DANA P. LOOMIS. An Investigation of Radon in North Carolina Groundwater and Its Relationship to Rock Type. (Under the direction of JAMES E. WATSON).

Previous studies of radon-222 in North Carolina groundwater supplies have shown that the state has some of the highest radon concentrations in the United States. Reanalysis of existing environmental sampling data from 272 public water systems shows that the distribution of radon in North Carolina follows distinct geographical and geological patterns; a simple average concentration based on these samples would not provide a meaningful estimate of public exposure to radon.

Variations in radon concentration are associated, in particular, with rock type. The highest radon concentrations in North Carolina groundwater supplies are found in waters from areas in the Piedmont and Blue Ridge regions underlain by granites, and the lowest concentrations (generally < 500 pCi/l) occur in aquifers of the coastal plain. Concentrations in most of the Piedmont region are intermediate (generally between 1000 and 5000 pCi/l). There appears to be no systematic relationship between radon concentration and water system size in North Carolina.

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INTRODUCTION

Environmental radon in air and water has recently been recognized as a major source of public exposure to ionizing radiation, largely as a result of measurements showing that radon concentrations in drinking water supplies and indoor atmospheres are high enough to produce a radiation dose significantly above that from outdoor background levels (e. g. Gessell, 1983; Hess et. al., 1985; Nero, 1983). Radon which may enter buildings in drinking water is transferred to air by household water uses such as showering and cleaning (Gessell and Prichard, 1980). In this way radon in drinking water may cause three types of radiation exposure which can potentially affect human health; an external whole-body dose, an ingestion exposure through drinking radon-bearing water, and an inhalation exposure from breathing the radon emitted to the indoor atmosphere. The airborne exposure pathway is of the greatest public health concern because the inhalation dose is larger by far than the dose from other pathways (Cross et al., 1985).

Radon may enter indoor air by direct emanation from the ground, from outdoor air, and from contaminated building materials as well as from water; the relative importance of radon contributed by these sources is not well understood.

Even if other sources of indoor radon are disregarded, however, many water supplies contain sufficiently high radon concentrations to provide a radiation dose which exceeds that from other environmental sources, both natural and artificial (Cross et al., 1985). Because of the widespread occurrence and often high concentrations of radon in ground water supplies this radiation dose presents a significant risk of lung cancer to the U.S. population. The lifetime lung-cancer risk from radon in drinking water has been estimated (e. g. Cross et al., 1985; Hess et al., 1985), although with considerable uncertainty (Crawford-Brown and Cothern, 1985), as approaching the total risk of lung cancer among U.S. non-smokers for radon concentrations which are common in North Carolina groundwaters, and may be much higher for individuals using waters with extremely high radon concentrations. Because of the magnitude of this estimated risk, it is important to understand the natural factors which control the distribution of environmental radon and are responsible for high radon concentrations in groundwater supplies.

North Carolina has some of the highest concentrations recorded in groundwater in the United States (Hess et. al., 1985; Horton, 1983; Sasser and Watson, 1978). Earlier studies of radon in North Carolina (Aldrich et. al., 1975; Sasser and Watson, 1978; U.S. EPA, 1982) showed that concentrations in the state range from near zero to over 46,000 pCi/l, but the distribution of radon concentration and the factors that control it have not previously been investigated on a statewide basis.

This study was undertaken for the Groundwater Section of the North Carolina Department of Natural Resources and Community Development to gain a better understanding of the distribution of radon in North Carolina groundwater, and to examine the relationship between radon concentration and aquifer lithology to determine if lithology could be used as a criterion for the identification of areas in North Carolina where groundwater use might be impacted by high radon concentrations. Reanalysis of radon data from other studies (Aldrich et. al., 1975; U.S. EPA, 1982) shows that radon concentration is related to aquifer lithology, and that distinct average radon concentrations are associated with major rock types. Groundwater from the North Carolina coastal plain has the lowest average radon level in the state, while the Piedmont and mountain regions have higher levels. The highest concentrations occur in areas in the Piedmont underlain by granitic rocks. These broad lithologic associations also imply variation in other geologic and hydrologic variables which are related to rock type, such as grain size, porosity, and groundwater flow rate (Tanner, 1964a).

This knowledge can be used for water management- and health-related purposes which include identifying high-risk areas, estimating radon concentrations in water supplies such as private wells which are not routinely monitored for radiological quality and estimating the cost and effictiveness of potential water quality standards for radon. In addition, the understanding gained through this investigation of the broad geologic affinities of radon in North Carolina groundwater may help to identify other factors which account for variability in radon concentration.

RADON IN DRINKING WATER

Radon in U.S. Drinking Water Supplies

The distribution of radon in U.S. drinking water supplies has been reported by the U.S. Environmental Protection Agency (U.S. EPA, 1982; Horton, 1983) and by Hess et al. (1985). Radon concentration in drinking water varies over several orders of magnitude, from near zero to over 100,000 pCi/l. In general, surface waters contain virtually no measurable radon, but high concentrations may occur in groundwaters. The highest concentrations recorded in the U.S. are in private wells in Maine and New Hampshire which exceed 300,000 pCi/l (Brutsaert et al., 1981). Table 1 shows geometric mean radon concentrations in public and private groundwater supplies from 40 states. The highest concentrations in groundwater occur, in addition to New England, in the Appalachian states, the Rocky Mountain states, and California (Hess et al., 1985; Horton, 1983). Groundwaters in the Atlantic-Gulf coastal plain and the midwest region have substantially lower radon concentrations

Table 1. Radon in U.S. drinking water supplies, modified from Hess et al. (1985).

Geometric mean Rn-222 in pCi/l; number of samples in parentheses.

STATE	PRIVATE WELL		PUBLIC GROUNDWATER SUPPLY	
AL	120	(22)	70	(132)
AR	230	(2)	12	(22)
AZ	-		250	(124)
CA	43	(6)	470	(15)
CO	-		230	(76)
DE	-		30	(72)
FL	6000	(34)	30	(327)
GA	2100	(2)	67	(225)
IA	-		220	(85)
ID	-		99	(155)
IL	-		95	(314)
IN	-		35	(185)
KS	-		120	(47)
KY	1500	(10)	32	(104)
MA	1000	(8)	500	(212)
ME	7000	(24)	-	
MN	1400	(1)	130	(233)
MO	0	(2)	24	(138)
MS	-		23	(104)
MT	4300	(8)	230	(71)
ND	-		35	(133)
NH	1400	(18)	940	(52)
NJ	-		300	(38)
NM	59	(59)	55	(171)
NV	-		190	(57)
NY	1500	(4)	52	(292)
OH	-		79	(165)
OK	-		93	(33)
OR	450	(18)	120	(69)
PA	910	(16)	380	(105)
RI	6500	(69)	2400	(575)
SC	1100	(23)	130	(384)
SD	4300	(2)	210	(155)
TN	0	(2)	12	(98)
UT	-		150	(195)
VA	560	(42)	350	(284)
VT	210	(23)	660	(71)
WI	730	(40)	150	(278)
WY	-		330	(32)
US	920	(434)	130	(6298)

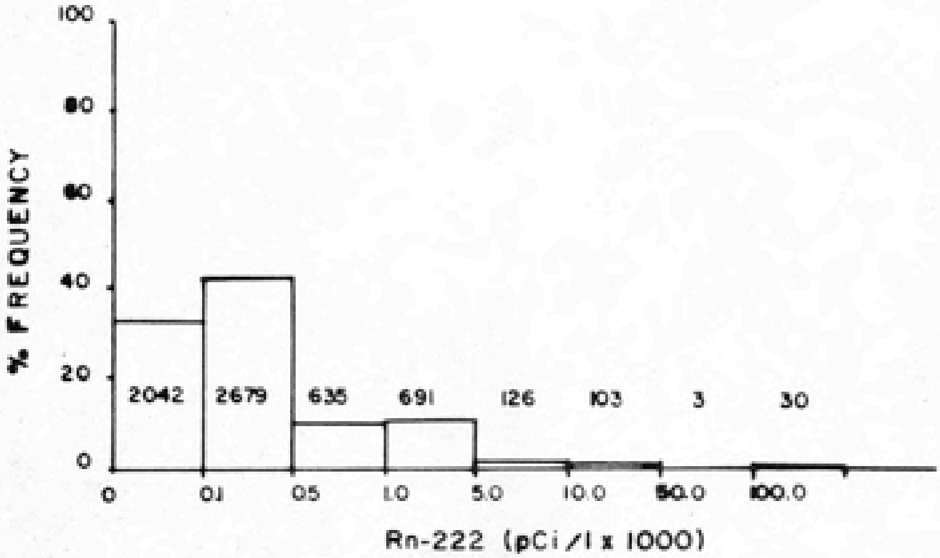
(Hess et. al., 1985; Horton, 1983).

In addition to this geographic variation, nationwide studies suggest that radon concentration varies among water systems of different sizes. Radon levels in private wells are often several times concentrations in public water supplies (Table 1), and among public water systems radon concentration tends to decrease as the size of the population served increases (Hess et al., 1985). This systematic difference between small and large water systems is apparently due to the longer retention times of water in large systems which allow some fraction of original radon to decay or escape to the atmosphere before the water is used (Brutsaert et al., 1981; Hess et al., 1985).

The frequency distribution of radon in U.S. public groundwater supplies is shown in Figure 1. The figure shows that radon concentrations in most of these systems (85%) is below 1000 pCi/l and that a substantial proportion (32%) are below 100 pCi/l. Although the percentage of water systems having extremely high radon concentrations over 10,000 pCi/l is quite small, users of these waters bear a risk far above associated with average levels of radon exposure.

Although the radon content of drinking water varies geographically and with water system size the largest variations are related to geologic factors. Radon in water has been shown, in particular, to be strongly associated with rock type. Because radon concentration is so intimately related to geologic variables average radon

Figure 1. Frequency of Rn-222 concentrations in U.S. public water supplies (modified from Hess et al., 1985).



concentrations and their associated radiation exposures must be determined in respect to these factors.

Sources of Radon in Groundwater

The occurrence of radon in water is governed in large part by the concentration of its parent nuclides, uranium and radium, in rock and soil, and by physical variables which control the emission of radon from solid materials into water. The concentration of parent nuclides in rocks is a function of geologic history and of the geochemistry of the uranium decay chain. Uranium is widely distributed in the Earth's crust, and average uranium concentrations in many minerals and rocks are well known. The following abundances are from Rogers and Adams (1969); uranium concentrations from 1 to 10 ppm are characteristic of silicic igneous rocks, (granites, rhyolites, quartz monzonites, etc.); somewhat lower concentrations are charactristic of intermediate rocks, and much lower concentrations, typically from .001 to 1 ppm, are found in mafic and ultramafic rocks. Variation between different rock bodies of the same type, or even within a single pluton, may be considerable, however. Sedimentary rocks have generally lower values, for example 0.45 ppm for quartz sandstones and 2.2 ppm for limestones, but marine phosphorites and some unusual shales may contain concentrations in excess of those found in normal granites. The uranium content of metamorphic rocks is quite variable as might be expected from their diverse origins.

Factors affecting radon concentration

Patterns of radon occurrence in groundwater reflect, in general, the major differences in the uranium content of common rocks. The consistent association of concentrations exceeding 100,000 pCi/l with granitic terranes has been observed repeatedly (Asikainen and Kahlos, 1979; Brutsaert et. al, 1981; Snihs, 1973). Lower concentrations (generally < 500 pCi/l) have been observed in various sedimentary rocks (Andrews and Wood, 1972; Gorgoni et al., 1982; Fukui and Katsurayama, 1983; King et al., 1982; Mitsch et al., 1984). It might appear from these relationships that rock type is the determinant of radon concentration in water. In fact, many variables, only some of which are covariates of lithology, intervene between decay of uranium and measurement of radon in a well water sample. These intermediate factors include

1) Geometry and mass of the radon source. Radon concentrations in water may be affected not only by the concentration of parent nuclides in surrounding rocks, but by the absolute quantity of parent present. The ability of radon to enter water may be affected, in addition, by the spatial distribution of nuclides within the rock.

2) Uranium-radium geochemistry. The extent to which radium, the immediate parent of radon, is in equilibrium with its progenitor uranium in geologic materials is largely a function of the degree to which the parent and its daughter products have been separated by geochemical

processes. Few data are available on radium in common rocks, but its chemical behavior is quite different from that of uranium, so geochemical separation of parent and daughter might be expected in certain situations.

3) Emanation Fraction. Not all of radon produced by the decay of radium in rocks escapes into air or water; the ratio of radon escape to production is frequently called the emanation fraction. The emanation fraction has been measured in the laboratory for a variety of minerals, rocks, and soils and varies considerably among mineralogic and lithologic types irrespective of uranium concentration (Baretto, Clark, and Adams, 1975). Emanation fraction is inversely proportional to grain size (Andrews and Wood, 1972), and is enhanced by deep weathering or pervasive microfracturing, and when parent nuclides are distributed near the surface of mineral grains (Tanner, 1964a).

4) Dissolved radium. Radon may also be concentrated in water by decay of radium already in solution. In general, dissolved radium levels are far lower than needed to support observed radon concentrations and most investigators have concluded that there is no relationship between dissolved radium and radon (Tanner, 1964b; Snihs, 1972). No such relationship has been observed in studies of radium and radon in most areas in North Carolina (Lee, Watson, and Fong, 1979), but correlation between Ra and Rn has been reported in the eastern Phosphate district of North Carolina (Strain, Watson, and Fong, 1979) and between logRa and logRn in South Carolina (King, Michel, and Moore, 1982).

5) Radon Migration. Radon atoms which enter water-filled pores may move from their original site by diffusion, in which the radon atom moves relative to the water, or by transport, in which the radon atom is carried by moving water. Because radon is chemically inert it is not removed from groundwater by process other than diffusion and radioactive decay. The factors which affect radon migration through rocks are primarily hydrologic ones; these factors include the diffusion constant of radon in water, degree of saturation, groundwater flow rate, and the size and geometry of pore spaces in the rock matrix (Tanner, 1964a). Migration is more rapid through fractures than in intergranular porosity. In saturated porous rock and soil, diffusion is inefficient and groundwater flow is the dominant factor in radon migration (Tanner, 1964a); the amount of radon transported from its original site is determined by the balance between flow rate and decay in which high flow rates favor the maintenance of radon activity near its original level (Andrews and Wood, 1972).

6) Temporal variations. Several investigators have reported conflicting findings on the temporal stability of radon concentration in wells repeatedly sampled over time. Michel and Moore (1980) reported stable radon concentrations during one year of observation of wells in the South Carolina coastal plain, but variation by a factor of 2-3 was reported in continuously monitored wells in Japan (Fukui and Katsurayama, 1983) and in England (Andrews and Wood, 1972).

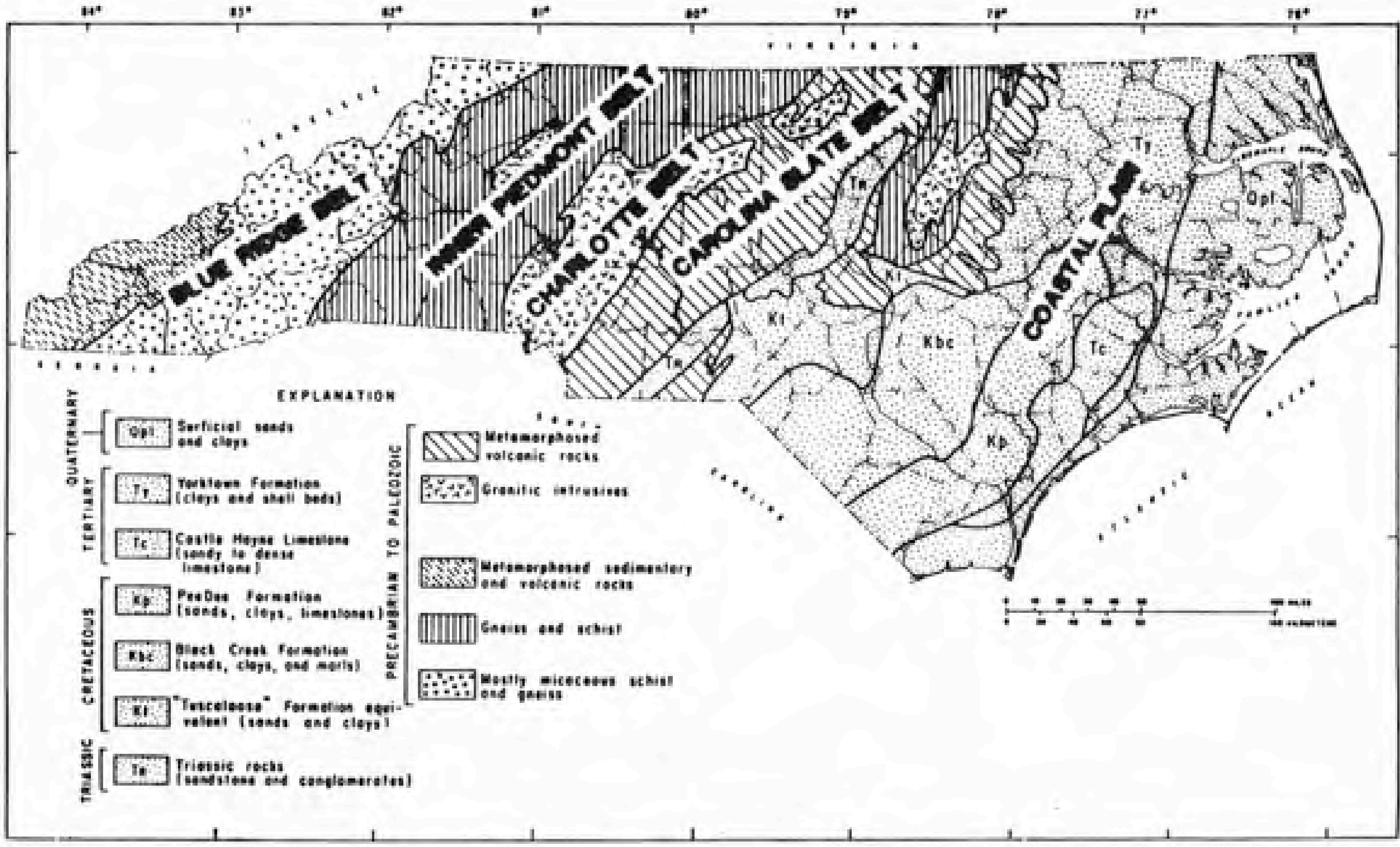
Radon concentration in these wells was positively correlated with rainfall, suggesting, perhaps, more efficient transport due to higher flow rates.

7) Well and water system design and use. Since groundwater samples must, in general, be obtained from wells, a number of variables related to well design and use may also affect the radon content measured in water samples. In addition to the inverse relationship of radon concentration and water system size (Hess et. al., 1985), Brutsaert et. al. (1981) reported a negative correlation between radon concentration and yield of 136 wells in Maine, and a positive correlation between radon concentration and well depth. Snihs (1973) found no evidence for the latter relationship in 37 wells in Sweden. Well pumping patterns may also affect the radon concentration of water samples; Fukui and Katsurayama (1983) reported small but consistent increases in radon activity after several hours pumping, presumably because induced flow brought new radon-laden water to the wellbore.

NORTH CAROLINA GEOLOGY AND HYDROLOGY

North Carolina can be geologically divided into a series of northeast-trending belts as shown in Figure 2. The boundaries of the major belts correspond to the three natural physiographic regions of North Carolina; the coastal plain, the Piedmont, and the Blue Ridge region. About half of the state is covered by late-Mesozoic and Cenozoic

Figure 2. Geologic map of North Carolina (modified from Heath, 1980).



sedimentary rocks of the Atlantic coastal plain; these strata form a sediment wedge which thickens eastward to over 10,000 ft at Cape Hatteras (Stuckey, 1965). The coastal plain sequence is divided into many formally-named geologic units, but a somewhat simpler system of nomenclature is generally used in hydrologic reports. Groundwater is obtained from four major aquifers in the coastal plain, which are known informally as the Cretaceous aquifer, the Castle Hayne aquifer, The Yorktown aquifer and the surficial aquifer, in decreasing order of depth (Heath, 1980). The relative positions of these aquifers in the geologic sequence reflect the order of their formally-designated counterparts, but the boundaries of these aquifers do not correspond precisely to those of the formal geologic units.

The Castle Hayne aquifer is the principal water source in the eastern coastal plain (Cederstrom et al., 1979; Heath, 1980). It is a confined aquifer of semi-consolidated limestone and shell beds which stores and transmits groundwater in voids created by limestone solution. Large wells which may yield over 1000 gal/min are common in the Castle Hayne aquifer (Heath, 1980). In the western portion of the coastal plain the Castle Hayne is absent and groundwater is obtained instead from sand and gravel aquifers in the Cretaceous, Yorktown, and surficial units. Many wells in this area are completed in multiple water-bearing zones and may mix groundwaters from aquifers of different ages and lithologies. The Cretaceous aquifer is particularly important in the southern portion of the inner coastal plain (Heath, 1980) where the Yorktown Formation has been removed by erosion (Stuckey and Conrad, 1958). The aquifer consists of three formal geologic units; the Tuscaloosa Formation, the Black Creek Formation and the Peedee Formation (Heath, 1980). The Peedee and Black Creek formations are lithologically complex units containing sands, clay, and shell beds (Stuckey and Conrad, 1958), but are not highly productive as aquifers (Cederstrom et al., 1979). The name Tuscaloosa is to be eliminated from formal use on the latest geologic map of North Carolina (in press). Nevertheless, because of its long use in the state and its frequent citation in hydrologic reports it will be used throughout this report. The Tuscaloosa is a highly productive sand aquifer which is a major groundwater source throughout the Atlantic-Gulf coastal plain (Cederstrom et al., 1979). Where it is exposed in North Carolina the Tuscaloosa consists of sedimentary material derived from erosion of Piedmont crystalline rocks, including granites, gneisses, and schists (Stuckey and Conrad, 1958).

The sediments of the coastal plain overlie much older Precambrian and Palezoic igneous and metamorphic rocks which emerge from beneath the coastal plain sequence along a northeast-trending zone called the fall-line. These complexly faulted and deformed rocks form the bedrock of the Piedmont region, and include the rocks of the Carolina Slate Belt and the Inner Piedmont Belt (Fig.2). The Carolina Slate Belt consists of low-rank metavolcanic and associated metasedimentary rocks, while the Inner Piedmont belt consists principally of gneisses and schists (King, 1955) metamorphosed up to the sillimanite zone (Butler, 1972; Overstreet, 1955). These rocks are intruded by clusters of Paleozoic plutons (Fullagar, 1971) which form the Raleigh belt and Charlotte belt (Fig. 2). These plutonic rocks are primarily granitic, but the Charlotte belt also includes significant areas of mafic igneous rocks (King, 1955). In several areas rocks of the Carolina Slate Belt and Inner Piedmont Belt are overlain by unmetamorphosed sedimentary rocks of Triassic age, deposited in northeast-trending basins (Fig. 2) bounded by faults. The lithology of the Triassic rocks is variable and includes sandstones, conglomerates, shales, siltstones and small amounts of limestone and coal (Stuckey, 1965). Small areas underlain by other rocks, including marbles and quartzites and conglomerates are also present in the Piedmont (Overstreet, 1955).

The Blue Ridge Belt in the far west is underlain by Precambrian granites and gneisses and Precambrian to early Paleozoic metasedimentary rocks (King, 1955). All of the Blue Ridge Belt rocks are metamorphosed to some extent. Blue Ridge granites and gneisses are similar in general to those in the Piedmont, and vary in degree of metamorphism from biotite to sillimanite zone (Butler, 1972). The Precambrian metasedimentary rocks of the Blue Ridge belt are significantly different from other metasediments in North Carolina, however. These rocks underlie a large area in southwestern North Carolina including Cherokee, Graham, and Swain counties and adjacent areas (Fig. 2). These rocks, formally assigned to the Ocoee Supergroup, were derived from the erosion of crystalline rocks similar to the granites and gneisses now exposed in the Blue Ridge, and their bulk composition is approximately that of granitic igneous rocks (Hadley, 1970). Small areas of other metasedimentary rocks are also exposed in the Blue Ridge Belt (King, 1955; Stuckey and Conrad, 1958).

Although the geology of the Piedmont and Blue Ridge regions is quite complex the hydrologic behavior of the major rock units is so similar that they can be regarded as a single hydrologic region (Heath, 1980). The crystalline rock aquifers of the Piedmont and Blue Ridge regions are much less productive than coastal plain sediments; typical well yields in the Piedmont are less than 100 gal/min (Cederstrom et al., 1979). In areas underlain by crystalline rocks groundwater is stored primarily in the saprolite (highly weathered rock material) which overlies bedrock and transmitted to discharge areas and wells by networks of fractures within the bedrock (LeGrand, 1967; Heath, 1980). Well yields tend to be higher in areas of dense fracturing where fractures are interconnected (Cederstrom et al., 1979). In general, rocks of the Ocoee Series and the Inner Piedmont Belt are the most productive hydrologic units in the region and the Triassic basins and Carolina Slate Belt are least productive (Heath, 1980).

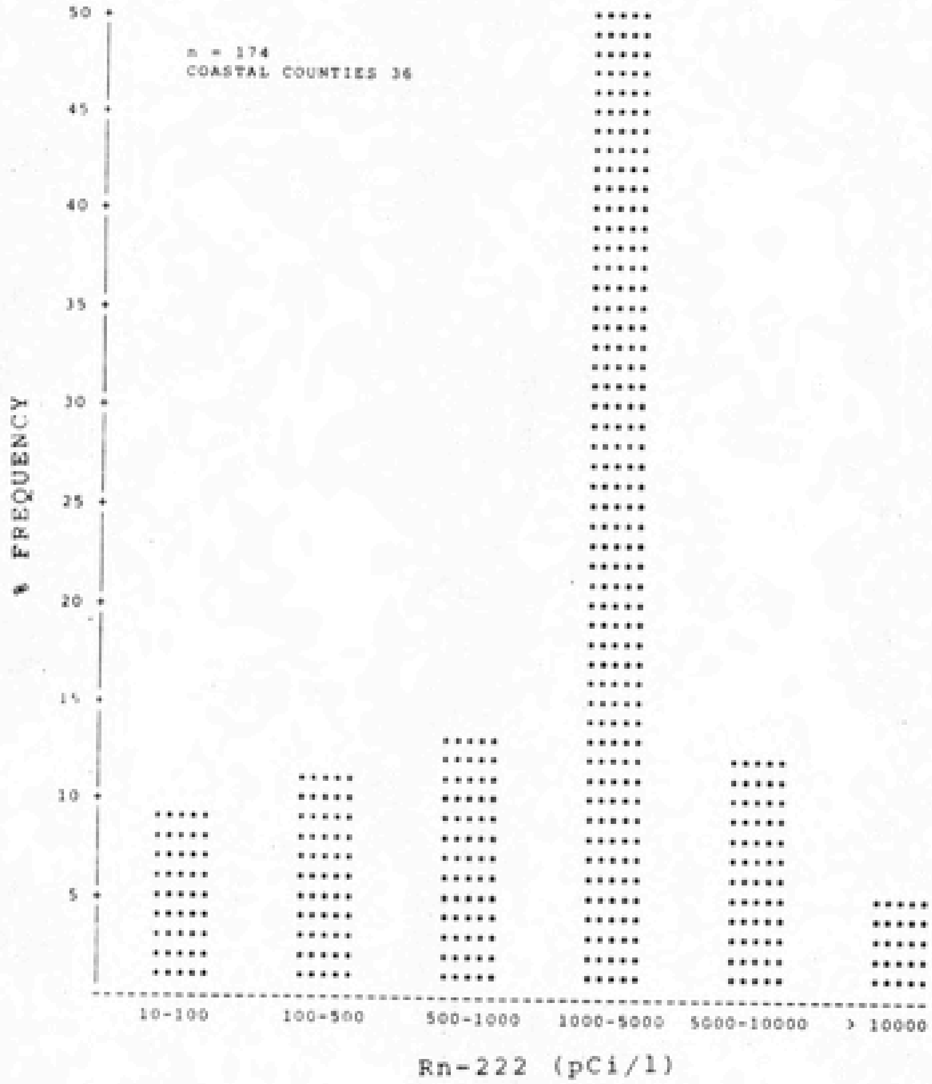
PREVIOUS RESEARCH

Several studies have been conducted on radon in North Carolina groundwater. Strain et al. (1979) and Mitsch et al. (1984) examined the local effects of phosphate ore extraction and processing in eastern North Carolina. Mitsch et al. found large variations in the radon content of groundwater samples from 116 wells producing from several aquifers in the phosphate district, but concluded that there was no pattern of radon distribution which could be attributed to the activities of the phosphate industry. The occurrence of radon in groundwater has been surveyed statewide in two environmental sampling programs; one conducted by the Radiation Protection Section of the North Carolina Department of Human Resources (Aldrich et al., 1975; Sasser and Watson, 1978) and the other by the U.S. Environmental Protection Agency (U.S. EPA, 1982; Horton, 1983).

Both statewide studies showed that measurable quantities of radon are present in many North Carolina groundwaters, and that the concentration of radon in these waters has a range of at least four orders of magnitude, from near zero to over 46,000 pCi/l. Geological data were not systematically collected and no geologic analysis of the radon data was presented in reports of either study. In spite of the simlar conclusions which may be drawn from these two studies, they differ significantly in intent and methodology.

Differences in sampling philosophy, in particular, appear to have had a major effect on the composition of the two data sets. The North Carolina Department of Human Resources (DHR) study of radon (Aldrich et al., 1975) was intended primarily to identify areas of high radon concentration. The investigators used prior knowledge to locate and sample wells where radon concentrations could be expected to be high (Aldrich et al., 1975; Felix S. W. Fong, personal communication, 1985). The locations sampled in this study are predominantly in the Piedmont and Blue Ridge regions of North Carolina, and are clustered in several counties which account for a large proportion of the sample. Wake County contains 10% of the locations and Catawba, Rockingham, Rowan, and Surry counties were also heavily sampled. The field procedures used in the DHR study were designed to obtain a sample of water as produced at the well. The reported concentrations can therefore be expected to represent the maximum concentrations to which users of water from the sampled wells could be exposed. Only groundwater supplies were sampled in the NC DHR study, and most of the samples were taken from small public water systems serving several hundred people, or less, such as trailer parks and subdivisions. Because of the selection of areas of known high radon concentration, the predominance of sample locations in the Piedmont and the generally small size of the water systems surveyed data from

Figure 3. Frequency of Rn-222 concentrations in NC DHR data of Aldrich et al. (1975).

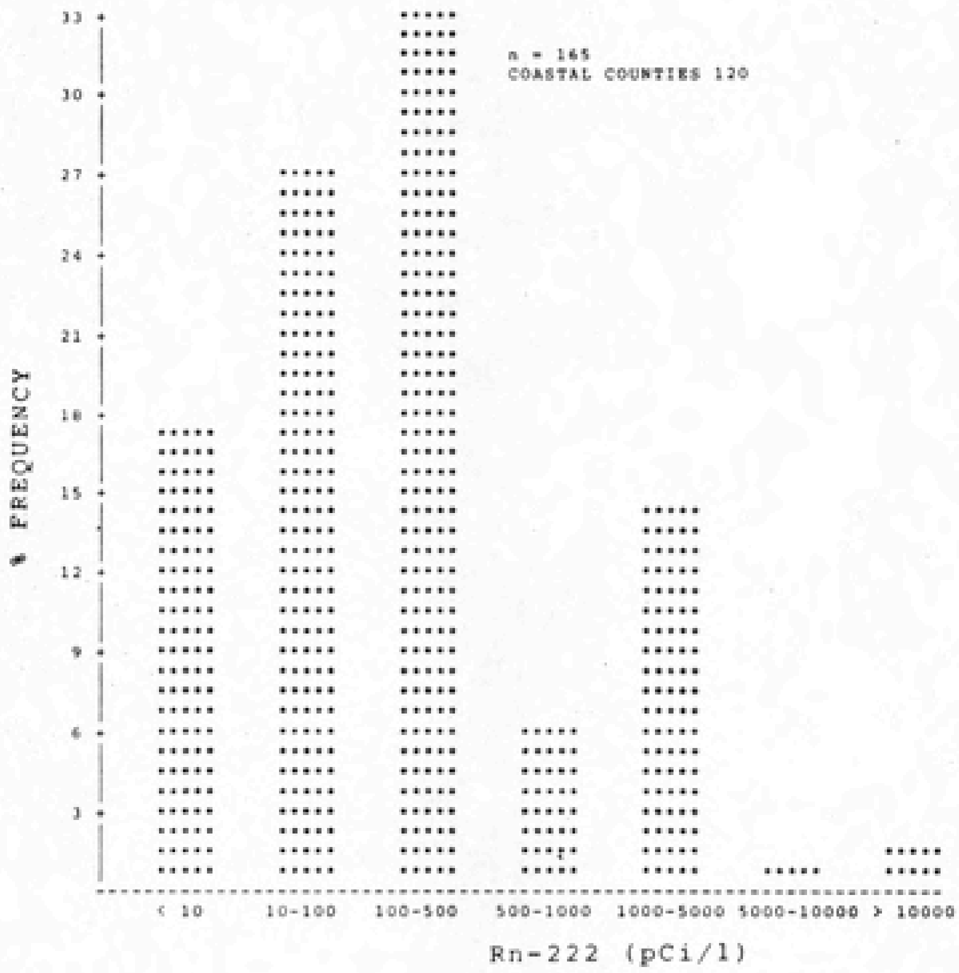


the DHR study could be expected, *a priori*, to be biased toward high radon concentrations. Figure 3 shows the frequency distribution of radon concentrations from this study and the number of observations from coastal versus Piedmont and mountain counties. The expected predominance of high values is manifested in the large number of observations in the 1000-10,000 pCi/l range.

The EPA study of radon in North Carolina groundwater was part of a nationwide program to estimate population exposure to radon and other naturally-occurring radionuclides in public water supplies (Horton, 1983). The design of the EPA study emphasized collecting a sample representative of the water actually consumed by the majority of groundwater users in the United States. EPA's sampling criteria called for the collection only of finished water from public systems serving 1000 people or more. Samples were collected as close to the water source as possible, to minimize the effects to radioactive decay, and composite samples were taken from multiwell systems. In North Carolina several of the systems sampled did use surface water or a mixture of surface and groundwaters, and many were smaller than the 1000 customer criterion. Actual sample collection was done for EPA by the Radiation Protection Section of NC DHR.

EPA's sampling criteria resulted in the collection of a more geographically uniform sample than in the DHR study, but most of the sample locations nevertheless tended to be

Figure 4. Frequency of Rn-222 concentrations in US EPA (1982) data.



in the coastal plain counties (Fig.4), perhaps because of the relative scarcity of large groundwater systems in the western portion of North Carolina. Other studies indicate that radon concentrations are generally low in the Atlantic coastal plain (Hess et al., 1985; Horton, 1983; King et al., 1982). As expecxted the EPA data (Fig. 4) shows mostly low radon concentrations.

In addition to these large differences in sampling criteria, the DHR and EPA radon studies also employed different analytical methods for determining radon activity. EPA used a liquid scintillation counting technique, while NC DHR use a gas-emanation (Lucas cell) technique. Although measurements by the two techniques should be comparable and calibration tests performed for the EPA research (Horton, 1983) indicate no significant differences between analyses measured with liquid scintillation versus other techniques, the comparabitly of North Carolina data using different analytical methods has been questioned (Fong and Penny, 1981). The comparability of these methods is discussed further in the description of methods used in the present study.

METHODS

The conclusions of this study are based on analyses of all available North Carolina environmental radon data collected in earlier statewide studies (Aldrich et al., 1975; Sasser and Watson, 1978; U.S. EPA, 1982). Names and localities of the sampled water systems were obtained directly from NC DHR and EPA. EPA also provided field data sheets for each water sample, giving a brief description of the sampled water system including number of wells in the system, number of individuals served, and for some systems, well depth and pumped aquifer. All of the water samples were collected from public water systems, so detailed information on system location, water source, and number of customers could be obtained through the Water Supply Branch of NC DHR, which maintains extensive records on all public water systems. Systems listed in Water Supply Branch records as using surface water or a mixture of surface water and groundwater were eliminated from the data set, leaving a working data set of 295 radon analyses.

For geologic analysis, radon concentrations were plotted on a 1:500,000 geologic map of North Carolina (N.C. Dept. of Conservation and Development, 1958) using well location information from records of the NC DHR Water Supply Branch and well completion reports obtained from the

Groundwater Section of the NC Department of Natural Resources and Community Development (NC DNRCD). Each of 272 samples which could be plotted on the map was assigned to one of the lithologic categories in Table 2 according to its location on the geologic map. An initial attempt was made to obtain lithologic descriptons of aquifers in each of the sampled wells through the well records of the NC DNRCD Groundwater Section, but this procedure had to be abandoned because data were only available for a small proportion of the wells and these were mostly in the form of unreliable drillers' logs. Although detailed lithologic descriptions of producing aquifers could not be obtained for most of the wells, the units shown on the geologic map are a sufficiently reliable and consistent reflection of the composition of aquifers in the Piedmont and mountain regions where groundwaters are drawn from crystalline rock bodies or from saprolite derived from them. Coastal plain locations were simply assigned to the general coastal plain lithologic group because the data which could be obtained for most wells were insufficiently detailed to allow aquifers in the sedimentary sequence to be differentiated. The practice of combining observations from different aquifers is imprecise, but should not distort the results of this study because differences between the radon concentrations of groundwater samples from the coastal plain relative to other areas can be expected to be larger than differences among the aquifers whch make up the coastal plain group. The few coastal plain wells for which aquifer

Table 2. Lithologic groups

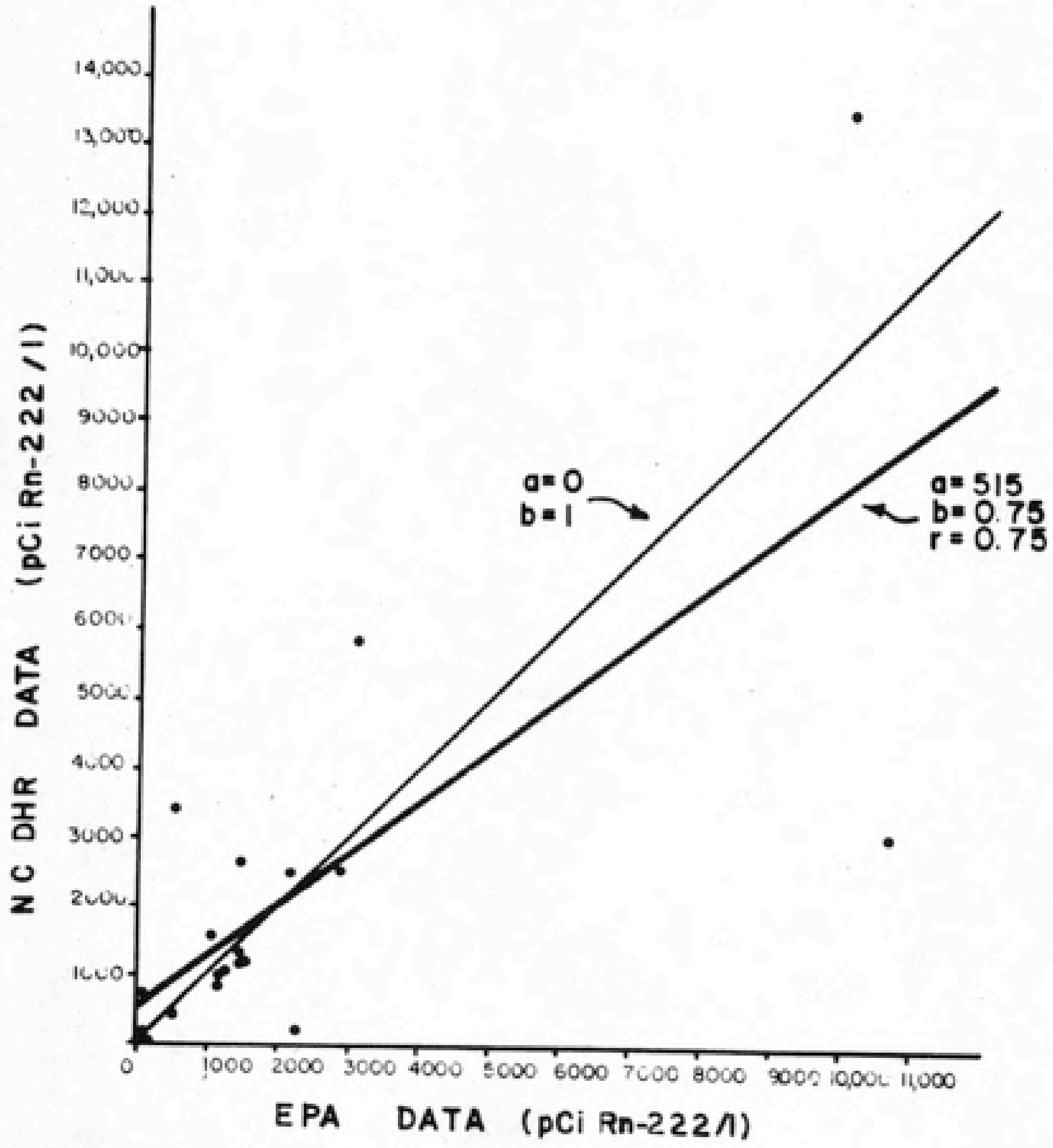
GROUP	DESCRIPTION
Granites	Paleozoic and Precambrian granitoid rocks of the Piedmont and Blue Ridge.
Precambrian sedimentary rocks	sandstone, conglomerate, graywacke and other sedimentary and metasedimentary rocks of the Ocoee Supergroup.
Gneiss & Schist	all gneisses and schists of Piedmont and Blue Ridge, including Brevard Schist, Henderson Gneiss, Cranberry Gneiss, and others.
Metavolcanic rocks	early Paleozoic metavolcanic and associated metasedimentary rocks of the Carolina slate belt.
Mafic igneous rocks	diorite and gabbro of the Charlotte belt.
Metasedimentary rocks	quartzite, marble, and associated metasedimentary rocks of the King's Mountain and Murphy belts.
Coastal plain sediments	all Mesozoic and Cenozoic clastic and carbonate sediments (primarily marine) and surficial deposits of the coastal plain.

data were avaliable were also treated separately with respect to aquifer in the data analysis.

The working data set was also examined to determine if loss of data might have altered its geographic or quantitative distribution. The frequency distribution of the radon concentratons in unaggregated data taken from the NC DHR and EPA studies is virtually the same as the distribution in the original studies. Likewise, the relative proportion of the sample in each North Carolina county is virtually unchanged in the final data set. The close match between the original data and those used in this study with respect to radon concentration and geographic distribution means that the results of the present study are unlikely to be biased by loss of data. This implies, in addition, that differences between the conclusions of this study and any which might be drawn from separate consideration of either primary data set are due to the effects of combining the earlier data and to the different analytic approach of this study.

The two original data sets were also tested for comparability because different methods of sample analysis were used in the NC DHR and EPA studies, as discussed above. This comparison was facilitated by the fact that twenty-one water systems were sampled in both studies. Regression analysis of EPA versus NC DHR results (Fig. 5) yields a slope of 0.75 and a highly-significant ($P < 0.0001$) Pearson correlation coefficient of 0.75. Results from the two methods are highly correlated and although the slope is not

Figure 5. Correlation of NC DHR and US EPA duplicate samples.



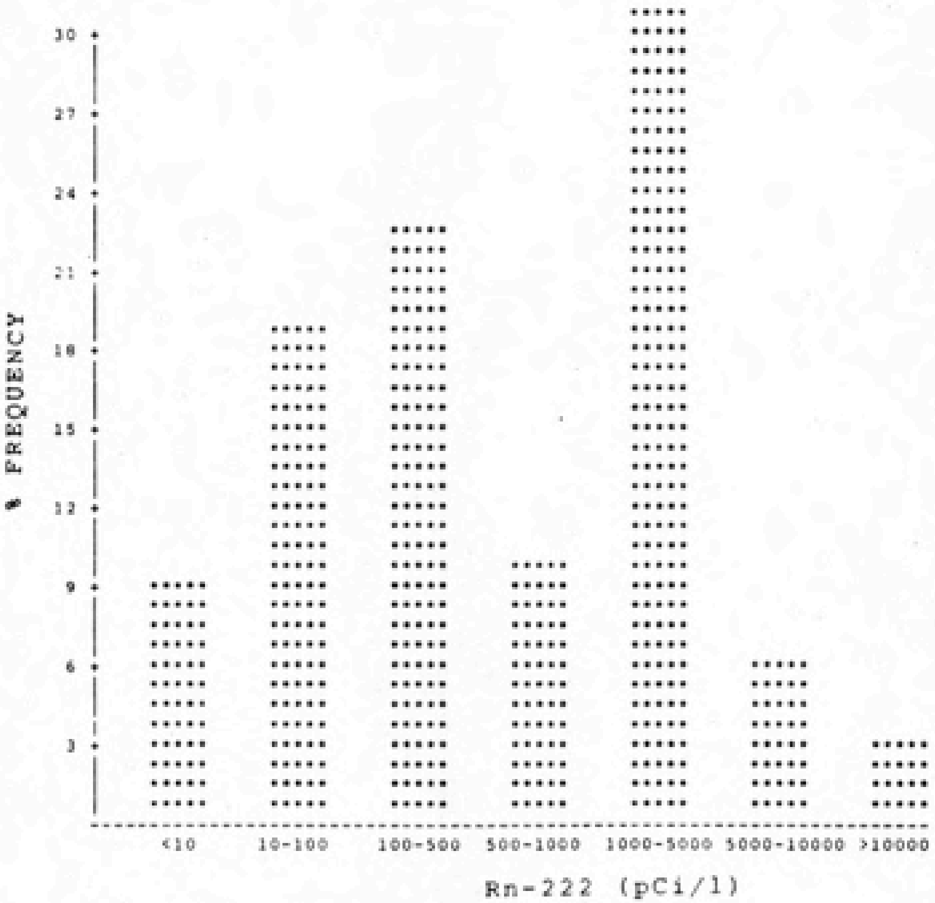
equal to 1 this does not appear to imply that there is a systematic difference in values between the studies. Visual inspection of the plot shows that most of the values fall approximately along a 1:1 line and that the deviation of the calculated slope from 1 is due primarily to several outliers in the data. In addition, a paired t-test for the difference in mean Radon concentration in the systems sampled in both studies yielded a non-significant value of t (t=0.0123), indicating that the data sets are indistinguishable at the 90% confidence level. Because of these results the data from both studies were used in raw form without any adjustment for differences in methodology.

RADON IN NORTH CAROLINA GROUNDWATER

The frequency distribution of radon concentrations in 272 groundwater samples from North Carolina public water supplies is shown in Figure 6. Note that combining data from the earlier studies smoothes the skewed distributions (Figs. 3 and 4) that resulted from sampling bias in these studies.

The distribution of radon in North Carolina is quite different from that for the entire United States (Fig. 1) in that a much higher proportion (41%) of the water systems sampled contain over 1000 pCi Rn-222/l. This difference suggests that population exposure to radon in North Carolina may be higher than average for the United States. A

Figure 6. Frequency of Rn-222 concentrations in North Carolina public groundwater supplies (272 samples).



geometric mean radon concentration of 180 pCi/l for 6300 public water supplies in the United States has been reported by Hess et al. (1985). Although it might be of interest to calculate such an average for North Carolina using these data, the result would not be meaningful because the sample is not randomized on geologic variables. A more meaningful estimate can be made by examining radon concentration in relation to lithology.

<u>Lithology and radon concentration</u>

Average radon concentrations and concentration ranges for eight North Carolina rock groups are shown in Table 3. Most of the radon values (94%) are from coastal plain sedimentary rocks, gneiss and schist, metavolcanic rocks, and granites; these rock types together make up most of North Carolina's land area (Fig. 2). Several other lithologic groups, including Precambrian sedimentary rocks of the Ocoee Supergroup, sedimentary rocks of Triassic basins, and mafic igneous rocks, which account for a minor proportion of the state's area, are also included in the sample.

The lowest radon concentrations are found in the coastal plain. Low values are also associated with mafic igneous rocks (5 observations) in the Charlotte Belt. The highest average concentration, as well as the highest measured values, are found in areas in the Piedmont underlain by granites. Intermediate average radon concentrations occur in the metavolcanic rocks, gneisses,

Table 3. Radon concentration (pCi/l) and lithology.

LITHOLOGY	NUMBER	ARITH. MEAN	GEOM. MEAN	5th PCT.	95th PCT.
GRANITE	24	10562.6	5909.7	515.67	43871.3
PRECAMBRIAN SED.	2	7090.65	5259.99	2335.13	1186.2
GNEISS/SCHIST	71	2244.38	1502.35	170.66	7703.6
METAVOLCANIC	21	1348.86	1183.9	481.52	3354.8
TRIASSIC	6	909.75	499.12	41.69	1766.8
METASEDIMENTARY	4	834.25	645.05	303.22	1510.55
MAFIC	5	527.35	263.99	34.2	1139.00
COASTAL PLAIN	139	426	48.28	ND*	2508.2
CASTLE HAYNE AQUIFER	22	93.6	14.07	ND	642.43
PEEDEE-BLACK CREEK	6	40.55	7.52	ND	178.6
TUSCALOOSA AQUIFER	15	563.01	215.53	ND	2278.37

*Not decectable above background.

TABLE 3

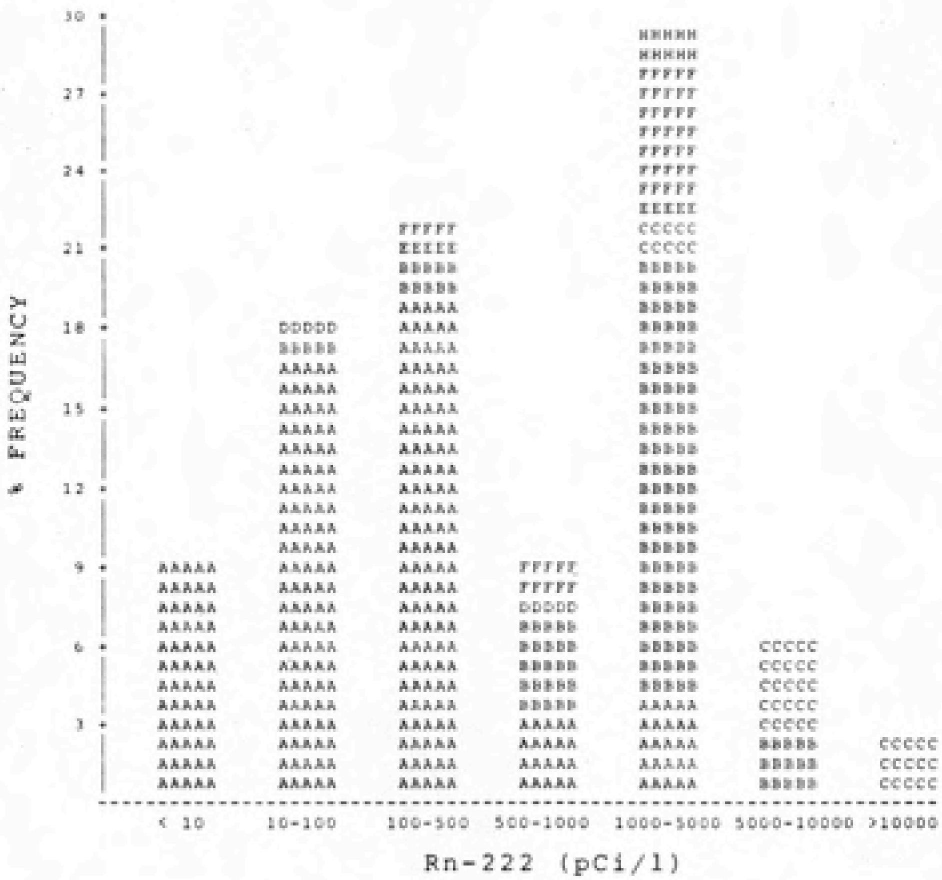
and schists which underlie most of the Piedmont and Blue Ridge regions. Although the radon levels associated with these widely-exposed rocks are low compared to those in granitic areas they are still significantly higher than the U.S. average. Figure 7 shows the influence of the major rock groups on the distribution of radon in groundwater. Observations from the coastal plain dominate the lower part of the concentration range, while values from igneous and metamorphic terranes in the Piedmont and Blue Ridge populate the upper part of the range, and virtually all of the high values over 10,000 pCi/l are associated with granitic areas.

The variation in average radon concentrations shown in Table 3 shows that radon concentration in groundwater is a function of lithology in North Carolina. Other variables, some of which are themselves related to lithology, may also have large effects on radon concentration. One such factor which has been cited by other investigators is water systen size; the relationship of radon concentration with water system size in North Carolina is discussed in the following section.

Effects of water system size

Some investigators have found evidence of an inverse relationship between water system size (i.e., number of individuals served) and radon concentration (Brutsaert et al., 1981; Hess et al., 1985). This relationship may be more than fortuitous. Hess et al. (1985) have made estimates of U.S. cancer mortality based on average radon

Figure 7. Frequency of Rn-222 in North Carolina groundwater by lithology (data as in Fig. 6).



A = Coastal Plain; B = gneiss; C = granite; D = mafic
E = metasidimentary; F = metavolcanic; G - Precambrian sediments;
H = Triassic.

concentrations in water systems serving populations of different sizes. This approach presumes that radon concentration is systematically related to system size and that radon levels can be predicted by population. It can be argued that such a functional relationship should exist between water system size and radon concentration because radon is more likely to escape to the atmosphere during the aeration that typically accompanies water treatment in larger public water systems, or to decay before it is transmitted to users through extensive distribution systems. Because of the potential importance of water system size as a predictor of radon concentration, North Carolina radon data were also analyzed with respect to this variable to determine if system size is systematically related to radon concentration in the state.

Table 4 gives average radon concentrations for population classes similar those used by Hess et al (1985). These values in the table decrease with water system size. But, Table 5 shows mean population sizes of water systems by lithologic group. It is apparent from this table that mean system size varies among lithologic groups, and that systems in the coastal plain tend to be larger than those in other parts of North Carlina. Therefore, the apparent variation of radon concentration with system size in Table 4 is not independent of lithology. To examine the relationship of water system size and radon concentration independent of lithology, simple linear regressions were performed on data

TABLE 4

Table 4. Radon concentration and water system size.

INDIVIDUALS SERVED	n	Radon (pCi/l) ARITH. MEAN	GEOM. MEAN	MIN.	MAX.
< 100	36	3342.72	1652.43	30.39	35552.45
100-1000	102	3006.02	671.82	ND	46644.53
1000-5000	110	583.33	92.76	ND	5830.41
5000-10000	16	276.56	21.33	ND	1598.03
> 10000	3	42.13	39.3	26.9	1.58

within each lithologic group using the model:

$$\text{radon} = A + B(\text{population}).$$

Regression coefficients are given in Table 5. If there is a systematic negative effect of population on radon concentration independent of lithology the slope (b) should be negative in each group and radon should be negatively correlated with population. Among lithologic groups, however, the regression lines have both positive and negative slopes and the degree of correlation between radon and population is low (Table 5). This suggests that in North Carolina water system size does not affect radon concentration independently of lithology. The apparent inverse relationship between water system size and radon concentration in Table 4 is due to differential distribution of different-size water systems among lithologic groups. This is largely a result of the different hydrologic properties of these rock groups. Because of the low productivity of most Piedmont aquifers, large municipal water systems in that part of North Carolina must generally use surface water, and only relatively small public water supplies rely on groundwater. Larger systems in the coastal plain, on the other hand, typically do use groundwater. The distinctly different hydrologic characteristics of Piedmont and coastal plain regions thus have a large effect on the overall relationship of radon concentration and water system size in North Carolina. Essentially the same aquifers and similar patterns of groundwater use are present throughout the southeastern United States (Cederstrom et al., 1979) and

TABLE 5. Regression models for radon and water system size.

LITHOLOGY	n	SIZE* MEAN	S.D.	a	b	r	p
coastal plain	139	2659	3323	585.8	-0.06	-0.16	0.07
gneiss/schist	71	644	760	2467.4	-0.27	-0.1	0.42
granite	24	343	304	6543.67	8.52	0.25	0.25
mafic igneous	5	1503	1230	318.58	0.21	0.35	0.56
metasediments	4	495	507	1406.6	-1.15	-0.95	0.05
metavolcanics	21	1208	1422	1355.43	-0.01	-0.01	0.96
triassic	6	1056	920	723.66	0.18	0.24	0.65
TOTAL	272	1715	157	2305	-0.29	-0.18	0.003

Model: radon = A + (B)number of individuals

* size = number of individuals served by water system.
n = number of samples.

may similarly affect radon distribution throughout the region.

These results further emphasize the important influence of geologic and hydrologic variables on radon concentration, and strongly suggest that water system size is not an independent predictor of radon concentration.

Discussion

The radon concentration of groundwaters from the aquifers considered here is largely consistent with results in other areas and with known relative average uranium concentrations for each rock type. The association of very high radon concentrations with granites has been observed in other areas (e.g., Asikainen and Kahlos, 1979; Brutsaert, 1981; Snihs, 1973), but the high values from the metasedimentary rocks of Ocoee Supergroup (2 observations) may be surprising. Although the estimated average concentration in this group is unstable, high radon concentrations might reasonably be expected to occur in this aquifer because its mineralogic composition is similar to granite (Hadley, 1970). In addition, the Ocoee is metamorphosed to high rank (sillimanite zone) in some areas. Brutsaert et al. (1981) reported an arithmetic mean radon concentration of 13,630 pCi/l in sillimanite-grade aquifers in Maine. The relative radon concentrations of groundwaters from other minor aquifers also appear to be generally consistent with relationships between lithology and radon concentration observed elsewhere. The average radon concentration of groundwater from the Triassic basins (6 observations) reflects the overall lithologic similarity of the Triassic sediments to the Piedmont rocks from which they were derived. Additional samples would probably be required to characterize the full range and distribution of radon concentration in the Triassic rocks because they are quite lithologically diverse. The low radon concentration of mafic igneous rock aquifers (5 observations) is consistent with the known average uranium concentration of these rocks, which is orders of magnitude below that of granite.

Groundwaters from metamorphic rock aquifers in the Piedmont and Blue Ridge together with those from coastal plain aquifers account for most of the observations in the sample; average radon concentrations from these groups show that a distinct break exists between radon levels in the coastal plain versus the Piedmont and Blue Ridge regions. The gap in average radon concentration between coastal plain and Piedmont aquifers in North Carolina is similar in magnitude to the discontinuity reported in geometric mean radon concentrations of 158.5 and 2511.9 pCi/l, respectively, in the South Carolina coastal plain and Piedmont (King et al., 1982). In spite of their distinct differences from each other, the coastal plain, gneiss/schist, and metavolcanic lithologic groups are broad categories which include rocks with different chemical compositions and physical and hydrologic properties which may affect radon concentration. Sufficient geologic data could not be obtained to evaluate other geologic and hydrologic sources of variability in radon concentration within lithologic groups in the Piedmont and Blue Ridge because well records are poor in crystalline rock areas of North Carolina. It was possible, however, to conduct a more detailed analysis of the coastal plain sample.

The coastal plain group, with 139 observations, is the largest and perhaps the most geologically varied of the lithologic groups considered here. Several aquifers of different age and lithology are utilized in the coastal plain, and primary data on the aquifers being pumped were available for a few of the sampled wells. Average radon concentrations were determined separately for each aquifer for which data were available (Table 3). The differences between these average concentrations are geologically significant. The average radon content of the Castle Hayne aquifer which supplies water to a large portion of eastern North Carolina is quite low (Table 3); this average is similar to that observed by Mitsch et al. (1984) in the Castle Hayne. The higher concentrations in the Tuscaloosa aquifer may be a result of its compositional similarity to the Piedmont crystalline rocks it overlies (Stuckey and Conrad, 1958). Differences between the average radon concentrations of coastal plain aquifers are a potential source of variability within the combined coastal plain group. There is also a spatial pattern to variations in concentration.

Anomalously high radon concentrations up to 12,000 pCi/l occur along the fall line at the dissected western edge of the coastal plain. Several of the highest anomalous values occur in wells located where the western edge of the coastal plain overlaps Piedmont granites. The coastal plain sedimentary rocks are thin near the fall line and it is possible that some of the anomalous wells may have been drilled through the coastal plain sediments to granites beneath. High radon concentrations in coastal plain aquifers may also be due to sedimentological concentration of rock and mineral fragments eroded from Piedmont crystalline rocks in sedimentary units such as the Tuscaloosa which overlie crystalline rocks. Alternatively, Michel and Moore (1980) suggested that uranium from Piedmont crstalline rocks may be geochemically concentrated in the upper coastal plain. Anomalous concentrations of radium, the immediate parent of radon, have also been observed along the fall line in North Carolina (Menetrez and Watson, 1983) and South Carolina (Michel and Moore, 1980). Although the reasons for these radiological anomalies are not clear, it appears that an average radon concentration which excluded observations from along the fall line would be more representative of the region. Removing measurements from along the fall line, including those which were not anomalously high, reduces the average radon concentrations and the range of concentrations significantly as shown in Table 6.

Table 6. Rn-222 in Coastal Plain minus fall-line samples.

n	ARITH. MEAN	GEOM. MEAN	MAXIMUM	MINIMUM
127	148.72	72.2	2727.33	ND*

*Not detectable above background.

It should be emphasized that although these revised values may be more representative of the coastal plain as a whole, the high concentrations along the fall line are an important determinant of radon exposure in that area.

The importance of other variables, in addition to lithology, which affect radon concentration should also be considered although they can not be evaluated directly from the data used here. The observed relationship between lithology and radon concentration may be a function, in part, of other factors which are correlated with lithology. Perhaps the most important of these lithology-related variables are the large differences between the hydrologic characteristics of aquifers in the coastal plain and the rest of North Carolina. All of these factors need additional study. These differences in hydrologic characteristics are so large that they may overwhelm the effects of other variables and create correlations between apparently unrelated factors like lithology and water system size. Although some investigators have found relationships between, for example, well yield and radon concentration, an overall analysis of this relationship in North Carolina might be deceptive because, as is the case with water system size, it would reflect primarily the differences in average yields between coastal plain and crystalline rock aquifers. A more useful treatment of this relationship might examine the relationship between well yield and radon levels within lithologic or hydrologic units.

MANAGEMENT CONSIDERATIONS

The relationship of radon in groundwater to aquifer lithology is potentially useful in risk assessment and management. Average concentrations indicate that groundwater in the North Carolina coastal plain has radon concentrations which are significantly lower than the 180 pCi/l United States average reported by Hess et al (1985). The low radon concentrations of the coastal plain contrast with concentrations on the order of 1000 to 5000 pCi/l in large areas of the Piedmont and Appalachian portions of North Carolina which are underlain by gneisses, schists and metavolcanic rocks, and with smaller areas underlain by granite where radon concentrations may exceed 10,000 pCi/l. These radon levels are significantly above average for the U.S.

The large discrepancy between radon concentrations in the coastal plain and the remainder of the state dictates that any efforts to evaluate or mitigate the risk associated with radon in groundwater should be focussed in the Piedmont and mountains initially. Granite bedrock areas should be of particular concern because groundwater users in these areas are at the greatest risk from radon exposure. The impact of radon on private wells should also be given additional consideration. Data from other studies (Hess et al, 1985) indicate that radon concentrations tend to be higher in private wells than in public water supplies. Since none of the water samples considered in this study were from private wells, the applicability of predictive criteria for radon levels in private supplies needs to be assessed by further sampling. The relationship between lithology and radon concentration shows that geologic factors which control radon concentration, rather than factors like water system size, should be considered in efforts to estimate radon exposures or their associated risks.

A complete assessment of radon exposure requires, in addition to knowledge of the geologic and hydrologic factors which control radon concentrations in water, a determination of the number of individuals exposed to specific concentrations of radon. This number could be obtained by estimating the number of groundwater users with wells in a particular aquifer and then applying average radon concentrations from each aquifer to the appropriate number of individuals to produce a geologically-weighted estimate.

SUMMARY

In North Carolina the highest concentrations of radon in groundwater occur in areas underlain by granite, and the lowest concentrations are found in the coastal plain and in areas where bedrock consists of mafic igneous rocks. Anomalously high radon concentrations also occur immediately downdip of the fall line, but the reasons for these high values are not apparent. In general radon concentrations are less than 500 pCi/l in the coastal plain and between 1,000 and 5,000 pCi/l in the Piedmont and Blue Ridge regions. Typical radon concentrations in the coastal plain are below average for the United States, while concentrations in the Piedmont and Blue Ridge are above average. This distinct break in radon concentrations reflects the large differences in the lithologic and hydrologic characteristics between the coastal plain and the remainder of North Carolina. There is no overall systematic relationship between radon concentration and water system size in North Carolina.

Relationships between lithology and radon concentration in North Carolina groundwater are consistent with relative concentrations that might be expected from the relative average uranium concentration in common rocks. However lithology is related to such factors as grain size, weathering, and many hydrologic properties which also affect radon concentration. Therefore the observed relatiohship between lithology and radon may be due in part to these other factors which are correlated with lithology. Additional study is needed to evaluate the importance of these variables.

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