)*
Ethane	3.67
Propane	8.48
Isobutane	1.43
Normal and unsplit butane	3.81
Natural gasoline	4.28

\*1 gallon of liquid propane yields 1.25 m<sup>3</sup> of gas at STP.

Table 3  
Liquefied Petroleum Gas (4)

Component	Percent of LPG*	Boiling Point (°C)
Methane	0.2-2.0	-161.5
Ethane	2.4-9.5	- 88.3
Radon	---	- 61.8
Propane	88-96	- 42.2
Butane	0.5-1.5	- 0.5

\*Measured in a survey of six retail sources of LPG in the Houston, Texas area.



Table 4

## Gas Processing Plants, Propane Production and Estimated Consumption in Ranges and Space Heaters by States and Regions

Division and State	Gas Processing Plants	Propane Production	Use (a)
United States (b)	805	$8.48 \times 10^9$	$2.24 \times 10^9$
Percentage of United States			
<u>New England</u>	0	0	3.5
Connecticut	0	0	0.8
Maine, New Hampshire, Vermont	0	0	1.5
Massachusetts	0	0	1.0
Rhode Island	0	0	0.2
<u>Middle Atlantic</u>	0.2	.02	7.0
New Jersey	0	0	0.9
New York	0	0	4.0
Pennsylvania	0.2	.02	2.1
<u>East North Central</u>	0.7	3.3	20.2
Illinois	0.1	2.8	5.4
Indiana	0	0	3.8
Michigan	0.6	0.5	3.5
Ohio	0	0	3.1
Wisconsin	0	0	4.4
<u>West North Central</u>	4.2	7.0	20.7
Iowa	0	0	1.3
Kansas	3.5	6.2	2.9
Minnesota	0	0	4.2
Missouri	0	0	8.1
Nebraska	0.2	0.2	2.6
North Dakota	0.4	0.3	1.1
South Dakota	0.1	0.3	0.5
<u>South Atlantic</u>	0.6	1.2	13.8
Delaware	0	0	0.2
Florida	0.1	0.30	3.5
Georgia	0	0	4.0
Maryland, District of Columbia	0	0	1.1
North Carolina	0	0	2.2
South Carolina	0	0	1.3
Virginia	0	0	1.1
West Virginia	0.5	0.90	0.4
<u>East South Central</u>	1.6	1.4	9.8
Alabama	0.1	0	3.0
Kentucky	0.4	1.2	2.7
Mississippi	1.1	0.2	2.7
Tennessee	0	0	1.4
<u>West South Central</u>	75.4	74.8	12.9
Arkansas	0.4	0.3	2.8
Louisiana	16.9	16.9	1.4
Oklahoma	11.6	10.8	3.0
Texas	46.5	46.8	5.7
<u>Mountain</u>	10.5	8.3	6.6
Arizona	0.1	0	0.6
Colorado	1.4	0.9	1.9
Idaho	0	0	0.5
Montana	0.7	0.2	0.4
Nevada	0	0.0	0.6
New Mexico	4.5	3.5	1.1
Utah	0.4	0.4	0.4
Wyoming	3.4	3.3	0.6
<u>Pacific</u>	6.8	3.9	5.5
Alaska	0.2	0.1	0.1
California	6.6	3.8	3.9
Hawaii	0	0	0.1
Oregon	0	0	0.7
Washington	0	0	0.7

(a) Note that this usage is for domestic ranges and space heaters only. It does not include other domestic appliances, commercial or industrial uses.

(b) Propane production and consumption in gallons per year.

The factors which affect the  $^{222}\text{Rn}$  content of propane as produced in the gas processing plant include the  $^{222}\text{Rn}$  content in the wellhead gas, the transit time from wellhead to processing plant, the makeup of the inlet gas, and the type of processing.

Gesell (4) has surveyed nine gas processing plants in the U.S. The locations of these surveys are shown by the circles in figure 3. Also shown in figure 3 are the locations of gas processing plants in the United States. The survey included measurement of the  $^{222}\text{Rn}$  content of the inlet stream(s), some of the products and the residue (outlet) gas. The results of this survey are given in table 5. It can be seen that  $^{222}\text{Rn}$  concentrations in the inlet gases are in the expected range for wellhead gases (see table 1). Furthermore, the  $^{222}\text{Rn}$  concentrations in the propane are much higher than those in the inlet natural gas, ranging up to 1119 pCi/liter. Figure 4 shows a plot of the  $^{222}\text{Rn}$  concentration in propane vs. the  $^{222}\text{Rn}$  concentration in the inlet gas for eight of the nine plants (the propane sample was lost from one plant). A least squares fit to the data gave a slope of 8.2 and a linear correlation coefficient of 0.94. Thus for a variety of processing plants, the average concentration of  $^{222}\text{Rn}$  in propane is about eight times the concentration in the inlet gas.

Bernhardt<sup>6</sup> reported that a radon concentration of 56 pCi/liter in the inlet natural gas was increased to 1100 pCi/liter in the propane fraction at a San Juan gas processing plant which indicates an increase in radon concentration by a factor of nearly 20. In addition, Fries and Kilgren (22) report data for a plant in California, which indicate a concentration factor for radon of about 14. The differences in these factors may be attributed to differences in hydrocarbon make-up of the inlet natural gas or to differences in plant operations and the composition of the LPG product, i.e., the relative quantities of ethane, propane, and butane. The factor of 20 reported by Bernhardt<sup>6</sup> was for a basically propane stream. The butane stream for this facility had a radon concentration factor of 7. The factor of 14 determined by Fries and Kilgren (22) appears to be for a mixed propane-butane stream. When these factors are combined with the data from Gesell (4), the average concentration of radon in propane is about  $10 \pm 5$  times that in natural gas at one standard deviation.

It should be noted that the study by Fries and Kilgren (22) indicated significant external gamma radiation levels may be found within the gas processing plant, arising from the short-lived radon daughters. However, these radiation levels were quite localized in particular process equipment. In some pieces of equipment, measureable quantities of long-lived radon daughters were also found. Fries and Kilgren (22) concluded that these quantities did not present a radiation hazard to plant maintenance personnel. Nevertheless, several precautions were recommended. Namely, process equipment with significant external radiation should be shut down

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<sup>6</sup>Bernhardt, D.E., "Radon in natural gas products--San Juan Plant," memorandum to C.L. Weaver, August 31, 1973.

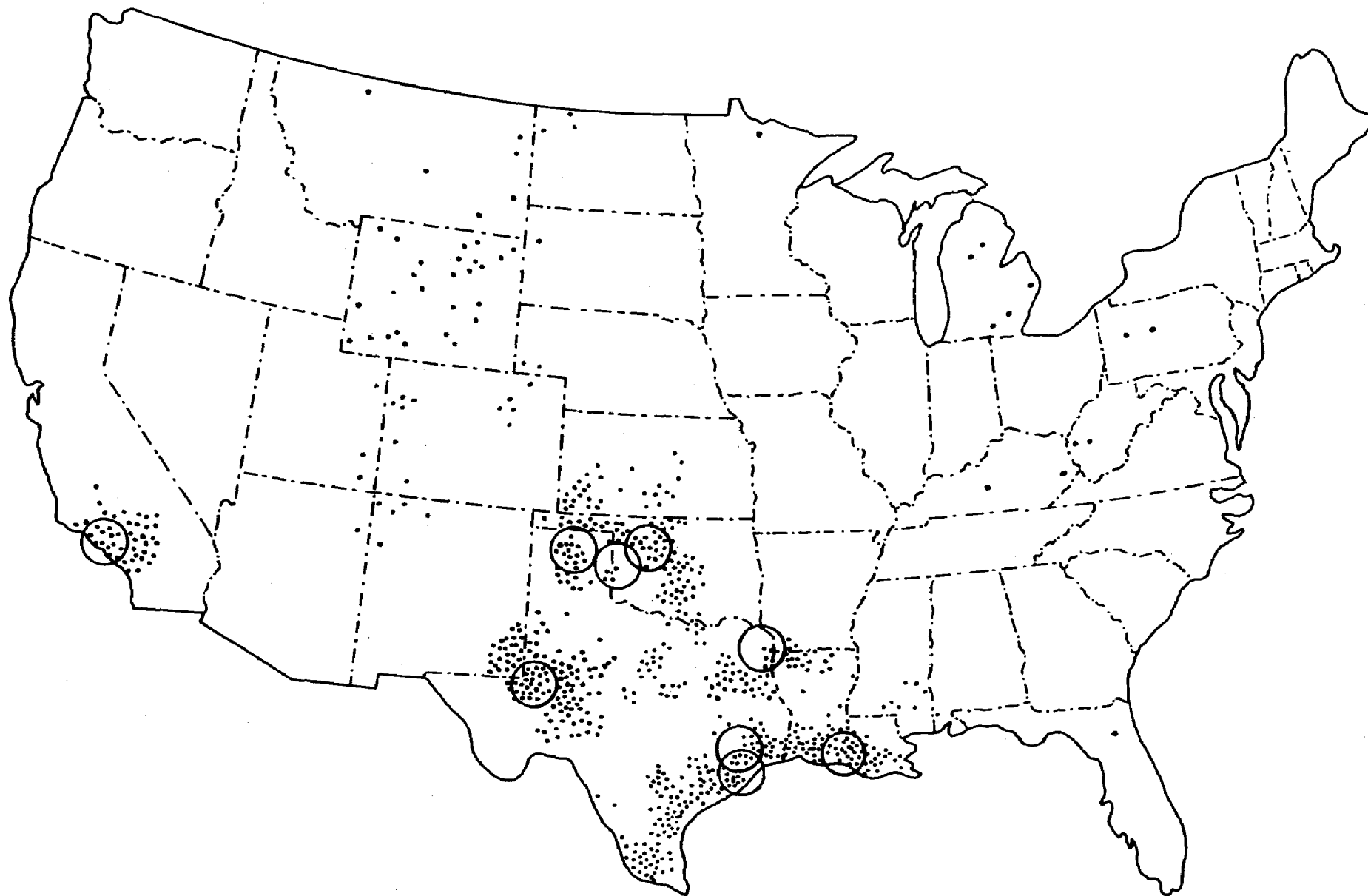


Figure 3. DISTRIBUTION OF GAS PROCESSING PLANTS IN THE CONTIGUOUS UNITED STATES. EACH SMALL DOT REPRESENTS ONE GAS PROCESSING PLANT AND EACH CIRCLE REPRESENTS A SURVEYED GAS PLANT (4)

Table 5

Radon-222 Concentration Measured in Gas Plant Processing  
Streams (pCi/liter at STP) (4)

Source	Plant Code								
	1	2	3	4	5	6	7	8	9
Inlet gas	10.5	26.3	78.4	118.5	35.4	18.4	2.9 5.0 5.0	1.0 1.5	23.6 41.3 17.3 28.6
Ethane	368.2	---	---	---	97.7 <sup>(a)</sup>	306.7	---	---	---
Propane	177.6	(b)	386.1	1,119.0	237.7	56.8	61.5	9.0	213.3
Butane	1.5	---	---	---	---	---	---	---	---
Sales gas (methane)	0.6	13.6	3.2	93.6	3.1	0.9	1.2 1.0	0.9 0.5	---

(a) Mixture of ethane and propane.

(b) Sample lost.

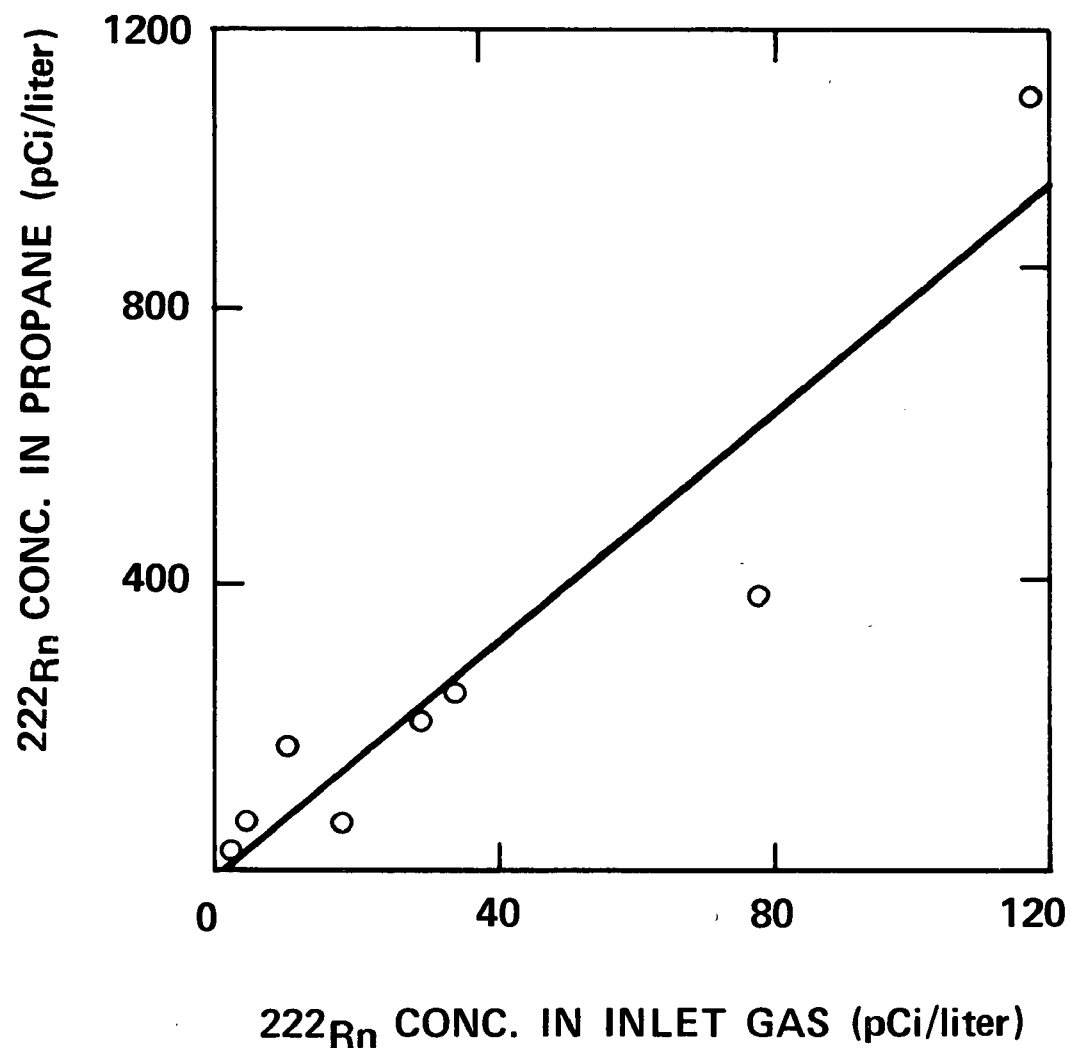


Figure 4. LEAST SQUARES FIT OF THE RADON-222 CONCENTRATION IN PROPANE VS. THE RADON-222 CONCENTRATION IN THE INLET GAS (4)

at least three hours before disassembly to allow decay of short-lived nuclides. Then for equipment requiring brushing or scraping, gloves and a face mask should be worn to prevent inhalation or ingestion of long-lived radon daughters. They furthermore recommended that decay time for radon in LPG be maximized by selling the oldest product first.

In addition to propane produced in domestic gas processing plants, propane is also produced by oil refineries and is imported. For the year ending in November 1973, gas processing plants produced approximately  $9 \times 10^9$  gallons of propane, refineries produced  $\sim 4 \times 10^9$  gallons and  $\sim 1 \times 10^9$  gallons were imported (25).

Refinery propane is not expected to contain high levels of radon-222, but concentrations should be checked. No information on radon-222 content of imported propane is available. However, radon levels should be low since shipping time would reduce the radon-222 content from the level at manufacture.

#### At the Retail Level

Although a knowledge of radon in wellhead gas and radon in LPG at the point of manufacture is basic to an understanding of the  $^{222}\text{Rn}$  in LPG problem, it does not provide direct information on radon levels to be expected in LPG as purchased by or delivered to the consumer. Transportation and storage networks are complex and the relatively short half-life of  $^{222}\text{Rn}$  can result in substantial decay of the activity. An indication of the complexity of the supply, transit, storage and delivery system is given by Gesell (4) who has studied the system for Houston, Texas. A flow diagram for a typical retailer is given in figure 5. Distribution systems in other areas may be more or less complex. For example, rail shipment is utilized for markets far from production, and for markets very near to production, the retail delivery truck may pick up directly from the gas plant on the day of delivery. Figure 6 shows a hypothetical delivery system which includes the various known sources, storage, transport, and delivery modes. It is clear from this figure that, because of the unknown storage and transport times, the best information on  $^{222}\text{Rn}$  levels in LPG at the consumer level would be obtained at the retail dealer level.

Gesell (3,4,16,17) has conducted a year-long weekly survey of LPG at the retail level in Houston, Texas, as well as a grab sample survey of retailers in 14 southern and southwestern states. He used the Lucas (26) method for radon analysis.

The year-long study of seven retailers was performed to examine any seasonal trends in radon concentration. The results of this survey (4) are given in table 6. Also included in table 6 are column and row averages. Retailer No. 7 was consistently low, so sampling was discontinued after six months. For purposes of the averages, retailer No. 7 was taken as 0.5 pCi/liter for February through June. The data are characterized

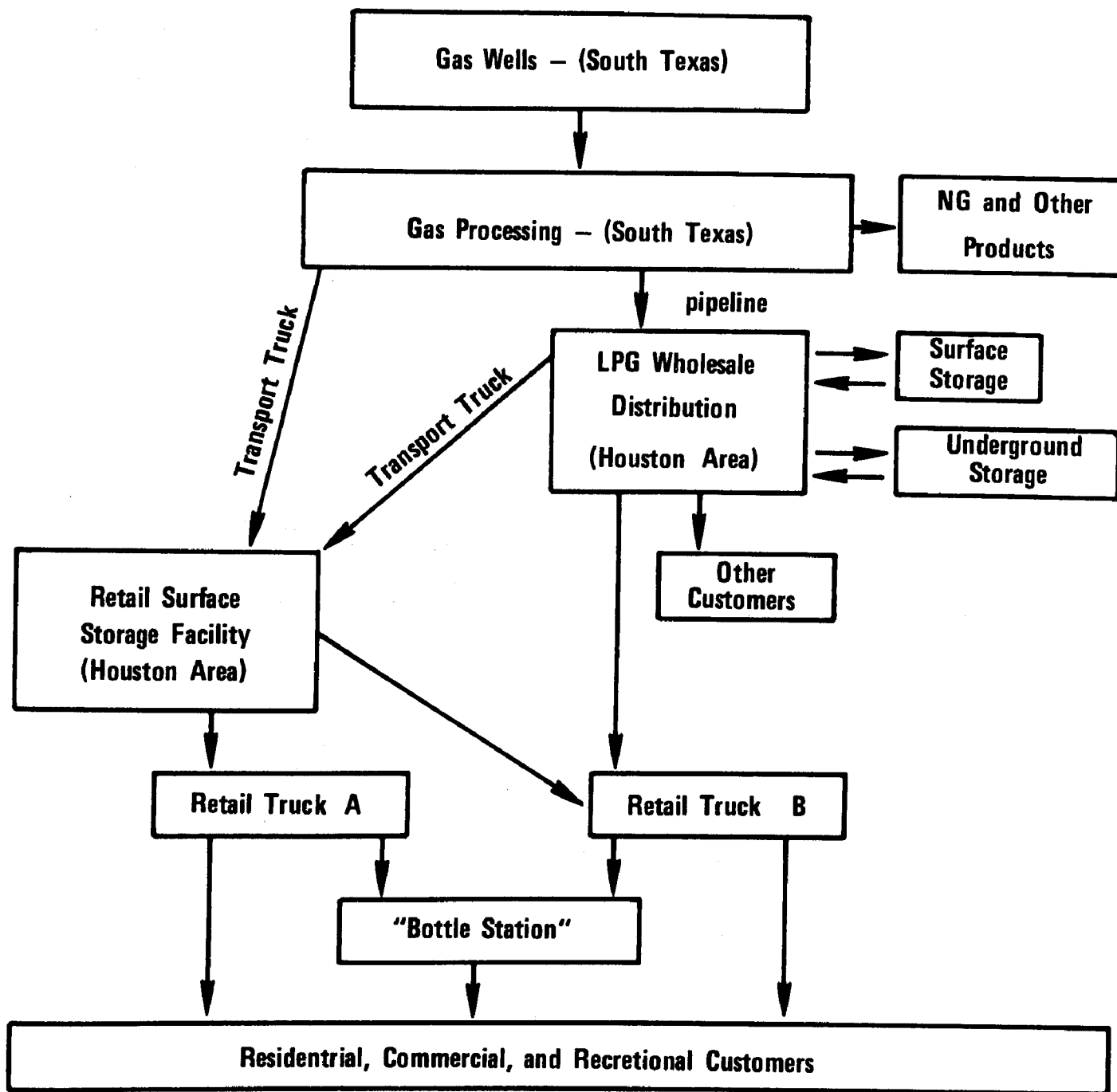


Figure 5. Flow Diagram from Wellhead to Consumer of LPG in the Houston, Texas Area (4)

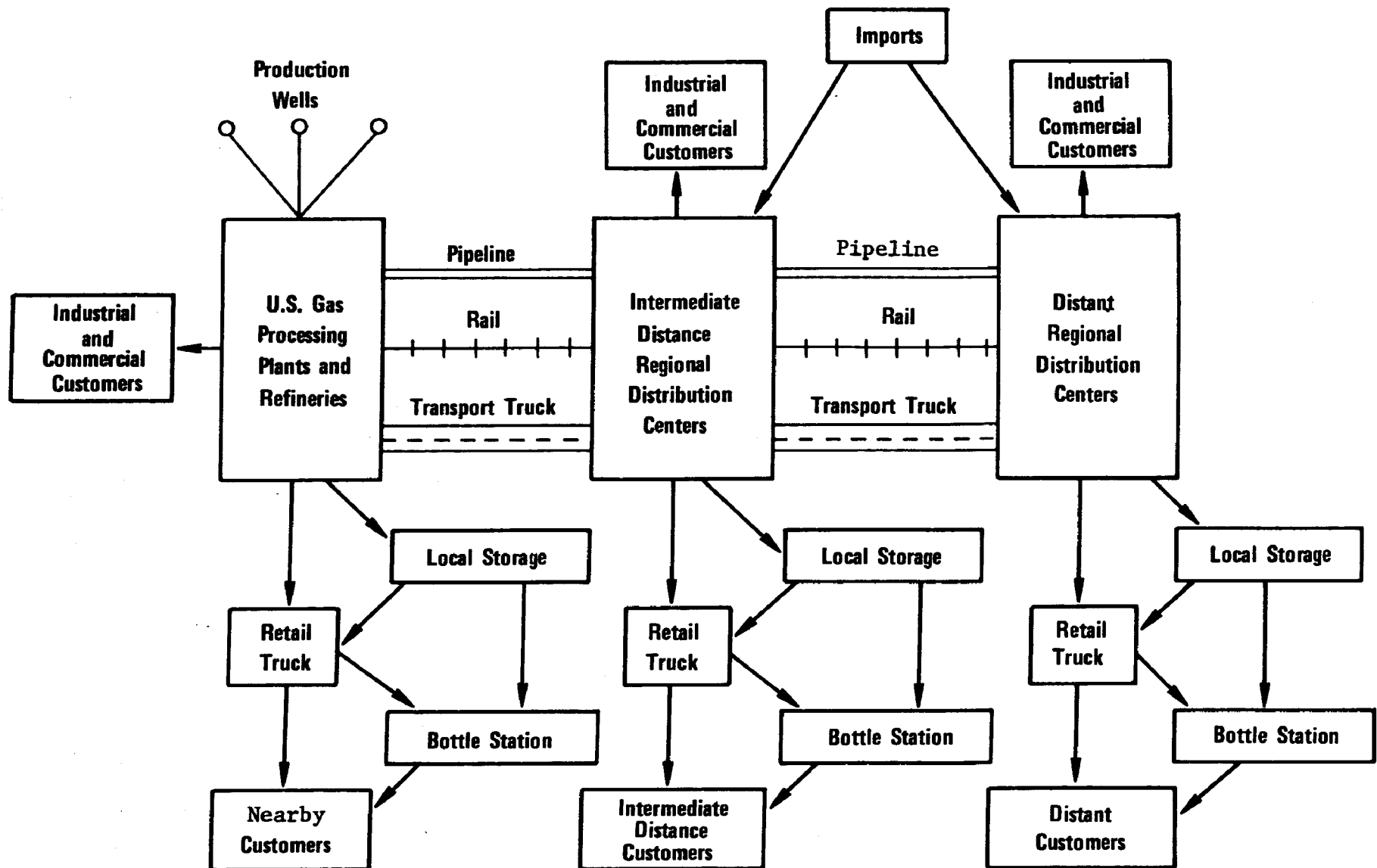


Figure 6. HYPOTHETICAL FLOW DIAGRAM FOR LPG FROM WELLHEAD TO MARKET.



Table 6 <sup>222</sup>Rn Concentration in Gaseous Retail LPG (pCi/liter at STP) in the Houston, Texas Area (4)

Week of	Retailer Code Number							Average
	1	2	3	4	5	6	7	
July 23 1972	9.7	5.8	8.8	141.4	79.2	316.3	<0.5	80.2
July 30	9.0(a)	5.3(a)	3.2(a)	120.5	35.9(a)	150.4(a)	<0.5(a)	46.4
Aug 6	23.9(b)	77.9(b)	84.1(b)	137.4	37.5(b)	181.9(b)	<0.5(b)	77.7
Aug 13	77.0(b)	46.8(b)	20.3(b)	140.4	35.0(b)	214.1(b)	<0.5(b)	76.2
Aug 20	106.3(b)	55.7(b)	42.2(b)	142.5	40.2(b)	236.5(b)	<0.5(b)	89.1
Aug 27	45.3	68.2	19.9	221.4	17.5	155.3	3.9	76.0
Sept 10	28.6	22.8	6.2	70.2	14.3	155.6	0.7	42.6
Sept 17	30.9	80.6	2.1	110.1	18.0	139.0	<0.5	54.5
Sept 24	23.7	24.1	21.8	51.0	32.9	153.2	0.8	43.9
Oct 1	67.4	7.4	13.6	86.8	67.8	133.8	5.5	54.6
Oct 8	5.4	31.3	35.5	11.8	14.1	17.0	4.1	17.0
Oct 15	12.9	7.3	24.8	149.4	14.0	306.5	2.9	74.0
Oct 22	3.8	95.1	46.1	81.3	22.8	151.6	4.9	57.9
Oct 29	11.9	101.1	31.1	100.3	54.1	211.1	6.5	73.7
Nov 5	0.7	2.4	71.0	71.1	15.9	173.1	0.7	47.8
Nov 12	72.8	100.7	33.2	55.8	23.8	277.1	1.2	80.6
Nov 19	8.9	1.0	35.9	65.0	17.3	177.4	7.4	44.7
Nov 26	1.4	8.1	114.0	2.2	3.4	215.4	7.6	50.3
Dec 3	0.6	215.1	10.6	11.9	1.8	209.9	1.5	64.5
Dec 10	<0.5	2.8	55.4	<0.5	1.0	170.4	6.7	33.9
Dec 17	<0.5	2.2	33.7	0.8	2.1	202.1	3.6	35.0
Dec 24	0.9	<0.5	5.6	<0.5	1.0	178.2	3.7	27.2
Dec 31	28.8	2.6	22.5	0.7	42.5	121.3	1.5	31.4
Jan 7 1973	6.3	1.1	---	<0.5	15.5	147.7	---	26.2
Jan 14	4.3	0.8	<0.5	0.7	3.1	133.2	---	20.4
Jan 21	2.1	1.2	4.3	<0.5	2.2	157.9	---	24.1
Jan 28	0.8	130.0	1.9	0.7	145.7	115.6	---	56.4
Feb 4	5.5	7.2	0.7	16.4	6.8	123.8	---	23.0
Feb 11	7.7	41.0	1.7	15.2	22.5	194.5	---	40.4
Feb 18	197.2	12.3	63.4	6.2	4.8	184.0	---	66.9
Feb 25	11.1	83.6	19.0	8.6	8.5	169.4	---	43.0
Mar 4	109.8	28.6	5.3	20.3	1.9	147.8	---	44.9
Mar 12	9.6	6.0	23.2	52.1	5.0	274.4	---	53.0
Mar 26	122.7	84.9	3.0	48.0	3.7	195.0	---	65.4
Apr 1	109.6	29.8	8.6	88.6	11.6	124.0	---	53.2
Apr 8	120.1	88.0	3.1	91.9	52.9	120.2	---	68.1
Apr 15	89.7	58.1	1.9	102.2	27.9	215.6	---	70.8
Apr 22	81.4	63.0	7.9	46.7	26.0	148.1	---	53.4
Apr 29	128.0	42.5	8.5	59.2	32.6	138.8	---	58.6
May 6	62.9	14.4	---	107.2	46.9	146.0	---	58.8
May 13	33.5	8.3	58.7	69.5	12.3	231.6	---	59.2
May 20	58.5	4.6	10.6	51.4	25.5	143.6	---	42.1
May 27	160.2	16.0	69.8	4.8	70.4	191.5	---	73.3
June 3	64.8	98.7	3.0	30.1	12.1	105.9	---	45.0
June 17	148.8	5.5	66.6	20.1	17.1	262.9	---	74.5
June 24	131.7	8.3	---	27.1	41.0	152.4	---	61.1
Average	49.2	38.5	25.9	43.4	26.7	170.4	1.8	50.8

(a) Average of two samples.

(b) Average of three samples.

by a large amount of scatter within and among dealers. This scatter is not too surprising in view of the previously discussed factors that control the radon in LPG.

Of the seven retailers sampled, No. 6 was on the average much higher in  $^{222}\text{Rn}$  concentration than the others and No. 7 was on the average much lower than the others. Inquiry revealed that retail dealership No. 6 obtains its LPG directly from a gas processing plant and that retail dealership No. 7 receives its LPG from an underground storage well. The other five retailers obtain their LPG from various wholesale distributors. Thus, the radon concentrations in the LPG from the several retailers are consistent with their individual supply practices which govern storage decay times for radon-222.

Figure 7 shows a second order polynomial fit of the monthly average data from the seven sampled Houston area dealers. Gesell (4) interprets the pronounced dip in the activity during the winter months as being due to the higher winter consumption rate and consequent appearance on the market of previously stored LPG. This interpretation is consistent with the known practices for manufacture, storage, and distribution of LPG. The production of LPG is maintained at a relatively constant level, with production going to storage or consumption according to demand.

It is worthwhile noting that the average concentration during the heating season is approximately one-half of the concentration during the summer months. Gesell (4) notes that the existing practice (at least in the Houston, Texas area) of storing during the summer and using from storage during the winter serves to reduce the exposure of those who utilize LPG in unvented heating from what it would be if production were simply adjusted to demand.

Gesell's (4) grab sample survey of retailers in southern and southwestern states included LPG samples at the locations indicated in figure 8. The results for these LPG samples are presented individually in table 7, and are grouped by region in figure 9. In each region of figure 9, the top number is the maximum concentration observed, the middle number is the average of all samples in the region and the bottom number is the lowest concentration observed. The most remarkable feature of this distribution is difference in radon concentrations in the Eastern and Western parts of the United States. Two major factors affect the concentration of radon in retail LPG, concentration at the point of manufacture and the time required to deliver the gas to the consumer. The low concentrations found in the two eastern-most regions could potentially be explained in terms of longer transport and storage times since these areas are fairly far from any major production. The low concentrations found in Louisiana and Arkansas, in view of the large amount of gas processing in Louisiana, invite interpretation in terms of low concentrations in the sources. This interpretation is consistent with the low  $^{222}\text{Rn}$  concentrations found in the gas processing plant surveyed in Louisiana and the reported low radon concentration in Gulf Coast gas (5,18).

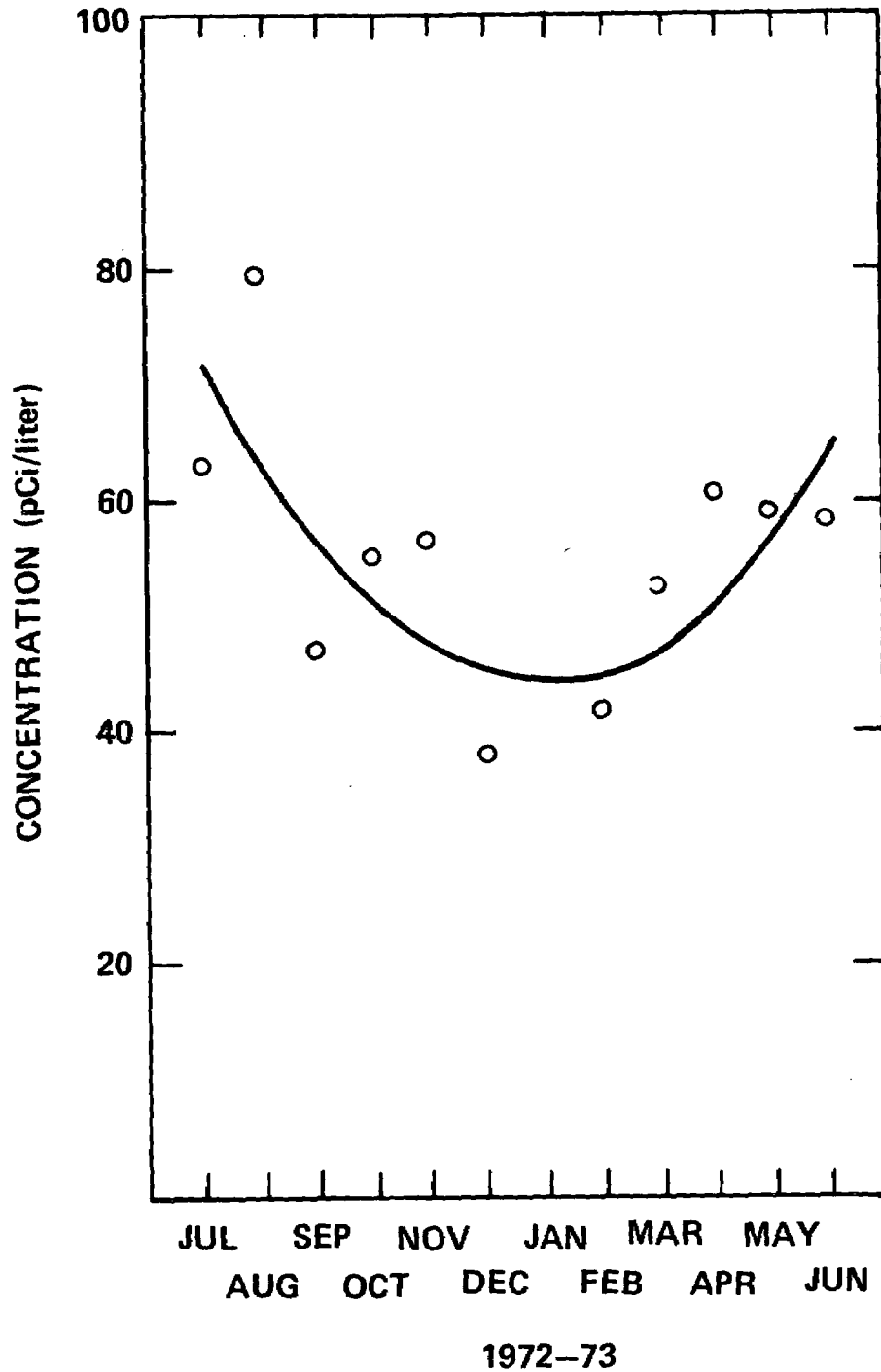


Figure 7. SECOND ORDER POLYNOMIAL FIT OF THE MONTHLY AVERAGE VALUES OF  $^{222}\text{Rn}$  CONCENTRATION FOUND IN WEEKLY HOUSTON, TEXAS, AREA RETAIL LPG SAMPLES (4)

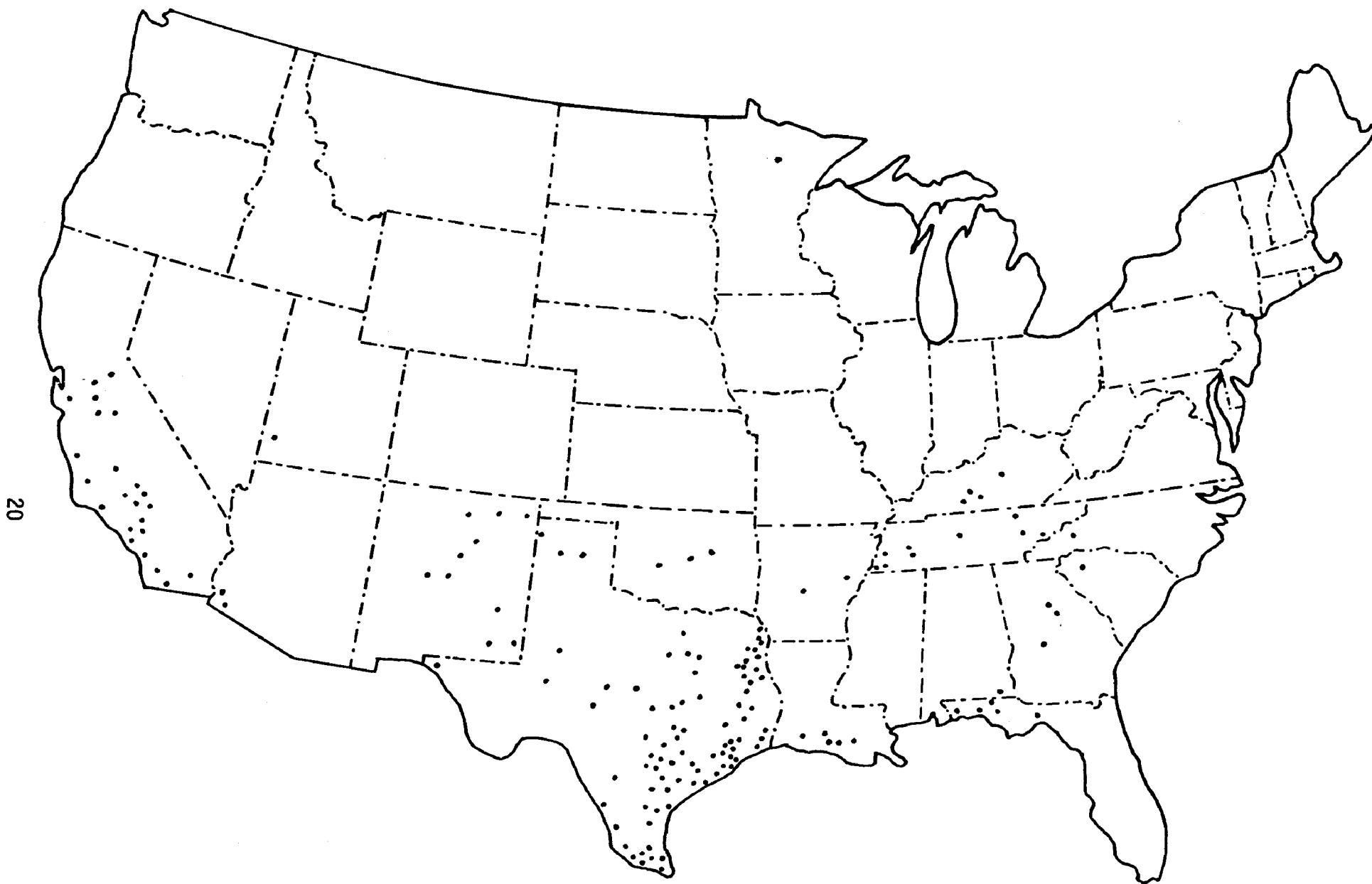


Figure 8. DISTRIBUTION OF RETAIL LPG SAMPLE SITES (4)

Table 7. Radon-222 Concentration in Retail LPG Samples (at STP) (4)

Location	Date	Radon-222 Concentration (pCi/liter)	Location	Date	Radon-222 Concentration (pCi/liter)
ALABAMA			GEORGIA		
Dothan	08/02/73	<0.5	Athens	08/03/73	1.1
ARIZONA			Athens	08/03/73	0.6
Yuma	08/30/73	6.8	Macon	08/03/73	<0.5
Yuma	08/30/73	34.1	KENTUCKY		
ARKANSAS			Bowling Green	08/07/73	2.7
N Little Rock	08/08/73	1.8	Bowling Green	08/07/73	<0.5
Forrest City	08/08/73	1.6	Georgetown	08/07/73	<0.5
			Glasgow	08/07/73	<0.5
CALIFORNIA			Lebanon	08/07/73	0.8
Angels Camp	08/24/73	7.9	LOUISIANA		
Bakersfield	08/28/73	55.4	Jeanerette	07/31/73	4.8
Bakersfield	08/28/73	15.1	Jennings	07/30/73	<0.5
Bakersfield	08/28/73	133.8	Lafayette	07/31/73	3.8
Carlsbad	08/22/73	84.0	Lafayette	07/31/73	<0.5
Delano	08/27/73	102.0	New Iberia	07/31/73	13.4
El Cajon	08/22/73	24.5	NEW MEXICO		
El Centro	08/30/73	3.4	Albuquerque	07/24/73	1.8
El Toro	08/22/73	45.6	Albuquerque	07/24/73	101.3
El Toro	08/23/73	41.9	Artesia	07/24/73	43.5
Escondido	08/28/73	27.9	Des Moines	07/20/73	120.2
Fresno	07/27/73	22.3	Espanola	07/23/73	7.4
Santa Barbara	08/23/73	47.0	Hobbs	07/25/73	62.9
Stockton	08/25/73	3.3	Las Vegas	07/23/73	<0.5
Visalia	08/27/73	710.8	Raton	07/23/73	57.1
King City	08/23/73	1049.3	Roswell	07/24/73	36.0
Lancaster	08/28/73	8.5	Socorro	07/24/73	3.5
Livingston	08/27/73	2.0	Taos	07/23/73	4.8
Los Angeles	08/22/73	144.2	NORTH CAROLINA		
Los Angeles	08/22/73	663.1	Asheville	08/06/73	1.0
Los Angeles	08/22/73	92.8	OKLAHOMA		
Los Angeles	08/22/73	88.0	Bristow	07/18/73	0.9
Los Gatos	08/24/73	5.9	Oklahoma City	07/18/73	10.1
Mercedes	08/27/73	283.5	Tulsa	07/17/73	2.7
Modesto	08/27/73	269.7	SOUTH CAROLINA		
Paso Robles	08/23/73	5.4	Greenville	08/03/73	7.0
FLORIDA			TENNESSEE		
Bonifay	08/02/73	<0.5	Covington	08/08/73	1.1
Chipley	08/02/73	1.3	Jackson	08/08/73	2.5
De Funiak			TEXAS		
Springs	08/02/73	<0.5	Henderson	04/12/73	4.0
Pensacola	08/01/73	3.8	Henderson	04/13/73	54.4
Tallahassee	08/02/73	1.2	Hitchcock	12/18/72	<0.5
TENNESSEE			Huntsville	07/16/73	0.7
Jackson	08/08/73	<0.5	Jefferson	04/13/73	118.5
Knoxville	08/06/73	1.1	Johnson City	05/11/73	10.7
La Follette	08/08/73	10.3	Katy	05/10/73	3.0
Memphis	08/08/73	<0.5	Kenedy	07/12/73	5.3
Memphis	08/07/73	<0.5	Kilgore	04/12/73	77.7
Nashville	08/06/73	0.6	Killeen	07/26/73	2.5
Sevierville			Kingsville	01/19/73	333.9
TEXAS			Lamesa	07/25/73	2.9
Alice	01/19/73	221.5	Lampasas	07/26/73	1.3
Angleton	01/18/73	21.3	Laredo	07/13/75	44.3
Angleton	01/18/73	3.2	Linden	04/13/73	231.7
Arlington	07/17/73	24.0	Longview	04/13/73	0.8
Atlanta	04/13/73	66.7	Livingston	04/12/73	4.5
Austin	05/11/73	51.0	Longview	04/13/73	14.9
Austin	05/11/73	13.7	Lufkin	04/12/73	3.9
Austin	05/11/73	45.2	Lufkin	04/12/73	85.0
Ballinger	07/26/73	3.7	Lufkin	04/12/73	21.0
Bay City	01/18/73	6.4	Luling	05/10/73	37.3
Bay City	01/18/73	269.1	Marshall	04/13/73	4.3
Beaumont	12/18/72	8.0	McAllen	02/23/73	469.2
Beaumont	12/18/72	82.5	Midland	07/25/73	7.8
Brownsville	02/23/73	294.5	Nacogdoches	04/12/73	21.3
Brownsville	02/23/73	21.3	Port Arthur	01/19/73	95.2
Brownwood	07/26/73	13.2	Port Arthur	12/18/72	286.8
Borger	07/19/73	191.3	Robstown	02/22/73	6.7
Carthage	04/13/73	112.6	San Angelo	07/25/73	26.6
Columbus	05/10/73	8.7	San Antonio	05/11/73	1240.5
Corpus Christi	01/19/73	65.4	San Antonio	05/11/73	23.8
Corpus Christi	01/19/73	237.1	San Antonio	05/11/73	7.6
Corsicana	07/16/73	4.7	San Antonio	05/11/73	9.2
Cortulla	07/13/73	2.3	San Benito	02/23/73	28.9
Dallas	07/16/73	14.2	San Benito	02/23/73	30.4
Dallas	07/16/73	3.5	San Benito	02/23/73	554.6
Denton	07/17/73	20.0	Seguin	05/10/73	4.9
Dumas	07/19/73	225.2	Sinton	02/22/73	46.5
Ennis	07/16/73	1.5	Temple	07/26/73	2.3
El Paso	08/30/73	6.8	Texarkana	04/13/73	58.4
Fredericksburg	05/11/73	2.1	Texarkana	04/13/73	22.5
Galveston	12/18/72	9.1	Texline	07/20/73	292.3
George West	07/12/73	1.8	Victoria	01/19/73	38.2
Goliad	07/12/73	104.9	Vidor	12/18/72	4.8
Harlingen	02/23/73	350.9	Weimar	05/10/73	6.8
Harlingen	02/23/73	185.5	Weslaco	02/23/73	168.0
			Zapata	07/13/73	138.5

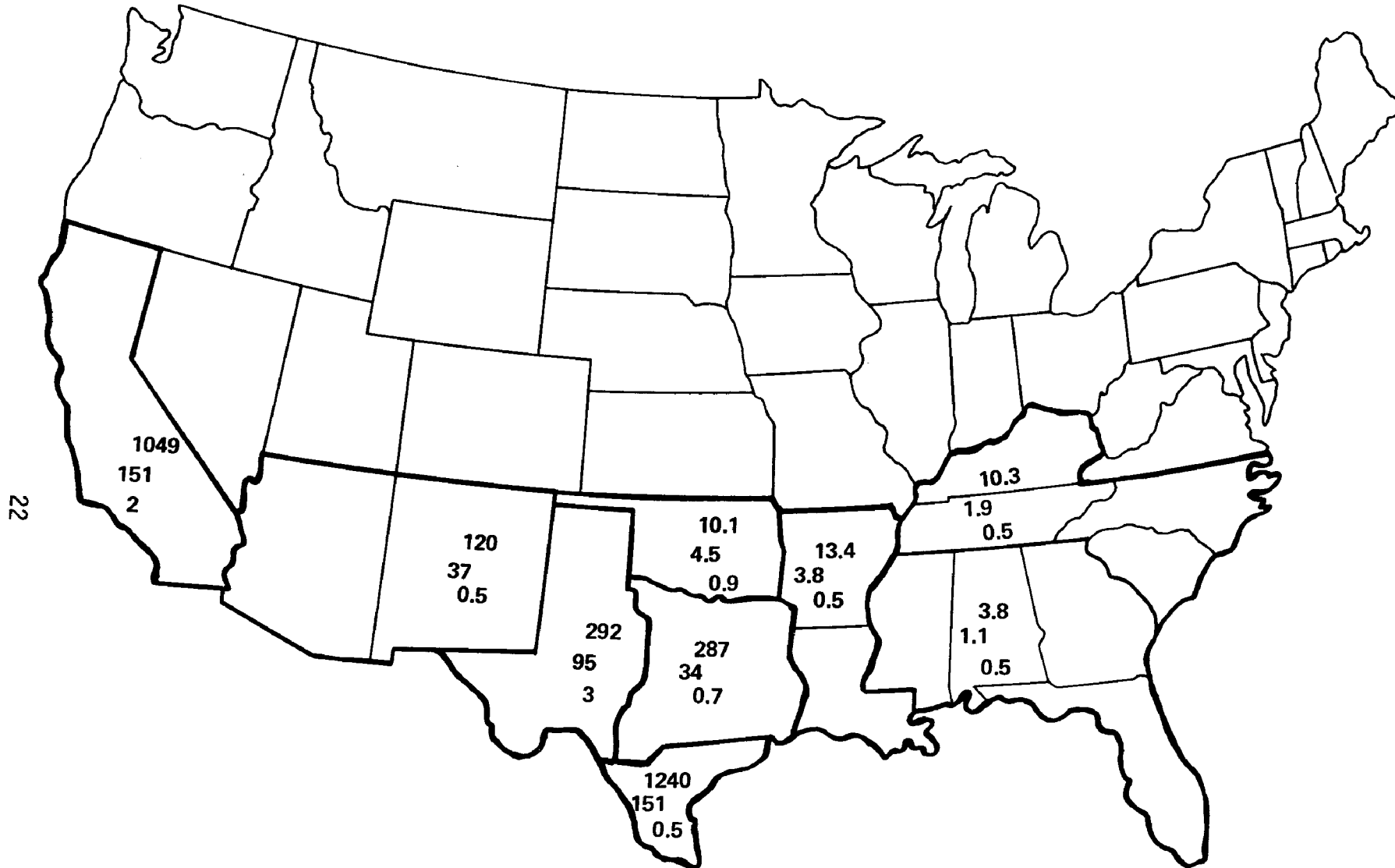


Figure 9. DISTRIBUTION BY REGION OF  $^{222}\text{Rn}$  CONCENTRATIONS FOUND IN RETAIL LPG (pCi/liter) IN EACH REGION. THE TOP NUMBER IS THE MAXIMUM VALUE FOUND, THE LOWER NUMBER IS THE LOWEST VALUE FOUND AND THE MIDDLE NUMBER IS THE MEAN OF ALL SAMPLES IN THE REGION. (4)

Gesell (6) examined the variation of  $^{222}\text{Rn}$  in LPG with distance from a major gas processing area by dividing the data from table 7 into three categories according to whether the samples were taken less than 25 miles, between 25 and 200 miles, or greater than two-hundred miles from a major gas processing area. The results are shown in table 8. The differences were tested by the non-parametric Wilcoxon two sample test (27). The mean for the 25-200 mile category was found to be significantly less ( $P < 0.1$ ) than the mean for the  $<25$  mile category and the mean for the  $>200$  mile category was found to be significantly less ( $P < 0.01$ ) than the mean for the 25-200 mile category. The results suggest that the unsampled northern states would tend to have lower radon concentrations than the sampled states since they are, with few exceptions farther from the major gas processing areas than the sampled states. This suggestion assumes that an insignificant amount of Canadian LPG enters the northern states market.

TABLE 8

RELATIONSHIP BETWEEN  $^{222}\text{Rn}$  CONCENTRATION IN LPG  
AND DISTANCE FROM A MAJOR GAS PROCESSING AREA (4).

Distance (miles)	Number of Samples	Average Concentration (pCi/liter)
<25	103	87.8
25-200	23	63.7
>200	31	45.6

Actually, about 7 percent of the annual LPG consumed in the United States is imported, mainly from Canada. Most of this Canadian LPG is used in the states nearest to Canada. Since the Canadian LPG has a generally higher radon-222 content than that from the United States (as shown in table 1), then the average radon content of LPG consumed in the northern states might be higher than would be predicted for LPG from the United States alone. However, no specific information was available for this study on radon concentrations in LPG in the northern states. Therefore, the contribution of radon from Canadian LPG was not included specifically in this study, although the general significance of Canadian LPG will be considered in the overall interpretation of data (see discussion section).

Gesell (4) also examined the variation of  $^{222}\text{Rn}$  in LPG with the type of container (retail storage tank, truck or small tank) from which the sample was drawn. Only Texas was included in this examination because in the other regions very few samples were obtained from trucks. Retailers fill bottles brought to their locations from the same, generally large (>10,000 gal.) tanks from which they fill their own delivery trucks, from the delivery trucks themselves, or from special "bottle stations" which are supplied by small (generally <2,000 gal.) tanks. The "bottle stations" are typically filled from the retail trucks. Thus, the LPG in the bottle stations would tend to be older, on the average, than the LPG in the trucks and that in the trucks slightly older than the LPG in the large storage tanks. The Texas samples (non-Houston) were categorized and averaged and the results are presented in table 9. The mean of the truck samples, although smaller than that of the large tank samples was not significantly smaller. The mean of the small tank samples is significantly smaller ( $P < 0.01$ ) than both the truck samples and the large tank samples. Significance was tested with the non-parametric Wilcoxon two sample test (27). This finding is consistent with the operational practices of the LPG retailers described above in that the samples which are anticipated to be older have, on the average, lower radon concentrations.

TABLE 9

RELATIONSHIP BETWEEN  $^{222}\text{Rn}$  CONCENTRATION IN LPG  
TYPE OF CONTAINER FROM WHICH THE SAMPLE WAS OBTAINED

Container	Number of Samples	Average Concentration (pCi/liter)
Large Tank	21	162.9
Truck	20	125.8
Small Tank	30	31.4

#### Radon Concentration in Home Storage Tanks

In order to estimate population dose and ultimately potential health effects, it is necessary to know or estimate the average radon levels in LPG contained in the consumers' home storage facilities. This could be best accomplished by a suitably large, well-distributed (geographically and in time) sampling of LPG in consumers' home storage tanks. Such data do not exist, however, so an alternative method will be employed to estimate the radon levels in delivered LPG as a function of geographical



location. This method is based upon Gesell's (4) data and knowledge of the locations of gas processing plants. Referring to figure 9, it is seen that California LPG has an average of ~150 pCi/liter. Texas exhibits an average of ~100 pCi/liter and Arizona-New Mexico exhibit an average of <50 pCi/liter. These regions have the highest radon in LPG levels, with the remainder of the sampled states exhibiting average radon in LPG levels of <5 pCi/liter.

It should be noted however that with the exception of Texas and California, very limited sampling was done in Gesell's study (4). For example, only three samples were gathered in Oklahoma. Even without resorting to statistical techniques, one can examine the data for Texas (table 7) and readily see that if only three samples were selected at random from Texas there would be a very good chance of missing the average value by a wide margin. Thus, excessive confidence should not be placed in the average concentration values for regions with only a few samples.

In addition to the measured concentration values in the fourteen states, the seasonal behavior must be considered. Furthermore, a rationale for estimating the average concentrations for the states not included in Gesell's survey must be developed. Gesell (4) found that for Houston, Texas the average winter  $^{222}\text{Rn}$  concentration was approximately one-half the average summer content. Because the fourteen state survey was performed in the summer months, one should consider applying a seasonal adjustment which would affect especially the dose due to unvented heaters which are used mainly in winter months. However, since the principle of radiation protection is to estimate on the conservative side, when reliable information is not available to indicate otherwise, and especially since the experience in Houston may not be directly applicable to other regions of the country, no seasonal adjustment is made for calculations in this study. This matter will be considered further in the discussion of uncertainties later in the report.

A scheme for estimating the radon concentration in LPG retailed in the unsampled states should reflect the known information in the sampled states as well as the distance from gas producing areas. Gesell has shown (table 8) that an expected relationship exists between average radon content and distance from a major gas processing area. Since this relationship indicates that concentrations decline as distance from a gas processing area increases, LPG from unsampled states far from gas processing areas should be estimated as lower in  $^{222}\text{Rn}$  content than states containing significant gas processing operations.

Based on the foregoing considerations, the following state average radon concentrations in LPG delivered to consumers have been assigned for the purpose of estimating potential health effects.

1. Based solely on Gesell's data (4), the following assignments have been made:

California:	150 pCi/liter
Arizona-New Mexico:	50 pCi/liter
Texas:	100 pCi/liter

2. Based partly on Gesell's data (4) and partly on the desire to be conservative, Oklahoma, Arkansas, Louisiana and the remaining surveyed southeastern states were assigned a value of 10 pCi/liter.
3. Based on proximity to states sampled by Gesell (4), Nevada, Utah and Colorado have been assigned a value of 50 pCi/liter.
4. Based primarily on the large distances from major gas processing areas and upon Gesell's data for states with gas processing, all remaining states have been assigned a value of 10 pCi/liter. (The significance of possible higher values in northern states due to import of Canadian LPG will be considered in the conclusions of this study.)

These radon concentrations are summarized in figure 10 which gives the state by state assigned values. All the calculations and estimates carried out later in the report utilize these values.

In previous studies on the potential radiological health effects of radon in natural gas (1-6), it has been generally assumed that the radon concentrations measured in the distribution systems are equivalent to the concentrations at the point of consumption. This assumption is a valid one because of the magnitude of the uncertainties in other aspects of the calculations and because transport pipelines do not represent more than a few hours to a few days delay between points of sampling and use. In the case of LPG, however, direct application of the concentrations found at the retail level would result in a serious over-estimation of the population dose. This over-estimation would occur because substantial storage time is involved in the residential storage tanks. If we consider that residential LPG storage tanks are filled at time intervals (t) with LPG of  $^{222}\text{Rn}$  concentration  $C_0$ , then the concentration C of the  $^{222}\text{Rn}$  in LPG at time  $t'$  after delivery would be

$$C = C_0 e^{-\lambda t'},$$

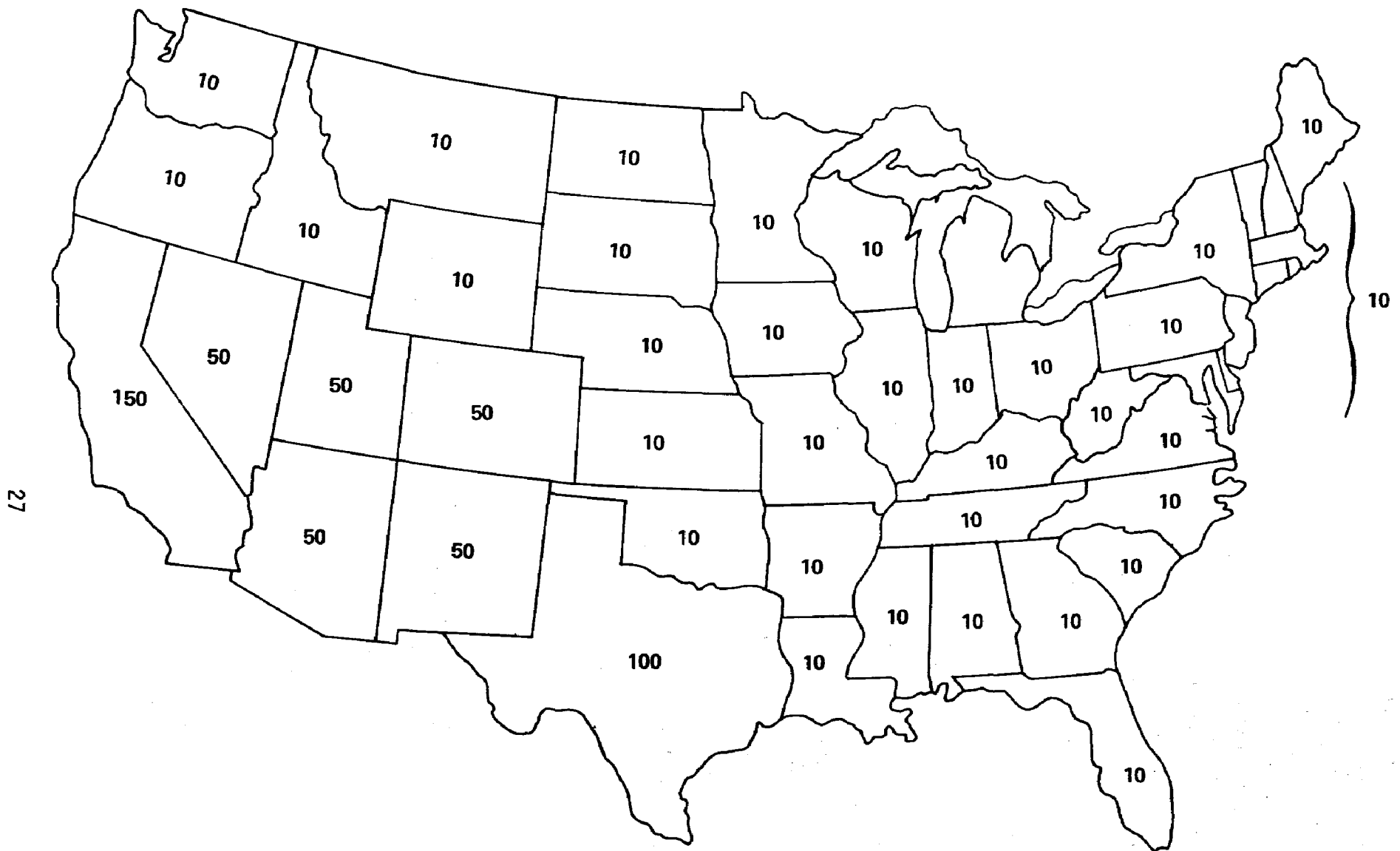
where  $\lambda$  is the decay constant for  $^{222}\text{Rn}$ . The average  $^{222}\text{Rn}$  content,  $\bar{C}$ , can be obtained by integrating C over the delivery interval (t) and dividing by t.

$$\bar{C} = \frac{1}{t} \int_0^t C(t') dt'$$

$$\bar{C} = \frac{1}{t} C_0 \int_0^t e^{-\lambda t'} dt'$$

$$\bar{C} = \frac{C_0}{\lambda t} (1 - e^{-\lambda t})$$

Discussions with dealers indicate that they prefer monthly deliveries and that domestic storage tanks are sized with that delivery interval in



**Figure 10. ASSIGNED VALUES OF  $^{222}\text{Rn}$  CONCENTRATION IN LPG DELIVERED TO CONSUMERS FOR THE PURPOSE OF ESTIMATING POPULATION DOSE AND HEALTH EFFECTS. ALL VALUES ARE IN PCI/L AT STP.**

mind. Accordingly, a time (t) of 30 days is chosen and

$$\bar{C} = 0.183 C_0.$$

This calculation assumes a constant rate of use and also assumes that the  $^{222}\text{Rn}$  content of the gaseous LPG bears a constant relationship to the  $^{222}\text{Rn}$  content of the liquid LPG.

### Radon in Indoor Air

The foregoing indicates how the radon content of LPG at the home appliance level may be estimated, including as parameters the retail sales level and radon decay in domestic storage tanks. The next step is to find the relationship between the radon concentration in the LPG and the radon concentration in dwelling air as a result of using the LPG in unvented appliances. It should be mentioned that LPG is by no means the only or, in most cases, even the dominant source of radon in dwelling air. Other sources include radon present in outdoor air, radon emanating from the construction materials of dwellings and radon in domestic water supplies. Gesell, et al. (3,4,16,17) and Johnson, et al. (5,6) have employed the simple concept of estimating the amount of radon released with gas combustion products from unvented gas ranges and space heaters into the dwelling space during 24 hours and dividing this quantity of radon by the volume of air available for dilution to give the dwelling radon concentration. The volume of air available for dilution is the product of the volume of the dwelling and the air exchange rate of the dwelling. The calculation implies uniform mixing, which may not occur. However, the error inherent in this assumption is partially offset because occupants move around the dwelling, thus exposing themselves to the different concentration levels.

This calculation also requires knowledge of the  $^{222}\text{Rn}$  concentration in LPG, the volume of LPG burned in each appliance, the degree to which the combustion products are vented to the dwelling, the volume of the dwelling, and the air exchange rate.  $^{222}\text{Rn}$  concentrations in LPG have been discussed earlier. The usage of LPG may be estimated from data on the usage of natural gas and the fact that the energy content of propane-based LPG is approximately 2-1/2 times that of natural gas on a volume basis. The "average" gas range utilizes  $0.765 \text{ m}^3$  ( $27 \text{ ft}^3$ ) of natural gas per day (28). This implies that an LPG range would require only  $0.306 \text{ m}^3$  ( $10.8 \text{ ft}^3$ ) of propane-based LPG. Jacobs et al. (29) have estimated that  $.354 \text{ m}^3$  ( $12.5 \text{ ft}^3$ ) per degree-day<sup>7</sup> of natural gas are required to heat a typical  $226.6 \text{ m}^3$  ( $8000 \text{ ft}^3$ ) dwelling. Thus,  $0.142 \text{ m}^3$  of LPG would be required for a degree-day.

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A degree-day is a term used by the heating industry to specify heating or fuel requirements as a function of outdoor temperature. (The number of degree-days on any given day is determined by the difference between  $65^\circ\text{F}$  and the daily mean temperature outdoors).

Gas appliances may be vented to the outside of the dwelling, partially vented to the outside, or unvented. Kitchen ranges are typically unvented, although a hood with a fan venting to the outside of the house is sometimes provided. Gas central furnaces are typically vented but there is widespread use of unvented space heating, particularly in the southern states. In Mississippi and Louisiana, over 30% of the dwellings utilize unvented space heating as the primary source of heat (24). In Texas, Oklahoma, Arkansas and Alabama, the figure is over 20% (24). Other gas appliances which may or may not be vented include clothes dryers and refrigerators.

Johnson et al. (5) and Handley and Barton (30) have recently reviewed the literature with regard to dwelling ventilation rates. The general disagreement in the literature is probably more reflective of actual differences in the characteristics of the dwellings than inadequacies in measurement. Values range from 0.5 to 9 air changes per hour. The American Society of Heating, Refrigeration and Air Conditioning Engineers (31) suggests values between 1/3 and 2 air changes per hour for various conditions for infiltration alone. Infiltration implies air passing through cracks and joints and is exclusive of air provided for ventilation by opening a window or providing a forced draft with a fan. Johnson et al. (5) and Gesell et al. (3,4,16,17) both employed one air change per hour in their calculations of radon exposure from natural gas and LPG. One air change per hour is conservative without being unrealistic. Furthermore, the calculated concentrations may be readily adjusted for other air exchange rates.

## DOSIMETRY

### $^{222}\text{Rn}$ and $^{222}\text{Rn}$ Daughter Dosimetry

Once the concentration of radon in dwelling air is established, the next logical step is to calculate the dose to an individual occupant of the dwelling under a prescribed set of circumstances. Unfortunately, the dosimetry of  $^{222}\text{Rn}$  and its daughters is neither simple nor well resolved. Johnson et al. (5) have provided a comprehensive review of the subject, especially as it applies to  $^{222}\text{Rn}$  in dwellings. Their task was complicated by the fact that most  $^{222}\text{Rn}$  dosimetry calculations have (justifiably) employed parameters typical of uranium mining. The following is a summary based partly on Johnson et al.'s (5) review to which the reader is referred for more comprehensive information and bibliographic material.

Table 10 gives a simplified relationship between  $^{222}\text{Rn}$  and its daughter products.  $^{222}\text{Rn}$  decays into a series of short-lived daughters which effectively terminate (insofar as airborne  $^{222}\text{Rn}$  and daughters are concerned) in 20.4 year half-life lead-210.

TABLE 10  
SIMPLIFIED DECAY SERIES FOR RADON-222

ISOTOPE	SYMBOL	HISTORICAL NAME	HALF-LIFE	TYPE OF DECAY	ALPHA ENERGY (MeV)
Radon-222	$^{222}\text{Rn}$	Radon	3.82 day	$\alpha$	5.49
Polonium-218	$^{218}\text{Po}$	Radium A	3.05 min.	$\alpha$	6.00
Lead-214	$^{214}\text{Pb}$	Radium B	26.8 min.	$\beta$	
Bismuth-214	$^{214}\text{Bi}$	Radium C	19.7 min.	$\beta$	
Polonium-214	$^{214}\text{Po}$	Radium C'	$164 \times 10^{-6}$ sec	$\alpha$	7.69
Lead-210	$^{210}\text{Pb}$	Radium D	20.4 yr.	$\beta$	
Bismuth-210	$^{210}\text{Bi}$	Radium E	5.0 day	$\beta$	
Polonium-210	$^{210}\text{Po}$	Radium F	138.4 day	$\alpha$	5.30
Lead-206	$^{206}\text{Pb}$	Radium G	Stable		

Numerous previous studies have yielded the following information about the dosimetry of airborne radon-222 and its airborne daughters (5).

1. The respiratory system and more specifically, the basal cell layer of the bronchial epithelium is the critical tissue.
2. The dose to this tissue is provided almost exclusively by the short-lived daughters of  $^{222}\text{Rn}$  i.e.,  $^{218}\text{Po} \rightarrow ^{214}\text{Po}$  rather than by the  $^{222}\text{Rn}$  itself. This occurs because radon, a noble gas, tends not to plate out on or concentrate in any particular tissue whereas the short-lived daughters, all metals, tend to deposit and adhere to the surface of the respiratory system.
3. Of the various radiations emitted by the short-lived daughters, the alpha radiation is by far the most important from a dose standpoint.
4. The principal adverse health effect associated with elevated levels of airborne  $^{222}\text{Rn}$  daughters is bronchial carcinoma.

The foregoing allows the dosimetry of  $^{222}\text{Rn}$  and its daughters to be reduced to a study of the dose imparted to the bronchial epithelium from the alpha emissions of the short-lived  $^{222}\text{Rn}$  daughters.

Ideally, estimation of the tracheobronchial dose (T-B dose) due to  $^{222}\text{Rn}$  daughters would begin with specification or measurement of the airborne concentrations of  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$  and  $^{214}\text{Po}$ . Subsequently the alpha dose resulting from each isotope would be calculated and the result summed. This task would be slightly simplified by the extremely short half life of  $^{214}\text{Po}$  which would allow one to treat the  $^{214}\text{Bi} - ^{214}\text{Po}$  pair as a single species. Despite this simplification, the difficulty of measuring the three isotopes under field conditions requires sophisticated equipment, and this led to the introduction of a special unit, the "working level" (WL) for specifying the collective airborne  $^{222}\text{Rn}$  daughter concentrations.

The "working level" was originally intended as a safe but not unduly restrictive level of airborne  $^{222}\text{Rn}$  daughters for application in uranium mining. However, the present occupational standard for radon daughter exposure is no longer one working level but rather 1/3 WL. The WL unit is still employed as a measure of airborne radon daughter concentrations and is equivalent to an amount of short-lived radon daughters in one liter of air whose decay would result in the release of  $1.3 \times 10^5$  MeV of alpha energy. It is also equivalent to the concentration of short-lived radon daughter products in radioactive equilibrium with 100 pCi/l of radon-222. The latter equivalency does not imply a direct conversion from radon-222 concentration to WL, however, because radioactive equilibrium among radon-222 and its daughters seldom exists in the field situation.

No practical means has been devised for directly measuring the dose imparted to bronchial tissue by  $^{222}\text{Rn}$  daughters so the relationship between airborne  $^{222}\text{Rn}$  daughter concentrations and dose is obtained by dose modeling and calculations. All of the dose models require specification of the degree of radioactive equilibrium which exists among the  $^{222}\text{Rn}$  daughters<sup>8</sup>, specification of the fraction of the radon daughters which remain unattached to aerosols (free ions)<sup>9</sup>, and the size distribution of aerosol particles. Jacobi (32) has recently shown, using the ICRP Lung Model, that the relationship between tracheo-bronchial dose per working level month (WLM; equivalent to exposure at one WL for one "occupational month" or 170 hours) can vary over an order of magnitude depending upon such factors as unattached daughter concentration and degree of equilibrium. Thus, a knowledge of WL only is insufficient to make dose estimations, and unattached daughter concentrations, particle size distribution, and the degree of equilibrium must be either measured or estimated.

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The degree of radioactive equilibrium between  $^{222}\text{Rn}$  and its daughters may be expressed as  $10, f_1, f_2, f_3$  where 10 is taken as the concentration of  $^{222}\text{Rn}$ ,  $f_1$  is the relative (to Rn) concentration of  $^{218}\text{Po}$  (RaA),  $f_2$  is the relative (to Rn) concentration of  $^{214}\text{Pb}$  (RaB) and  $f_3$  is the relative (to Rn) concentration of the  $^{214}\text{Bi}-^{214}\text{Po}$  (RaC-RaC') pair. Thus perfect equilibrium would be expressed as 10, 10, 10, 10.

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Airborne  $^{222}\text{Rn}$  daughters which are unattached to aerosols are called "free ions". The fraction of free ions for each species is usually expressed as percentage of the total number of each species.

Johnson et al. (5) have compiled a table summarizing the existing work on  $^{222}\text{Rn}$  daughter dosimetry which is reproduced here as table 11. The conditions used by each investigator have been normalized for continuous exposure for one year (8,760 hours) at one working level. The results of each investigator, expressed in rads/year are not directly comparable because different assumed values for free ion concentration and equilibrium conditions were employed. Furthermore, different atmospheric dust loading and lung models were utilized. Table 11 provides information on the order of magnitude and range of the various conversion factors. Presumably the appropriate conversion factor(s) for the situation of radon daughters in dwellings are encompassed by the values in table 11. Measurements of the various parameters in dwellings could be employed to narrow the range of conversion factors.

#### *Dose conversion factor for this study*

In earlier studies on radon in natural gas, Barton, et al. (1) and Johnson et al. (5) used the conversion factor of 100 rads per year to the tracheobronchial epithelium for continuous exposure at 1 WL. This corresponds to 1000 rem per WL for a quality factor of 10 for alpha particles. Upon further evaluation, Johnson et al. (5) concluded that several parameters involved in deriving this conversion factor were overly conservative. Subsequently, a lower conversion factor was derived for evaluating doses from radon emanation from uranium mill tailings piles. [As part of the environmental analysis of the uranium fuel cycle made by EPA (42).] This lower factor is used for determining dose from radon in LPG.

This factor was derived from data presented by Haque and Collinson (39) and in the 1972 UN SCEAR Report (43-p. 35,81). These data are based on exposure to radon and daughters in a home with adequate ventilation for a period of 6000 hours. Absorbed doses were calculated for test conditions with a radon concentration of 0.164 pCi/l and a daughter distribution of 0.9, 0.5, 0.35; i.e., ratio of the radon daughters RaA, RaB, RaC (RaC') respectively, to the radon concentration. Variations on two lung model parameters were taken into account for these dose calculations. One was the thickness of the mucous sheath and epithelium, i.e., the distance for an alpha particle to penetrate from the deposited radon daughter to the critical basal cell nuclei of the bronchial epithelium. The other parameter was the lung region of concern as denoted by the generation number of the Weibel dichotomous lung model (39). The particular variations on these parameters selected for this study were a thickness of 60 microns and a generation number of 5 which corresponds to the segmental bronchi region of the lung. These selections were made on the basis of state-of-the art knowledge of lung models and may require modification as more data become available. <sup>10</sup>



Table 11. Summary of dose conversion factors for radon and radon daughters (5)

Radon-daughter equilibria	Exposure conditions	Lung model, critical tissue	Dose factor <sup>(a)</sup> rads/year	Reference
10,10,10,10 <sup>(b)</sup> 4% free RaA	0.3 $\mu$ particles Rn-100 pCi/l annual occupational exposure	Weibel model (A) 15 l/min, segmental bronchi	12	Harley and Pasternack (33)
10,6,3,2 4% free RaA	"	"	18.5	(33)
10,9,6,4 8.5% free RaA	"	Landahl model	86	(33)
Nonequilibrium, little free RaA	WLM = 170 hrs. <sup>(c)</sup>	Epithelial base cells of large bronchi	25.8-51.5	BEIR (34)
Nonequilibrium	500 hours per month in homes	Bronchial epithelium	34	Toth (35)
Nonequilibrium, 1-2% free RaA	Clean air MPAI of 4 WLM <sup>(c)</sup>	Revised ICRP model (38), bronchial region	19.3-51.5	Jacobi (32)
"	High aerosol conc. 0.05-0.2 $\mu$ m particles	"	15.5-25.8	(32)
10,10,6,4 <sup>(d)</sup>	Normal room air change-1 hr <sup>-1</sup> , 10,000 particles cm <sup>-3</sup> , 0.09 $\mu$ m 10 pCi/l-Rn	Findeisen-Landahl model, bronchial epithelium, 14 l/min.	88	Jacobi (36)
10,10,10,10 25% free RaA	Natural radiation exposure, 0.09 $\mu$ m 0.1 pCi/l-Rn	"	140	(36)
10,9,6,4 <sup>(d)</sup> 8.5% free RaA	0.3 $\mu$ m particles Rn-100 pCi/l, occupational exposure	Landahl model, segmental bronchi, 15 l/min., mouth breathing	103	Altshuler et al. (37) Lundin (38)
"	"	" Nose breathing	56	(37)
10,9,5,3.5	Adequately ventilated room, 6,000 hr/yr	Segmental bronchi, 15 l/min., mouth breathing	89-620	Hague and Collinson (39)
10,9,6,4	>0.1 $\mu$ m particles Rn-100 pCi/l	Segmental bronchi 15 l/min.	111	Burgess and Shapiro (40)
Range 12-620				

(a) Dose factor = rads/year for continuous exposure (8,760 hours) to one working level. One WL = any combination of short-lived radon daughters (through <sup>214</sup>Po, RaC') leading to a total emission of  $1.3 \times 10^5$  MeV of alpha energy per liter of air (41). One WL is also defined as 100 pCi/l of radon in equilibrium with its daughters.

(b) Relative concentrations of <sup>222</sup>Rn, RaA, RaB, and RaC (RaC').

(c) WLM, 1 working level month = 170 hours exposure at 1 WL. Jacobi (32) defines 1 WLM as  $2.6 \times 10^{10}$  MeV potential alpha-energy inhaled at 20 l/min. for 166.7 hours/month. MPAI = maximum permissible annual intake.

(d) These conditions represent typical dwellings.

The dose calculated for the above exposure conditions was 0.04 rad. This dose must then be multiplied by 1.46 for continuous exposure for one year (8766 hours); by 6.1 to convert from 0.164 pCi/l of radon to 1.0 pCi/l; and by a quality factor of 10 to convert to dose equivalent in rem. Thus, for continuous exposure to an atmosphere of 1.0 pCi/l of radon-222 with a daughter ratio of 0.9, 0.5, 0.35, the radiation dose would be 3.56 rem/yr to the bronchial epithelium.

For comparison, the calculation of dose may also be done on the basis of working levels of exposure by using the BEIR Report (34) estimates to derive a dose equivalent. This is done by converting the above radon and daughter concentrations to working levels according to table 12.

Table 12. Calculation of Working Levels

Nuclide	pCi/l	Atoms/l	Alpha Energy per atom (MeV)	Total Potential alpha energy (MeV)
Rn	1	$1.77 \times 10^4$	----	---
RaA	0.9	8.79	13.68	120
RaB	0.5	42.9	7.68	329
RaC	0.35	22.1	7.68	170
RaC'	0.35	$10^{-6}$	7.68	---
				<u>619</u>

$$\frac{619 \text{ MeV}}{1.3 \times 10^5 \text{ MeV/WL}} = 0.00485 \text{ WL}$$

Exposure at 0.00485 WL for 170 working hours per month corresponds to 0.00485 WLM (working level months). Using the BEIR Report (34) conversion factors of 5-10 rem/WLM, this exposure corresponds to 0.024 to 0.048 rem per working month. These doses are converted to continuous annual dose by multiplying by 12 and by 4.3 (ratio of hours in a year to hours in 12 working months). The yearly doses are then in the calculated range 1.24 rem to 2.48 rem. Since the BEIR conversion factor of 10 rem/WLM is applicable except for individuals with chronic bronchitis, then 2.48 rem/year is the better estimate for dose equivalent delivered by continuous exposure at 1 pCi/l of radon-222.

Of the two methods for estimating doses, the method based on Haque and Collinson (39) gives the highest dose for the lung region of concern.

The BEIR Report does not estimate dose for a particular part of the bronchial tree and therefore does not emphasize a critical region. Furthermore, the BEIR estimates are for occupational exposure to uranium miners, and therefore do not apply directly to normal living conditions. The cleaner air and smaller aerosol particle sizes in homes, as opposed to mines, would result in an increased dose for the same radon concentration (or WL) due to a larger fraction of free ions and the fact that more small particles penetrate to the critical region of the lung. Thus, the dose conversion factor that will be used in this study is 3.56 rem/year, which will be rounded off to

$$1 \text{ pCi/l radon-222} = 4 \text{ rem/year}$$

for daughter ratios of 0.9, 0.5, 0.35, and a penetration depth of 60 microns to the nuclei of the cells at risk, which are the basal cells of the segmental bronchi. This conversion factor is equivalent to 800 rem/year at one WL, which is 200 rem/year at one WL lower than the conversion factor used by Johnson et al. (5) in the previous study on radon in natural gas.

#### Postulated Exposure Conditions

The exposure conditions for determining dose to an individual using LPG in home cooking and heating are summarized in table 13. Most of these conditions are the same as those used by Johnson et al. (5) to estimate the radon dose from natural gas. The main differences are in the radon concentrations involved, the smaller quantities of LPG used, and the lower dose conversion factor as discussed above. As noted previously, less LPG is used because this fuel has a higher B.t.u. content than natural gas.

Using the parameters from table 13 the following general equation may be derived for estimating the airborne concentration,  $C_A$ , of radon-222 in homes using LPG in unvented appliances.

$$C_A \text{ (pCi/l)} = \frac{C_o \times Q \times DF}{V \times R}$$

Where:  $C_o$  = radon concentration in LPG delivered to homes  
(pCi/l)

$Q$  = quantity of LPG used ( $\text{m}^3/\text{day}$ )

$DF$  = decay factor due to storage in home tanks (0.183 for 30 days)

$V$  = house volume ( $226.6\text{m}^3$ )

$R$  - air exchange rate (24 house volumes/day)

For example, a home using LPG in a kitchen range, with the highest radon concentration measured (1240 pCi/l) and monthly deliveries, would have an average indoor radon concentration from this source of

Table 13. Exposure conditions employed in the estimation of dose from radon in LPG

Parameter	Condition Used in this Analysis <sup>a</sup>	Possible Variation <sup>b</sup>
<sup>222</sup> Rn concentration in delivered LPG	An average value has been assigned to each state (10 -150 pCi/l)	0 - 1500 pCi/liter
LPG delivery interval	one month	2 weeks - 3 months
Storage tank decay factor	0.183	0.363 - 0.0613
LPG use		
Cooking ranges	0.306 m <sup>3</sup> /day	up to 0.476 m <sup>3</sup> /day
Space heaters	0.142 m <sup>3</sup> /degree-day	0.112 - 0.168 m <sup>3</sup> /degree-day
Ventilation conditions (appliances)	unvented	ranges may be potentially vented
Dwelling volume	226.6 m <sup>3</sup> (8000 ft <sup>3</sup> )	142 - 425 m <sup>3</sup>
Degree days	average for each state	± 25% within states
Air exchange rate	1 dwelling volume/hour	0.25 - 5 per hour
Number of persons occupying each dwelling	4 (continuous occupancy)	1 - 10 (or partial occupancy)
<sup>222</sup> Rn and daughter equilibrium		
in LPG	1, 0, 0, 0 <sup>c</sup>	up to 1, 1, 1, 1
in dwelling air	1.0, 0.9, 0.5, 0.35	1.0, 0.5, 0.25, 0.1 to 1, 1, 1, 1
Percent free <sup>218</sup> Po	35%	5 - 50%
Critical pathway	inhalation of radon daughters	<sup>222</sup> Rn accounts for <1% of dose
Critical organ	bronchial epithelium	some exposure to remainder of respiratory tract and other organs
Dose conversion factor <sup>d</sup>	4 rem/yr for continuous exposure at 1 pCi/l	2 - 5 rem/year
Quality factor	10	1 - 10

<sup>a</sup>Conditions are intended to be typical and to err on the conservative side in the case of less well-understood parameters.

<sup>b</sup>These variations are intended to cover a large fraction of actual exposure conditions.

<sup>c</sup>Ratio of <sup>222</sup>Rn, <sup>218</sup>Po, <sup>214</sup>Pb, <sup>214</sup>Bi (<sup>214</sup>Po).

<sup>d</sup>This factor incorporates radon daughter equilibrium conditions, free ion fraction, critical pathway, and dosimetry model parameters.

$$C_A = \frac{1240 \times 0.306 \times 0.183}{226.6 \times 24} = 0.0128 \text{ pCi/l}$$

If the same home used unvented space heating, an additional 0.142 m<sup>3</sup> of LPG would be required for each degree-day. To pick a "worst case" example, Duluth, Minnesota requires 10,000 degree-days of heating per year (31). Thus, heating would result in a radon concentration of

$$C_A = \frac{1240 \times 0.142 \times 10,000 \times 0.183}{226.6 \times 24} = 0.162 \text{ pCi/l}$$

for this example of the highest radon concentration in LPG and the coldest weather conditions. For the average daily heating requirement in the U.S., which is 7.77 degree-days (5), the radon concentration in this same home would be

$$C_A = \frac{1240 \times 0.142 \times 7.77 \times 0.183}{226.6 \times 24} = 0.046 \text{ pCi/l}$$

#### Dose to an Individual

The dose to an individual is estimated by multiplying the dose conversion factor (4 rem/year per pCi/l of radon) times the radon concentration in the home air. Individual doses for various combinations of exposure conditions are shown in table 14.

Table 14. Doses to individuals from radon-222 in LPG

Radon-222 in home tank (a) pCi/l	Dose equivalent (mrem/year)		
	cooking ranges (b)	space heaters (c)	Total
40	0.41	1.48	1.89
150	6.2	22.3	28.5
1240	51	184	235
1240	51	648 (d)	699

(a) Concentration at time of delivery to home tank

(b) Average LPG use 0.306 m<sup>3</sup>/day

(c) Average LPG use 1.1 m<sup>3</sup>/day

(d) For 10,000 degree-days/year and 1240 pCi/l of radon-222

NOTE: It is not likely that the highest radon concentration and highest LPG use would occur together.

This table indicated that for average LPG radon concentrations of 10 to 150 pCi/l, the average annual dose to individuals would be less than 30 mrem even for combined use of unventing cooking ranges and space heaters. The maximum individual dose for the conditions of highest LPG radon concentration and gas use could be about 700 mrem/year although it is not likely that both conditions would occur together. Existing data are insufficient to determine the significance of potential high individual doses. This matter will be further evaluated by ORP.

### Population Dose

The tracheobronchial (T-B) doses to the United States population were estimated on a state-by-state basis in the same manner as for individual doses. Within each State, the concentration of radon-222 in LPG and the annual degree-day heating requirements were assumed to be constant. The individual dose for these conditions was then multiplied by the number of dwellings using LPG in unvented appliances and by four (number of occupants per dwelling) to obtain the State population dose.

The number of dwellings using LPG in cooking ranges was obtained directly from census data (24). However, determining the number of unvented space heaters using LPG was more involved. While census data gave the number of dwellings with unvented heaters and also the number using LPG fuel, these data do not directly give the number of dwellings which use LPG in unvented space heaters. Therefore, the number was estimated by assuming that unvented space heaters are fueled by natural gas, LPG, or kerosene in the same proportion as all home heating systems in each State. Fuels such as coal or wood were not considered because they are obviously unsuited for unvented heating. The estimate was further refined by separating each State into rural and urban components prior to making the estimate. The urban and rural estimates were combined for the total number of unvented space heaters burning LPG in each State.

The population T-B doses and the parameters for estimating these doses are given in table 15. The total population T-B dose equivalent for use of LPG in kitchen ranges was estimated to be about 18,400 person-rem per year. The total dose equivalent for unvented space heaters was about 10,600 person-rem year.

By assuming four occupants per dwelling, the population at risk for exposure to radon from LPG use in kitchen ranges is about 21.3 million persons or about 10 percent of the United States population. The average individual dose equivalent from this source is about 0.87 mrem/year. For space heaters, the potential population at risk is about 2.8 million persons or about 1.3 percent of the population. The average dose to individuals from LPG in space heaters is about 3.7 mrem/year.

The combined population T-B dose equivalent for exposure to radon daughters from use of LPG in unvented kitchen ranges and space heaters is estimated to be about 29,000 person-rem per year for the United States.

Table 15

Estimated Dose Equivalent to the U.S. Population Due to  $^{222}\text{Rn}$  in LPG

State	Assumed $^{222}\text{Rn}$ Concentration in LPG (pCi/l)	Estimated Dwellings with Unvented LPG Heaters	Estimated Annual Degree- Days	Estimated Population T-B Dose- Heaters (person-rem/yr)	Dwellings with LPG Ranges	Estimated Population T-B Dose- Ranges (person-rem/yr)	Total Estimated T-B Dose (person-rem/yr)
Texas	100	160,710	1,940	6,524	336,334	5,555	12,079
California	150	9,928	2,756	859	164,848	4,083	4,942
Florida	10	80,813	742	126	425,837	703	829
Mississippi	10	97,762	2,190	447	147,351	242	689
New York	10	787	6,266	9	377,931	624	633
Georgia	10	78,318	2,435	399	135,235	224	623
Alabama	10	76,886	2,368	382	104,154	171	553
Arkansas	10	44,012	3,015	278	143,146	237	515
Oklahoma	10	42,074	3,792	335	108,146	179	514
Missouri	10	7,675	4,921	79	258,522	427	506
New Mexico	50	3,441	4,646	167	32,318	267	434
Pennsylvania	10	529	5,531	6	256,977	425	431
Colorado	50	1,205	6,310	79	38,656	320	399
Illinois	10	2,279	5,899	28	192,504	317	345
Louisiana	10	48,956	1,627	167	108,175	178	345
North Carolina	10	5,389	3,284	38	185,263	306	344
Indiana	10	3,512	5,694	41	181,434	299	340
Arizona	50	2,201	3,295	75	29,554	244	319
Ohio	10	1,092	5,840	13	175,161	289	302
Michigan	10	1,516	7,372	23	157,708	261	284
Wisconsin	10	2,134	7,779	34	149,184	246	280
Minnesota	10	2,116	8,892	39	144,861	239	278
Kentucky	10	6,189	4,869	64	125,216	207	271
Iowa	10	251	6,870	4	142,002	235	239
Virginia	10	832	3,776	6	137,600	227	233
South Carolina	10	9,238	2,336	45	87,000	143	188
Nevada	50	678	6,194	43	17,197	143	186
Maryland	10	408	4,617	4	109,729	180	184
New Jersey	10	211	4,794	2	100,276	165	167
Massachusetts	10	186	6,518	2	90,133	148	150
Tennessee	10	11,248	3,485	83	36,811	60	143
Maine	10	76	8,639	2		137	139
Connecticut	10	127	5,916	2	75,317	124	126
Kansas	10	1,618	5,282	19	59,575	98	117
Nebraska	10	1,348	6,681	19	45,061	75	94
Utah	50	290	6,109	19	8,534	70	89
South Dakota	10	254	7,802	4	40,934	68	72
New Hampshire	10	69	7,383	2	42,144	70	72
West Virginia	10	1,631	4,838	17	31,247	51	68
Vermont	10	159	8,269	4	35,691	58	62
North Dakota	10	514	9,303	9	25,665	43	52
Oregon	10	835	6,045	11	24,814	41	52
Washington	10	966	5,368	11	24,314	39	50
Delaware	10	76	4,930	--	26,234	43	43
Rhode Island	10	53	5,879	--	21,738	36	36
Montana	10	746	8,094	13	14,329	23	36
Idaho	10	497	6,128	6	12,152	21	27
Wyoming	10	419	7,585	8	11,159	19	27
Alaska	10	33	12,097	--	14,365	23	23
Hawaii	10	26	---	--	12,860	21	21
District of Columbia	10	18	4,617		7,308	11	11
Totals		713,317		10,564	5,315,373	18,415	28,979

### *Possible variations in population dose estimates*

The estimation of population dose was performed using nominal values of the many parameters entering into the calculation. Different estimates would result for other exposure conditions. To provide some insight on how the dose estimates may be adjusted for different conditions, a listing of possible corrections is given in table 16. It is seen that reasonable variations in several parameters could result in changes in the calculated doses by a factor of two.

### POTENTIAL HEALTH EFFECTS

Although radon was not known to be the cause, health effects were reported as excessive pulmonary disease among groups of miners in the Joachimsthal and Schneeberg mining areas of central Europe over three hundred years ago (47). Herting and Hesse (48) reported the first autopsies on deceased miners and identified malignant growth in 1879. In 1911 these growths were demonstrated to be carcinomas of the lung. Epidemiological studies reported by Thiele et al. (49) in 1924 and by Peller (50) in 1939 denoted highly significant increases of pulmonary cancers among miners. Careful epidemiological work on uranium miners, in particular, has demonstrated significant increases in bronchial carcinomas (47). The agents responsible for a significant proportion of these increases have subsequently been identified as airborne radon daughters which, upon inhalation, become attached to pulmonary surfaces where they release their alpha decay energy into these tissues.

### Conversion from Dose to Potential Health Effects"

The general approach for deriving an appropriate factor relating health effects to dose from radon daughter irradiation of lung tissues involves two steps. The first step is to determine an age and population adjusted estimate of excess somatic cancer deaths. Then, the estimate of excess lung cancer deaths can be derived as a fractional part of the somatic cancer deaths.

The estimation of excess lung cancer mortality in this study will be based on the report by the National Academy of Science on the biological effects of ionizing radiation (BEIR report) (34). The excess risk of death from lung cancer due to exposure to one rem of ionizing radiation may be determined using risk model data from table 3-1, p. 169 and table 3-2, p. 171 of the BEIR report.

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The health effects conversion factor derived in this study is based on information provided by Neal S. Nelson, Ph.D., U.S.E.P.A., Office of Radiation Programs, in a memorandum to R. Johnson, November 14, 1974.



Table 16

Corrections to Adjust Estimated Population Doses for  
Different Exposure Conditions (5)

Parameter	Value	Adjustment Multiplier
Ventilation rate-air changes per hour	0.25 (1.0)(a) 2.0	6.0 1.0 0.34
$^{222}\text{Rn}$ concentration in LPG	varies with state (10 - 150 pCi/l)	linear(b)
Quantity of LPG Used	varies with heating requirements	linear
Dwelling size	(226.6 m <sup>3</sup> ) (8000 ft <sup>3</sup> )	inverse
Daughter equilibrium ratio	1,1,1,1 (1, 0.9, 0.5, 0.35) 1, 0.75, 0.5, 0.3 1, 0.5, 0.25, 0.1	1.9 1.0 0.75 0.37
Percent free $^{218}\text{Po}$	3 10 25 (35) 50	0.27 0.38 0.77 1.0 1.3
Dose conversion factor	(4 rem/yr at 1 pCi/l)	linear
Quality factor	(10)	linear

(a) Values in parentheses were used in this study.

(b) A linear correction means the correction is proportional to the variation in the parameter.

Data from both the absolute risk model and the relative risk model of the BEIR report were combined to determine the best estimate of excess cancer deaths. The absolute risk model estimate is based on the product of assumed risk times the total population at risk and gives the number of cases that may result from a given exposure to a given population. The relative risk model estimates excess deaths on the basis of the ratio of the risk in those exposed to the risk versus those not exposed (incidence in exposed population to incidence in control population).

The age adjusted estimates from both models of excess somatic cancer deaths for a population exposure of a million persons/year/rem (i.e.  $10^6$  person-rem/year) are shown in table 17. These estimates were derived from 1967 U.S. population data after converting from 0.1 to 1.0 rem/year (table 3-1, 34). Data are given for a duration of risk (plateau region) of 30 years and a lifetime. The data for both plateau regions are combined in the estimate for excess lung cancer deaths by taking the geometric mean. This was done instead of taking the arithmetic mean to compensate for uncertainty in the lifetime plateau estimate for the relative risk model.

The estimates for excess lung cancer mortality were determined by assuming that the cancer distribution in both models is the same for all ages. Then the proportion of lung cancers was taken as 26 percent of all excess somatic cancer deaths using the data from table 3-2 of the BEIR report.

The estimates from both models were combined by taking the average of the geometric means for the two plateau regions. The combined estimate is 38.5 excess lung cancer mortalities for each million persons exposed/year/rem. This estimate will be rounded off to one significant figure to give the following health effects conversion factor for use in this study

$$4 \times 10^1 \text{ excess lung cancer deaths}/10^6 \text{ person-rem.}^{12}$$

for continuous exposure to radon daughters.

It should be noted that 100% fatality is assumed here. In addition, it should be recognized that very few data are available on carcinogenic alpha dose to the lung. Furthermore, the lung cancer estimates here are based on data derived from dose levels considerably higher than anticipated for radon in LPG. The BEIR report points out that extrapolation to low doses for large numbers of people has inherent uncertainties such that zero cannot be excluded as a possible conversion from radiation dose to cancer incidence for the low dose rate conditions of this study. These considerations should be kept in mind when evaluating the potential health effects which may result from radon-222 in LPG.

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This conversion factor may be modified as more data become available from ongoing studies.

Table 17.

Estimates of excess somatic and lung cancer deaths

Risk Model	Excess somatic cancer deaths <sup>(a)</sup> for 10 <sup>6</sup> person-rem/year		
	<u>30 year plateau</u>	<u>lifetime plateau</u>	
Absolute	61.2	75.0	
Relative	123.1	421.5	
	Excess lung cancer deaths <sup>(b)</sup> for 10 <sup>6</sup> person-rem/year		
	<u>30 year plateau</u>	<u>lifetime plateau</u>	<u>geometric mean<sup>(c)</sup></u>
Absolute	15.9	19.5	17.6
Relative	32.0	109.9	59.3
Combined Models	$\frac{17.6 + 59.3}{2} = 38.5$		

(a) Excluding leukemia.

(b) Assumes lung cancers represent 26% of somatic cancer deaths. (BEIR Report, p. 171).

(c) The geometric mean was used to compensate for uncertainty in the lifetime plateau estimate for the relative risk model.

### Health Effects Estimate

The total dose equivalent for continuous exposure to radon daughters from use of LPG in unvented kitchen ranges and space heaters in the United States is estimated to be about 29,000 person-rem/year. When the health effects conversion factor is applied to this dose the estimate of potential health effects is

$$[29 \times 10^3 \text{ person-rem/yr}] [4 \times 10^{-1} \text{ effects}/10^6 \text{ person-rem}] = \\ 1.16 \text{ effects/year}$$

or about one excess lung cancer mortality a year.

### CONTROL COSTS

At the present time the most practical method for controlling the concentration of radon in LPG would be storage to take advantage of the relatively short half-life of radon-222 (3.83 days). Storage is already an integral part of the LPG industry, therefore the technology is readily available for use of storage to control radon levels. Approximately 75 percent of existing storage is underground in depleted oil wells or in caverns within salt formations. The remaining 25 percent of storage for LPG is in above ground tanks mainly at distribution centers or points of LPG consumption.

Storage serves a primary function of providing the necessary balance between a constant production rate and seasonal demands for LPG. However, at present all the available underground storage capacity is being used. Therefore, LPG is in an oversupply status which has resulted in recent increases in charges for LPG storage due to the storage demand.

Since existing underground storage capacity is fully utilized, new storage requirements for control of radon could require extensive additions to above ground storage. However, for the purpose of this control cost analysis, the assumption will be made that development of new storage capacity would be done in the same ratio as existing above and below ground storage, i.e. 25 and 75 percent respectively.

The following cost analysis for control of radon-222 in LPG is intended to provide order-of-magnitude estimates to evaluate the significance of potential health effects in relation to control costs. The actual costs for individual storage facilities can vary widely as functions of facility size, production rates, pumping costs, and distribution logistics.

The costs for storing LPG will be estimated by the present worth method for annualized costs. This method estimates the yearly capital

cost by an annual fixed charge rate. The fixed charge rate used in this analysis was 16 percent per year, including interest, taxes, insurance, and depreciation for a thirty year facility life. The sum of the annual capital costs and annual operating costs gives the annualized costs.

The cost data for this analysis are given in table 18. The major factor in the operating costs is that of pumping the LPG into and out of storage. These pumping costs are approximately the same for each storage method and average about 0.25 cents/gal for pumping into storage and the same for pumping out.

For this analysis, the control costs will be estimated for a two week storage of all the LPG produced in the two states (Texas and California) with the highest average radon-222 concentrations in LPG. Two weeks of storage will allow radon-222 to decay to less than 10 percent of its original concentration. Texas and California together produce about 51 percent of the LPG in the United States. Their combined production for two weeks is about  $1.6 \times 10^8$  gal.

The annualized capital costs are calculated on the basis of capital costs for a two week storage capacity plus annual operating costs consisting of pumping charges for cycling the LPG through storage every two weeks. The summary of annualized costs is shown in table 19.

#### Comparison of Radon Control Costs to Reduction in Potential Health Effects

The estimated cost for control of radon in LPG by storage would be from 38 to 48 million dollars a year for about half of normal United States production. For this storage to be effective it should be applied in the production areas with highest radon-222 concentrations, i.e. Texas and California. Since it is not known which states use LPG from Texas and California, it is not possible to calculate specifically the potential reduction in health effects which may be attributed to control of radon in LPG from those two states. However, the maximum benefit that could be derived from this control would be the elimination of estimated health effects (about one excess lung cancer mortality a year).

Thus, if the proposed storage control could eliminate the potential of one excess lung cancer a year, the control cost per health effect reduction would be in the order of 38 to 48 million dollars. Since other uncontrolled LPG sources would continue to contribute to population lung dose, then the potential number of excess lung cancers could at best only be reduced to some fraction per year by storage control. Therefore the cost per health effect reduction could be considerably in excess of 50 million dollars. Investments of this magnitude for controlling radon-222 in LPG are clearly not cost effective in relation to the possible reduction in health effects.

Table 18. Costs for LPG Storage (1974 Basis)

<u>Storage Method</u>	<u>Capital Costs</u>	<u>Operating Costs</u>
Underground		
Oil Wells	\$0.02-0.03/gal/yr	\$0.005/gal
Salt Caverns	\$0.04-0.05/gal/yr	\$0.005/gal
Above ground	\$0.35-0.50/gal/yr	\$0.005/gal

Table 19. Annualized Cost Estimate for Storage of LPG<sup>(a)</sup>

<u>Storage Method</u>	<u>Annualized Costs</u>
Underground <sup>(b)</sup>	$\$18.5 \times 10^6$ to $\$22.2 \times 10^6$
Above ground <sup>(b)</sup>	$19.7 \times 10^6$ to $25.8 \times 10^6$
Total	$\$38.2 \times 10^6$ to $\$48.0 \times 10^6$

(a)

For two weeks production of LPG from Texas and California ( $\sim 1.6 \times 10^8$  gal).

(b)

75 percent storage underground, 25 percent above ground.

## DISCUSSION

### Review of Uncertainties

A detailed review of the applicable uncertainties was presented in the earlier study on natural gas (5). Therefore, only a few of the main points will be reviewed in this report. First, it should be emphasized that throughout this analysis values have been assumed for exposure conditions or population at risk that were conservative, i.e., so that the calculated doses or health effects would be overestimated rather than underestimated. At the same time, however, an effort has been made to obtain values that were realistic in order that the final health effects estimate might be in reasonable perspective.

Possible variations in exposure conditions which could be reasonably encountered by large portions of the population at risk are summarized in table 13. Factors for correcting the calculated dose to adjust for other exposure conditions are given in table 16. Individuals or small groups could conceivably receive doses significantly higher than estimated in this study, but no data are available for assessing this possibility.

The observations which initially led to this study on LPG were that radon is separated from natural gas with the LPG fraction and that very high radon concentrations in LPG had been measured at processing plants. The distribution and use patterns for LPG were not known at that time. Since radon-222 has such a short half-life, the significance of high concentrations at the gas processing plant has to be considered in terms of distribution and storage time prior to home use. Assessment of these factors indicated that average radon concentrations were a function of distance from the processing plant. More importantly, this study showed that storage in home tanks is a significant factor which allows radon to decay to two-tenths or less of its original concentration between tank refills. Consequently, the postulated radon concentration in LPG is quite low for most States, and the baseline level of 10 pCi/l assigned to 41 States for dose calculations is probably high for LPG produced in the United States.

However; as noted previously in reference to figure 10, the average radon content of northern and eastern states may be greater than 10 pCi/l due to use of Canadian LPG. Since no data are available on radon in LPG consumed in these states, an estimate of the significance of dose from Canadian LPG was made by assuming a radon content of 50 pCi/l for LPG in each of 16 states near the Canadian border. Another 10 states adjacent to these were assigned values of 30 to 40 pCi/l. With these radon concentrations an additional population T-B dose equivalent of about 8600 person-rem/year was estimated. This additional dose could potentially raise the total estimated health effects by 0.3 excess lung cancers per year in the United States.

## Comparison With Other Sources of Radiation

Radon-222 in LPG is by no means the only contributor of radiation dose to the respiratory system. Other sources of radon-222, as well as radon-220 (thoron), contribute alpha radiation dose to the bronchial epithelium of the lung. Doses are also received from natural terrestrial beta and gamma radiation, cosmic radiation, internally deposited isotopes of the thorium and uranium decay series, and potassium-40. Additional radiation doses to the population are received from man-made sources, including medical radiation, fallout radiation, and nuclear facilities. In order to place the contribution of dose from radon in LPG in better perspective, the doses to persons exposed or individuals-at-risk and the doses to the U. S. population-at-risk were estimated for various radiation sources as noted in table 20.

It is recognized that there are a number of limitations which should be noted when attempting to compare doses from various sources. In particular, doses to the lung or bronchial tissues are not directly comparable to whole body doses which include doses to other tissues and organs as well. However, the dose estimates in table 20 do allow a number of observations.

For example, it is readily seen that background radon-222 contributes by far the greatest dose to the lung of all sources. Since the population-at-risk for background radon includes the entire U. S. Population, then the population dose is also the largest. Also, radon from natural gas contributes almost 100 times as much dose as from LPG on a national basis.

## Interpretation of Estimated Health Effects

Since no lung cancers have been reported for exposure to radon daughters at less than 0.33 WL a year, it should be emphasized that the health effects estimate in this study is a statistical projection only for assessing potential effects on large populations. This estimate is based on the assumption of a linear non-threshold dose response to alpha radiation from radon daughters.

In this study, the use of LPG in both kitchen ranges and space heaters in the average home would result in less than  $1 \times 10^{-5}$  WL. Furthermore, the estimate for this analysis of one excess lung cancer a year is probably high according to the analysis of uncertainties outlined in detail by Johnson, et al. (5) in the natural gas study. Using the approach from that study, the health effects estimate for LPG would be reduced to much less than one effect a year. (This estimate would not be affected significantly by contributions from Canadian LPG). Therefore, it can be concluded that the use of LPG containing radon-222 does not contribute to the incidence of lung cancer in the United States.

The evaluation of control costs for reducing the estimated health effects from radon in LPG indicate that 50 million dollars or more would be required for each health effect. Such an expenditure would not be cost effective in terms of the possible reduction in health effects. Consequently, it is not considered necessary to control the content of radon in LPG.



Table 20. Comparison of bronchial epithelium doses from various sources

Source	Average <sup>(a)</sup> individual dose mrem/yr	U.S. Population <sup>(b)</sup> dose person-rem/yr	Ref <sup>(c)</sup>
Radon-222 in LPG <sup>(d)</sup> (e)			
kitchen ranges	0.87	18,400	
space heaters	<u>3.70</u>	<u>10,600</u>	
Total	4.57	29,000	
Radon-222 in natural gas <sup>(d)</sup>			
kitchen ranges	12.0	1,499,200	(5)
space heaters	<u>43.2</u>	<u>683,200</u>	
Total	55.2	2,182,400	
Background radon-222 at 0.13 pCi/l <sup>(d)</sup>	520	109,200,000	(1)
Natural terrestrial radiation	40	8,400,000	(44)
Cosmic radiation <sup>(f)</sup>	44	9,240,000	(44)
Internally deposited potassium-40	17	3,570,000	(45)
Fallout (1969) <sup>(f)</sup>	4	840,000	(45)
Diagnostic radiography (1970) <sup>(g)</sup>	153	11,536,200	(45)
Nuclear power reactors <sup>(f)</sup>	0.056	1,650	(46)

- (a) Dose to persons exposed or individuals-at-risk.  
(b) Summation for U.S. population-at-risk for each source of exposure.  
(c) References from which dose information was derived.  
(d) Dose conversion factor, 1 pCi/l = 4 rem/yr.  
(e) Not including possible contribution from Canadian LPG.  
(f) Lung dose assumed equal to whole body dose.  
(g) Lung dose assumed equal to abdominal dose.

## CONCLUSIONS

The conclusions from this assessment of potential radiological health effects from radon in liquefied petroleum gas are summarized as follows:

- (a) The use of LPG containing radon-222 in average homes with unvented kitchen ranges and space heaters does not contribute to lung cancer incidence in the United States.
- (b) Controls for reducing radon concentrations in LPG by storage methods would cost over \$50 million for a reduction of one potential excess lung cancer. Therefore, it would not be cost effective to require controls on radon in LPG by storage on a national basis.
- (c) High concentrations of radon-222 have been measured in LPG, but distribution and storage times allow most of the radon to decay prior to use of the LPG.
- (d) The average population tracheobronchial dose-equivalent was estimated to be 29,000 person-rem/year.
- (e) The average dose to individuals and the U. S. population-at-risk from radon in LPG is very small compared to other natural and man-made sources of ionizing radiation.
- (f) Individuals could possibly receive dose equivalents greater than 500 mrem/year. However, existing data are not sufficient to determine the significance of such potentially high individual doses. This matter will be further evaluated by ORP.

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