Radon-222 and Other Naturally-Occurring Radionuclides in Private Drinking Water Wells and Radon in Indoor Air in Buncombe, Henderson, and Transylvania Counties, North Carolina, 2005, by Ted R. Campbell

Ground Water Circular Number 20

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Ted R. Campbell

N.C. Department of Environment and Natural Resources

**Division of Water Quality, Aquifer Protection Section,
Asheville Regional Office**

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ABSTRACT

High levels of carcinogenic radionuclides – most notably radon-222 (radon) - naturally occur in ground water drinking supplies in the Blue Ridge and Piedmont Provinces of Western North Carolina. This is problematic because about half of the residents in the region rely on ground water as their principal potable supply. Further, eight counties in North Carolina - all in Western North Carolina – are classified as EPA Zone 1 counties, with predicted indoor air radon concentrations above the action level of 4 picocuries per liter (pCi/L). The main source of these radionuclides is uranium rich rock – including granites and gneisses – prevalent across much of the region.

Ground water samples collected from 103 private wells within Buncombe, Henderson, and Transylvania Counties were found to contain ubiquitously high levels of radon (109 to 45,600 pCi/L; median = 6060 pCi/L). Radon exceeded EPA's proposed maximum contaminant level (MCL) of 300 pCi/L in 98 percent of the wells, and exceeded the proposed alternate MCL of 4000 pCi/L in 64 percent of the wells. Uranium (maximum = 63 ug/L) and gross alpha activity (maximum = 56 pCi/L) exceeded the EPA MCL in about 2 to 3 percent of wells. Radium-226, -224, and -223 were relatively low in all sampled wells (less than 1.4 pCi/L).

Radon was significantly higher in wells located in meta-igneous rocks (median = 7480 pCi/L; maximum = 45,600 pCi/L) than in meta-sedimentary rocks (median = 4040 pCi/L; maximum = 14,300 pCi/L). One exception to this was the migmatitic biotite gneiss, a meta-sedimentary rock, which had median radon levels (8110 pCi/L) comparable to that of the meta-igneous gneisses. Although maximum levels of uranium and gross alpha were significantly higher in wells in meta-igneous rocks than meta-sedimentary rocks, little difference was noted between the two rock types for median levels. Radium-226 levels were comparable in the two rock types. Granite and granitic gneiss formations such as the Henderson Gneiss, granite gneiss, and Caesars Head Granite generally were associated with elevated radionuclides in well water.

Wells in oxidizing ground waters (n = 98) were significantly higher in radon and uranium (average = 8018 pCi/L and 1.8 ug/L, respectively) and lower in Ra-226 (average = 0.15 pCi/L) than wells in reducing ground waters (n = 5) (radon average = 1930 pCi/L; uranium average = near zero; Ra-226 average = 0.37 pCi/L). This corroborates findings of other researchers who have shown that uranium is more soluble in oxidizing ground water, while Ra-226 is less soluble. The plated Ra-226 serves as a continuing source of radon to the ground water.

Radon was positively correlated with the uranium/radium-226 ratio, suggesting that the ground water conditions which dissolve and mobilize uranium may in turn result in sorption of radium-226 on fracture walls acting as an ongoing source of direct radon emanation to ground water in the vicinity of the well. Further, radon was 3 to 5 orders of magnitude higher than radium-226, implying that the vast majority of radium-226 - the source of radon - was sorbed on bedrock fractures near the well. Because of the very large disequilibrium between radon and its parent radium-226, any changes in ground water geochemistry near the well could result in dissolution and mobilization of radium, and subsequent impact of the drinking supply.

Uranium and gross alpha activity were positively correlated with pH and alkalinity (as bicarbonate). Radium isotopes tended to increase as DO decreased. Radon was inversely correlated with Ra224/Ra223 (R=-0.45), suggesting that, as expected, rock with a presumed higher *uranium* content (parent of Ra-223 and radon) than *thorium* content (parent of Ra-224) resulted in higher dissolved radon concentrations.   Radium-223 was positively correlated with Ra-224 (R=0.76) and well depth (R=0.38) and inversely correlated with casing depth (R=-0.38), DO (R=-0.37) and ORP (R=-0.35). Radium-224 was positively correlated with well depth (R=0.37) and inversely correlated with DO (R=-0.44) and Ra-226/Ra-223 (R=-0.48). Wells were grouped into hydrologic/topographic settings of either ground water recharge, mid-slope, or ground water discharge, and differences in radionuclide concentrations between the three settings were not noted.

Indoor radon ranged from 0.3 to 22.8 pCi/L, with a median value of 2.7 pCi/L. About one third of the sampled homes had indoor air radon above the EPA action level of 4 pCi/L. Data obtained from a commercial laboratory for 15,280 homes in the three county study area showed indoor radon levels between 0.4 and 2146 pCi/L with a median value of 3.2 pCi/L.

INTRODUCTION

Elevated levels of naturally occurring carcinogenic radionuclides are found in ground water and indoor air (radon) in the Blue Ridge and Piedmont Provinces of Western North Carolina. This is due to the presence of uranium rich rocks – including granites and gneisses - across much of the region. Ductile and brittle faulting also is common in the area and may increase the mobility of radionuclides in ground water and air.

About half of the citizens of Western North Carolina rely on ground water as their principal drinking supply (U.S. Geological Survey, website http://nc.water.usgs.gov/wateruse/data/Data_Tables_2000.html, accessed May 22, 2006). Existing data from public and private wells in the region indicate that elevated concentrations of dissolved uranium, radium, and gross alpha-particle activity occur locally, and elevated concentrations of dissolved radon-222 (radon) are widespread. (In this report, "radon-222", "radon", and Rn-222" are used interchangeably.) Anecdotal

evidence suggests that few residents are aware that their domestic ground water supply may be contaminated with elevated levels of naturally occurring radon or other radionuclides.

Radionuclides are human carcinogens and have been linked to bone, kidney, and lung cancers, among others. To protect the public from these health hazards, public water suppliers are required to collect radionuclide samples at regular intervals as part of the U.S. Environmental Protection Agency's (EPA) "Radionuclides Rule" (EPA, 2000). This rule defines maximum contaminant levels (MCLs) for various radionuclides in public water supplies (table 1). Although the MCLs do not specifically apply to *private* (domestic) drinking water supplies, the standards often are used as guidelines in determining potential health risks associated with consuming drinking water from domestic wells.

To date, studies of radionuclides in ground water in Western North Carolina have been limited in scope. Further, the correlation between radionuclide content in parent rock and radionuclide concentrations in ground water and air is not well understood. A study conducted from 1991 to 1993 found that the average radon concentration in 277 private wells sampled in the mountain region was about 2000 picocuries per liter (pCi/L) (University of North Carolina, 1993); a study in 1975 found that the average radon concentration in 20 wells sampled in the mountain region was about 4100 pCi/L (Aldrich and others, 1975).

Homes, schools, and businesses in Western North Carolina also are susceptible to elevated levels of indoor air radon. Eight counties in North Carolina - all in Western North Carolina – are classified as EPA Zone 1 counties, with predicted indoor radon concentrations above the EPA recommended action level of 4 pCi/L (EPA Radon Map, accessed via internet, 8/19/05, http://www.ncradon.org/zone.htm).

Because radionuclides are known to occur in the region and because they are linked to increased risk of cancer, several key questions must be addressed. What is the occurrence and distribution of dissolved radionuclides in the region? Are the observed levels safe to drink? Are the dissolved radon levels high enough to cause a substantial increase in the overall exposure to inhaled radon? Is it possible to develop regional radionuclide susceptibility maps on the basis of knowledge of local geology, geochemical conditions, and topographic settings? Are well owners aware of the implications of elevated levels of dissolved radionuclides in their drinking water? Is current policy regarding radionuclides in drinking water adequately protective of public health?

This study was designed to evaluate the extent to which rock types, geologic structure, topographic setting, and other hydrogeochemical factors are associated with elevated radionuclide concentrations in air and ground water. The study was targeted to specific areas of Western North Carolina and therefore is limited in scope. It is part of a multi-phased approach to help officials and the public to understand the quality of the ground water supply and the extent to which radionuclides may pose a health threat to the citizens of Western North Carolina. Findings in this report will help the planning,

regulating, scientific, and well owner communities to more fully understand the mechanisms and settings that contribute elevated radionuclides to the region's ground water supplies and indoor air. This study is a direct response to the North Carolina Division of Water Quality's mandate to help ensure that North Carolina's ground water resources are safe and sustainable. This study was made possible by a grant from the EPA, and carried out in consultation with the North Carolina Division of Environmental Health's Radiation Protection Section.

Purpose and Scope

The purpose of this report is threefold. First, the report documents the occurrence and distribution of selected radionuclides in drinking water collected from private wells in targeted areas of Buncombe, Henderson, and Transylvania Counties of Western North Carolina. Second, the report describes the extent to which elevated radionuclide concentrations are associated with local geology (rock type, structure, depth to bedrock), geochemistry (dissolved oxygen (DO), pH, specific conductance (SC), alkalinity as bicarbonate (alk), iron, manganese, and selected individual radionuclides), and (or) well characteristics (topographic setting, and others). And third, the report provides background information on the occurrence and behavior of radionuclides common in Western North Carolina, and touches on associated health risks, and therefore serves as a useful reference for planners and citizens of the region.

Data used to draw conclusions in this report were obtained from raw, untreated, unfiltered ground water samples collected using a consistent method at 103 private drinking water wells and one abandoned public supply well. All wells sampled in the study were open boreholes completed in fractured bedrock with the overlying soil and regolith sealed by polyvinyl chloride or steel casing. The wells were sampled for total uranium (uranium), gross alpha-particle activity, radium isotopes (radium-226 (Ra-226), radium-224 (Ra-224), and radium-223 (Ra-223)), radon-222, iron, manganese, lead, arsenic, and field parameters. Additional data obtained at the wells included well-construction details, yield, latitude and longitude, topographic setting, and surrounding rock type and structure information.

In addition, indoor air radon was measured in 67 of the 103 homes associated with the sampled private wells. To augment the indoor radon dataset, AirChek, Inc. provided an additional 15,280 indoor air radon results for homes in the three-county study area. Finally, historic compliance monitoring data obtained from public water suppliers across Western North Carolina also were evaluated.

Areas sampled during this study were selected because of their proximity to uranium-rich geologic formations and because of their location within a county that has been identified as having predicted indoor radon screening potential greater than the EPA action level of 4 pCi/L (EPA Radon Map, accessed via internet, 8/19/05, http://www.ncradon.org/zone.htm). EPA Radon Zone Map, accessed via internet, 8/18/05, http://www.ncradon.org/zone.htm). A number of other areas of Western North

Carolina also are suspected to contain potentially elevated radionuclide concentrations (the area of Whiteside Granite in Macon County, for example), and these areas may be addressed in subsequent phases of investigation.

Acknowledgments

Background on Radionuclide Chemistry and Occurrence

Radionuclides are naturally occurring elements that undergo radioactive decay. This decay occurs when an unstable "parent" element releases energy and becomes a new "daughter" element (progeny) with new properties. Energy released during this decay process occurs as ionizing particles (alpha or beta particles) or rays (gamma rays). Naturally occurring radionuclides are found primarily found to originate from one of three decay chains: uranium-238, uranium-235 (rare), and thorium-232 (common, but mostly insoluble). An example of the decay process is that of uranium-238, which decays through a series of intermediate daughter elements (with each decay step releasing either alpha or beta particles) and ends as a stable form of lead.

Radiation is a measure of the *rate* of radioactive decay, which is also referred to as *activity*. Activity is expressed as the "curie", a measure of the number of disintegrations per unit time (one curie = 3.7 x 10^{10} atomic disintegrations per second). Activity in water is expressed in pCi/L, where one pCi/L is equivalent to 2.2 atomic disintegrations per minute per liter of water. A very large mass of a slowly decaying radionuclide can have a lower overall radiation than a very small mass of rapidly decaying radionuclide. Each type of radiation has unique properties, but, generally speaking, most are energetic enough to break chemical bonds and potentially result in tissue damage.

Radionuclides are colorless, odorless, and tasteless. They are ubiquitous in rock, soil, and water. Their concentration and occurrence in ground water are controlled, in large part, by geochemical conditions and by the degree to which the local geology contains original sources of all radionuclides, namely uranium or thorium. Radionuclides include the very slow decaying elements that originally were incorporated into the earth's crust (uranium and thorium), as well as their shorter-lived progeny. Every radioactive element has a unique "half-life" which is a measure of the amount of time required for half of the initial amount of the substance to decay. Half-lives vary widely, as shown in figure 1 which illustrates the two most common decay series, uranium-238 and thorium-232, as presented from Hall and others (1985).

Because radioactive elements have different radiochemical properties, they tend to behave differently in the subsurface. Among the properties that vary are solubilities, decay rates, sorption rates, physical states (gas or solid), and geochemical reactivity. As a result, observed concentrations of a parent element are not necessarily correlated with observed concentrations of its daughter elements. For example, high concentrations of dissolved radon are not necessarily correlated with concentrations of dissolved Ra-226. Further, because the geochemical properties of uranium-238 differ from those of Ra-226, they are mobilized under different conditions and their co-occurrence is not common (Szabo and Zapecza, 1991; Menetrez and Watson, 1983).

Adverse health affects are associated with long-term exposure of radionuclides, so the EPA has established drinking water standards for selected constituents (table 1). In 1991 the EPA proposed an MCL standard for radon in water of 300 pCi/L, and in 1999 proposed an alternate MCL of 4000 pCi/L for water suppliers that have established an indoor radon mitigation program. The primary radon health risk is from inhalation exposure rather than ingestion, and radon-rich water can slightly increase levels of indoor air radon as it volatilizes during routine in-home water usage. As of the date of this report, these proposed levels have not yet been enacted.

Studies have shown that radionuclide concentrations in ground water and indoor air in Western North Carolina are above EPA MCLs and are therefore high enough to pose potential health risks. Anthropogenic sources of radionuclides (from nuclear testing, radiopharmaceuticals, and mining) are not a significant source of contamination to ground water supplies in the region.

The radionuclides of interest in this study include uranium, Ra-226, radon, and gross alpha-particle activity. Radium-228 is a common constituent that was not included in this study due to cost and laboratory constraints. Thorium-232 is another common radionuclide with its own decay series, but was not analyzed directly during this study. Instead, Ra-224, a decay product of thorium-232, was used as a proxy to identify potential thorium sources. Thorium is relatively insoluble in most ground waters.
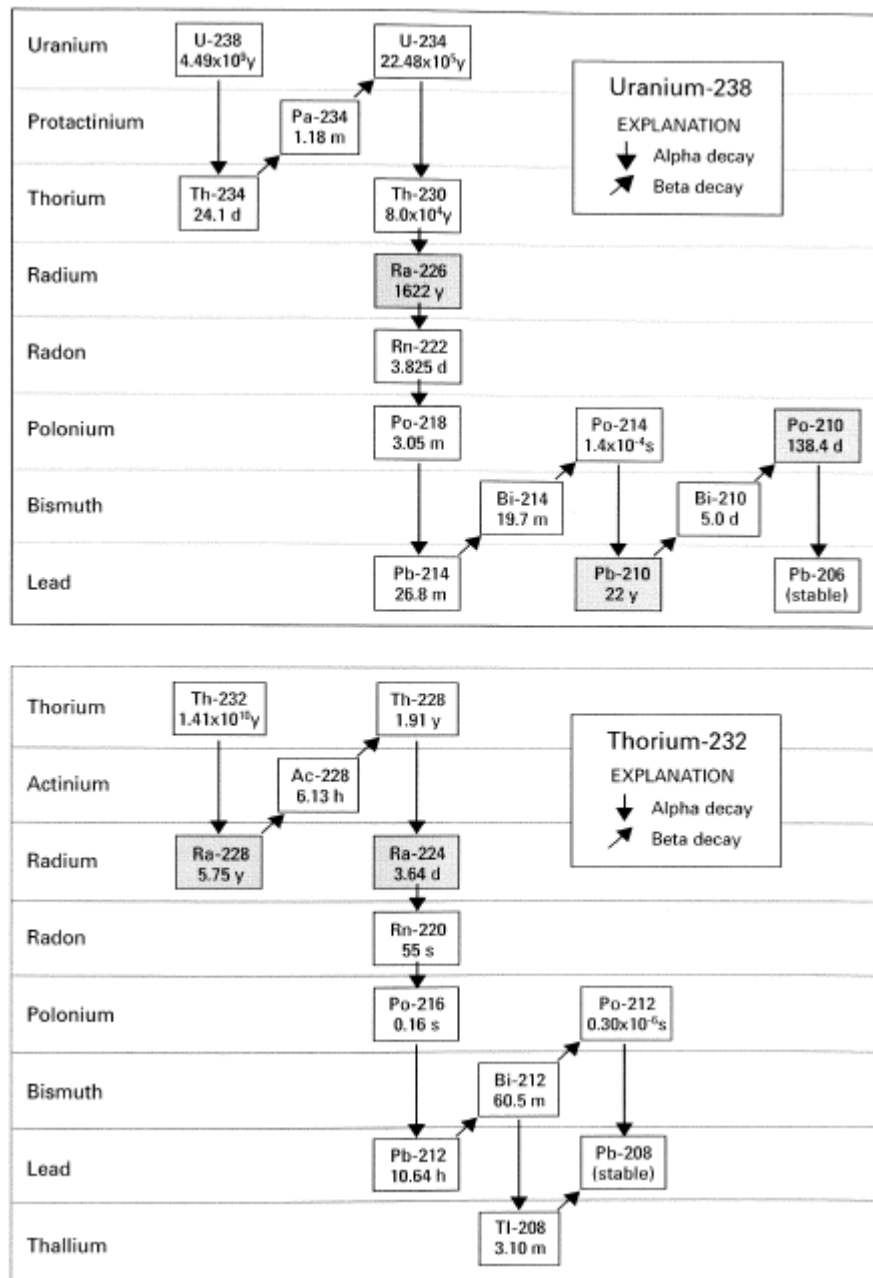
Figure 1. Schematic showing radionuclide decay chains with parent and daughter elements and half-lives (in seconds (s), minutes (m), hours (h), days (d), or years (y), from Focazio and others (2000), as adapted from Hall and others (1985).

Table 1. EPA maximum contaminant levels for selected radionuclides.

Radionuclide	USEPA MCL
Radon, in pCi/L	300* / 4000**
Uranium, in ug/L	30
Radium-226, in pCi/L	5***
Radium-228, in pCi/L	5***
Gross Alpha, in pCi/L	15****
Gross Beta, in millirems/yr	4
Indoor air radon, in pCi/L	4*****

*   proposed MCL
**   proposed alternate MCL
***   combined value, radium-228 + radium-226
****   excluding uranium and radium-226
***** air standard is an "action level"
MCL, maximum contaminant level
pCi/L, picocuries per liter
ug/L, micrograms per liter

Radium-223, a daughter of uranium-235, also was analyzed. Descriptions are provided below for the environmental occurrence of selected radionuclides.

Gross alpha-particle activity

Gross alpha-particle activity is a measure of the total amount of radioactivity in a sample that is attributable to the decay of alpha-emitting radionuclides (refer to fig. 1). Both short-lived and long-lived radionuclides emit alpha particles, therefore the length of time between sample collection and laboratory analysis can greatly affect the results that are obtained. Gross alpha activity often is used as a screening tool to predict the presence of other regulated radionuclides such as Ra-226 and uranium, or isotopes that are not yet regulated but pose concern because of widespread occurrence, such as Ra-224 (Focazio and others, 2000). Exposure to elevated levels of alpha emitters is linked to an increased likelihood of incidence of cancer. The EPA has established an MCL of 15 pCi/L for gross alpha activity, excluding uranium and Ra-226, which are regulated separately.

Uranium

Uranium is a naturally occurring radionuclide that is common in crustal rocks around the world. It is a reactive metal and combines with other elements in the environment to form various uranium compounds. It is the parent (source) of a long series of intermediary progeny, including Ra-226, radon, and others. Because it has an extremely long half-life of 4.5 billion years, it is present in quantities similar to those that existed when the earth was formed. Long-term exposure to uranium has been linked to

increased incidence of liver and kidney toxicity and cancer. The EPA has established an MCL of 30 ug/L for uranium.

Natural uranium occurs as three isotopes that are found in the following predictable percentages, by weight: 99.28% uranium-238; 0.72% uranium-235; and 0.006% uranium-234 (International Atomic Energy Agency, website http://www.iaea.org/NewsCenter/Features/DU/du_qaa.shtml, accessed May 23, 2006). Uranium-238 and uranium-234 are part of the same decay chain (that is, uranium-238 is a parent of uranium-234) and are much more common in terms of mass and radioactivity, respectively, than uranium-235, which is part of the highly radioactive and rare actinium decay chain. Because these isotopes and their daughter progeny occur in predictable percentages when in radioactive equilibrium, a comparison of ratios can reveal the predominant original source of radionuclides which can, in turn, lead to a greater understanding about the geologic influence on radionuclide occurrence in ground water. For example, in equilibrium daughter progeny occur in the following relative ratios: Ra-223 (uranium decay series) 0.05 : Ra-226 (uranium decay series) 1.0 : Ra-228 (thorium decay series) 1.5 : Ra-224 (thorium decay series) 1.5.   Measuring these daughter elements can reveal whether thorium or uranium is the primary geologic influence on radionuclide occurrence.

Szabo and others (2001) found elevated concentrations of uranium in a large number of wells across the Appalachian Physiographic Province. These elevated levels were associated with ground water of moderate DO and neutral to alkaline pH. Concentrations of dissolved uranium were as high as 100 ug/L at some locations and tended to be highest in private wells (Landmeyer and Rueber, 2001).

Uranium is particularly soluble in the presence of oxygenated ground water, though it can, in some rare cases, be soluble in reducing environments. Zapecza and Szabo (1988) observed that uranium is particularly soluble in oxidizing, bicarbonate-rich ground water. Uranium concentrations also are associated with oxidizing waters low in iron and manganese and high in sulfate and bicarbonate. Uranium tends to precipitate out of solution and sorb onto fracture openings under reducing conditions, as iron- and sulfate-reducing bacteria reduce the soluble form of uranium (VI) to the insoluble form (IV) (Sani and others, 2004). The presence of carbonates (from the geologic matrix or bacterial respiration) and neutral or alkaline pH will tend to dissolve uranium into very stable uranium (VI)-carbonate complexes (Hsi and Langmuir, 1985) . That is, the dissolution of uranium is limited in many geochemical environments and is controlled in large part by adsorption and complexation reactions. In short, dissolved uranium generally is associated with somewhat increased levels of DO, pH, alkalinity, and sulfate and bicarbonate, and generally is associated with somewhat decreased levels of iron and manganese.

Radium, a naturally occurring reactive metal linked to bone cancer, occurs in the form of four primary isotopes that are produced during the decay of uranium (Ra-226 and Ra–223) and thorium (Ra-228 and Ra–224) (fig 1). This study is concerned with Ra-226, Ra-224, and Ra-223. Radium-223 typically occurs at very low concentrations in ground water because it originates from uranium-235, the least abundant of the long-lived parent nuclides. Nevertheless, Ra-223 was measured to help identify parent source rock. Due to budget constraints, Ra-228 was not measured. The EPA has established an MCL of 5 pCi/L for combined Ra-226 plus Ra-228.

Radium-226 is a daughter of uranium-238 and has a half-life of 1622 years (fig. 1). Radium-224 is a daughter of thorium-232 and has a half-life of 3.6 days. Radium-223 has a half-life of 11.4 days and is a daughter of uranium-235. Radium-228 is a daughter of thorium-232 and has a half-life of 5.75 years.

Radium concentrations in ground water are dependent on concentrations of parent elements, geochemical conditions that control dissolution and sorption, travel time versus half-life decay, and the occurrence of alpha recoil (Szabo and Zapecza, 1991). Radium forms as parent elements decay, and parent concentrations depend on rock type and solubility. Geochemical conditions also are critical in determining whether radium is in solution or sorbed onto soil/rock. Sorption depends in part on the ionic strength of the solution. As ion concentrations in ground water increase, there is greater competition for sorption sites and solubility of radium tends to increase. Sorption also depends on the particle surface area, with greater surface area (manganese oxides, for example) leading to increased sorption. Dissolved concentrations also are controlled by half-lives, with the longer lived isotopes traveling "unsupported" (in the absence of the parent element) further than short lived ones. Finally, alpha recoil can account for varying radium concentrations dissolved in ground water. When a parent element decays to the daughter radium isotope, it emits an alpha particle (a positively-charged helium atom). By virtue of the conservation of momentum, the newly created radium atom will "recoil" in the opposite direction. If the decay occurs close enough to the surface of the solid phase (soil, rock, fracture plating) surface, then the recoil action can propel the atom out of the mineral surface and into solution (Fleischer, 1983).

Radium tends to be only slightly soluble, and concentrations have been associated with reducing waters high in iron and manganese and low in sulfate (Szabo and Zapecza, 1991). The solubility of radium generally is low, but increases with decreasing pH (Szabo and others, 2005). Thus, solubility is controlled mostly by sorption conditions that are controlled largely by pH. Increased concentrations of total dissolved solids also increase the solubility of radium as other ions compete for limited sorption sites, a phenomenon known as the "common ion effect".

Radium-226 is a daughter produced by the decay of uranium-238. Because uranium-238 is distributed widely and can readily dissolve and move in ground water, its daughter elements such as Ra-226 and radon tend to be common in ground water.

Because Ra-226 has different geochemical properties (sorption, solubility, and complexation) than its parent uranium-238, their co-occurrence is not common. Similarly, Ra-226 concentrations often are not correlative with radon concentrations (Szabo and Zapecza, 1991).

In Western North Carolina, 128 public water supplies had an average Ra-226 concentration of 0.5 pCi/L (written communication, M. Fillinger, North Carolina Department of Environmental Health, Public Water Supply, Jan 12, 2005). Radium-226 has been found in elevated concentrations in ground water along the Fall Line in states from Georgia to New Jersey (Zapecza and Szabo, 1988). These aquifers tend to be composed, in part, of unconsolidated sand fragments that are rich in uranium minerals derived from the Blue Ridge and Piedmont provinces. In a study by Szabo and others (2001), elevated concentrations of radium were associated with reducing or acidic conditions and with quartz, sandstone, and shales.

Radium-224 is a daughter produced by the decay of thorium-228 (thorium-228 is a decay element of thorium-232). Although the abundance of thorium is higher than that of uranium, the solubility of thorium is very limited, and the occurrence of Ra-224 generally is less widespread than that of Ra-226 (Focazio and others, 2000). The short half-life of Ra-224 (3.64 days) also limits its occurrence in ground water. While some studies have shown that Ra-224 can be two times higher than Ra-228, their concentrations were nearly the same (1:1 ratio) in a study of 99 wells across the US (Focazio and others, 2000). This study also found radium to be somewhat mobile in the presence of ground water with low DO, high total dissolved solids, and high chloride content. Due to the common ion effect, high radium also was found in disturbed agricultural recharge areas where competing ions such as H, Ca, and Mg were present (Szabo and others, 1997).

Radium-223 is a daughter of the rare uranium-235 (part of the actinium decay series). Historic Ra-223 data from ground water samples collected in Western North Carolina could not be located, though Focazio and others (2000) measured concentrations less than 1 pCi/L in a limited national reconnaissance.

In 1983, Menetrez and Watson (1983) examined radionuclide data from North Carolina public water supply systems and found that about 2.5% of the 2089 samples were above regulatory standards for either combined Ra-226 plus Ra-228 (39 were above the MCL of 5 pCi/L) or gross alpha activity (12 were above the MCL of 15 pCi/L), and over half the systems with elevated gross alpha activity (above 15 pCi/L) also had elevated uranium concentrations (for example, above 10 pCi/L). Results of well sampling in the Piedmont of S.C. indicated that elevated Ra-226 concentrations were not strongly associated with elevated uranium-238 concentrations (South Carolina Department of Health and Environmental Control, website http://www.scdhec.net/eqc/water/html/urotest.html, accessed January 12, 2004).

Radium isotope ratios may be used to evaluate the sources of radium in ground water. For example, the ratio of Ra-224/Ra-223 is used to determine whether thorium

(the parent element of Ra-224) or uranium (the parent element of Ra-223) is predominant. Radium isotope ratios also may be used to evaluate the different processes that affect the emanation/leaching of radium from the rock into the ground water. Because the ratio of uranium-238 to uranium-235 is constant in nature, the ratio of daughter elements is likewise expected to be constant. Therefore, the ratio of Ra-223 to Ra-226 should be constant under equilibrium conditions and should equal 0.046. If the ratio is greater than the expected 0.046 then the shorter-lived isotope Ra-223 predominates suggesting that recoil processes are more important than dissolution. However, if the ratio is less than 0.046 then the longer lived isotope Ra-226 predominates suggesting that dissolution processes are more important than recoil.

A typical granitic rock, such as those common in Western North Carolina, contains 3.6 disintegrations per minute per gram (dpm/g) of uranium-238, 0.17 dpm/g of uranium-235, and 5.3 dpm/g of thorium-232 (International Atomic Energy Agency, website International Atomic Energy Agency, website http://www.iaea.org/NewsCenter/Features/DU/du_qaa.shtml, accessed May 23, 2006). Therefore, at equilibrium, the rock will contain 3.6 dpm/g of Ra-226 (a daughter of uranium-238), 0.17 dpm/g of Ra-223 (a daughter of uranium-235) and 5.3 dpm/g of Ra-224 and Ra-228 (daughters of thorium-232). At equilibrium, the ratios would be expected to be 1.0:0.05:1.5:1.5 for Ra-226:Ra-223:Ra-224:Ra-228. If the ratios are substantially different, the system is in non-equilibrium and it may be inferred that preferential transport is occurring for one or more radium or thorium isotopes.

Radon-222

Radon-222 (radon) is a naturally occurring radioactive gas that is invisible, mobile, and dissolves in water. It is produced during the decay of uranium (fig. 1). Uranium-238 decays through several daughter elements to Ra-226, and Ra-226 decays directly to radon. Radon is found nearly everywhere in soil, air, and water; even outdoor air contains very low levels of radon (typically about 0.4 pCi/L).

Radon is a known human lung carcinogen and is the second leading cause of lung cancer in the US (National Academy of Sciences, 1999). It is estimated that radon is responsible for 15,000 to 20,000 lung cancer deaths per year in the U.S. (National Academy of Sciences, 1999). It is the largest source of radiation exposure to the public and is considered a serious health risk.

By far, the primary health risk for radon is exposure through inhalation, not ingestion. Drinking radon-rich water appears to result in only a very minor increase in the risk of stomach cancer (National Research Council, 1999). The primary concern of exposure to radon-rich water is its contribution to radon in indoor air as it volatilizes out of the water phase. This occurs during normal household water use, particularly during showers and during the washing of dishes and clothes (Cothern and others, 1986; Fitzgerald and others, 1997; Prichard, 1987). Some research has shown that acute doses of de-gassed radon (during showering, for example) may spike at levels that could

increase the overall long-term exposure (and therefore health risk) over that of ambient indoor radon levels alone (Fitzgerald and others, 1997; Bernhardt and Hess, 1996).

Most radon that is inhaled is also exhaled, but radon's daughter progeny are readily retained in the lung. These daughter progeny, over the long term, can damage sensitive lung tissue. Even very small exposures to radon can, over time, result in lung cancer, and researchers have not yet been able to determine a safe lower threshold. Smokers exposed to elevated levels of radon are particularly susceptible to contracting lung cancer because of the synergistic relationship between radon, smoking, and lung cancer. Generally speaking, health risks associated with radon are due to long-term exposure, from about 5 to 25 years (National Academy of Sciences, 1999).

The EPA has proposed an MCL for radon in water of 300 pCi/L, and an alternate MCL of 4000 pCi/L for public water suppliers that have radon mitigation programs for their customer base (EPA, 1999). Radon dissolved in public water supply systems nationwide averages about 250 pCi/L (National Research Council, 1999).

The source of radon gas is uranium-rich rock. Rock types that contain higher than normal amounts of uranium are granites, felsic volcanic rocks, black shales, phosphate-rich sedimentary rocks, and metamorphic rocks derived from these rocks (U.S. Geological Survey, 2000). Western North Carolina is underlain by many of these, most notably granitic and metamorphic rocks. Along these lines, rock type has been strongly associated with concentrations of dissolved Radon, with ground water in granites often containing high levels, up to 100,000 pCi/L (Asikainen and Kahlos, 1979; Brutsaert and others, 1981; Snihs, 1973) and ground water in sedimentary rocks often containing much lower levels, often less than 500 pCi/L (Andrews and Wood, 1972; King and others, 1982; and Mitsch and others, 1984).

Because of its relatively short 3.8-day half-life, radon typically is not transported long distances from its source. Because radon cannot move long distances without decaying, and because radon often is found in the absence of dissolved uranium and radium, it may be concluded that radon concentrations often are derived from fracture coatings that contain uranium and radium precipitate. In general, elevated radon concentrations generally are associated with fractured, uranium-rich bedrock and thin or highly permeable, well drained, overlying soils. There tends to be little relationship between dissolved concentrations of radon and Ra-226.

Radon is mostly chemically inert (non-reactive) and is soluble in water. Because it is a gas, it is much more mobile than other radionuclides such as radium and uranium. Radon migrates from areas of higher pressure to those of lower pressure. For example, radon tends to move from underlying rock and soil into buildings (through foundation cracks and utility openings), particularly into basements. This effect is pronounced in buildings situated on cut-slope embankments (LeGrande, 1987).

Radon also is drawn into the low pressure zone within a pumping well's cone of depression. In the Piedmont and Mountains of North Carolina the cone of depression

often is overlain by a semi-confining clayey saprolite. As the well pump cycles off and on, the water table fluctuates up and down creating turbulent conditions, particularly pronounced at the zone between unsaturated and saturated conditions. The result is that radon is drawn from remote crevices and fractures into the low-pressure zone and pump (LeGrande, 1987). In the absence of pumping, radon in the unsaturated zone tends to be pushed downward with the infiltration of rainfall and then upward as the water table rises after being recharged by a rainfall event. Because radon moves more quickly in air than water its mobility may tend to increase as the soil dries, resulting in increased indoor concentrations. An increase in soil moisture content after a precipitation event also effects soil radon emanation. For buildings whose water is supplied by a well, radon dissolved in ground water can enter through the water distribution system, becoming volatilized into the indoor air space as the water is used, especially showers and washing machines (Fitzgerald and others, 1997).

Dissolved radon tends to be higher in private wells than in public systems (Hess and others, 1985). Private wells are small, closed systems that can result in radon build up, while public systems generally have a longer residence storage time with longer distribution systems which allows for some decay before consumption.

Radon concentrations also can be affected by the timing of recent rainfall, volume purged from a well before sampling, volume purged from a well in recent days prior to sampling, depth of water table, and other factors. Radon levels tend to be less influenced by geochemical (dissolution) conditions than radium and uranium because radon is relatively inert, whereas dissolved radium and uranium are observed only when they are solubilized from the solid phase into water. Dissolved radon levels may increase somewhat when the water table is higher (when the soil is saturated) because alpha recoil in dry grains can result in radon particles becoming embedded in adjacent soil grains rather than becoming freed into the pore-water space between grains.

Studies show that radon concentrations in ground water vary widely across North Carolina, from detection limits to over 45,600 pCi/L (Campbell, 2006; Aldrich and others, 1975; Sasser and Watson, 1978; Loomis and others, 1987; Loomis, 1987a; Horton, 1983; 1985). Loomis (1987b) found that regional variability in dissolved radon concentrations was mostly consistent with relative abundances of uranium-bearing rocks. Additional research was recommended to evaluate the variabilities found within given rock types. Loomis and others (1987) also reported a high degree of variability between radon concentrations within any given rock type. For example, wells in granites had the highest *and* the lowest dissolved radon concentrations. Rock type and geologic region, taken in combination, were the best predictors of elevated dissolved radon concentrations. Highest radon concentrations were found in settings characterized by granites and gneisses within the Blue Ridge, Inner Piedmont, and Raleigh Belt regions. Lesser concentrations were found in mafics and metavolcanic settings. In their study pumping well factors such as discharge, specific capacity, depth, and casing length were *not* significant predictors of radon concentrations. In a study by Senior (1998), radon activities did not correlate with well characteristics such as depth or yield, nor with pH, dissolved major ions, or other chemical constituents in water.

Radon concentrations in ground water in North Carolina have been among the highest observed in the U.S. (Hess and others, 1985; Horton, 1983; 1985; Aldrich and others, 1975; Mitsch and others, 1984; Strain and others, 1979). A 1993 study reported that of 400 private wells sampled across North Carolina, 67% had radon concentrations above 300 pCi/L, and 11% were above 4000 pCi/L (University of North Carolina, 1993). Of the 277 wells sampled within the mountain region, the average radon--222 was about 2000 pCi/L, 83% were above 300 pCi/L, 56% were above 1000 pCi/L, and 10% were above 5000 pCi/L. In general, the highest concentrations of radon in ground water across the U.S. occur in New England, Southeastern Appalachian and Piedmont provinces, Rocky Mountain States, and California (Hess and others, 1985; Horton, 1985). Hess and others (1985) noted that dissolved radon concentrations in surface water typically are very low due to volatilization of radon into the atmosphere. As a result, communities that rely on surface water reservoirs for their drinking water supply typically have little cause for radon concern.

Another isotope of radon known as thoron (or "radon-220") is a very short-lived daughter of thorium. Because of its short half life (54.5 seconds) it is very difficult to measure. Its short half life also prevents it from traveling more than very short distances (perhaps several feet) from its source. For this reason, exposure to thoron typically is limited. Thoron was not addressed in this study, and all references to "radon" in this report are to radon-222.

Indoor air radon

Ambient air radon concentrations range from less than 0.1 pCi/L to about 30 pCi/L (U.S. Geological Survey, 2000). The amount of radon in a given sample of air is dependent on many factors, including the proximity of source material, soil porosity, moisture, and interconnectivity (permeability), depth to ground water, barriers to upward movement such as clay confining layers or well-sealed slab foundations, interior ventilation, and others. The EPA has established an action level of 4 pCi/L for indoor radon.

Eight counties in North Carolina are listed as Zone 1, counties that are projected to have indoor radon levels greater than 4 pCi/L on average. These include Watauga, Alleghany, Mitchell, Buncombe, Henderson, Transylvania, Cherokee, and Rockingham Counties (fig. 2). According to a statewide statistical survey of indoor air in homes (North Carolina Radiation Protection Division, 1990), average radon concentrations for 94 samples in Buncombe County were 2.18 pCi/L, for 8 samples in Cherokee were 3.44, 45 samples in Henderson were 4.46, 5 samples in Mitchell were 1.76, and for 17 samples in Transylvania were 4.44. Concentrations were somewhat higher in a statewide non-statistical data compilation study. It is suggested that an estimated 10,000 pCi/L of Radon dissolved in ground water supplied to a home results in about 1 pCi/L of Radon in indoor air (Prichard, 1987). Indoor radon concentrations obtained from a commercial laboratory ranged from 0.4 to 1082 pCi/L (written communication, S. Price, Airchek, Inc., March 8, 2005) (fig. 3).
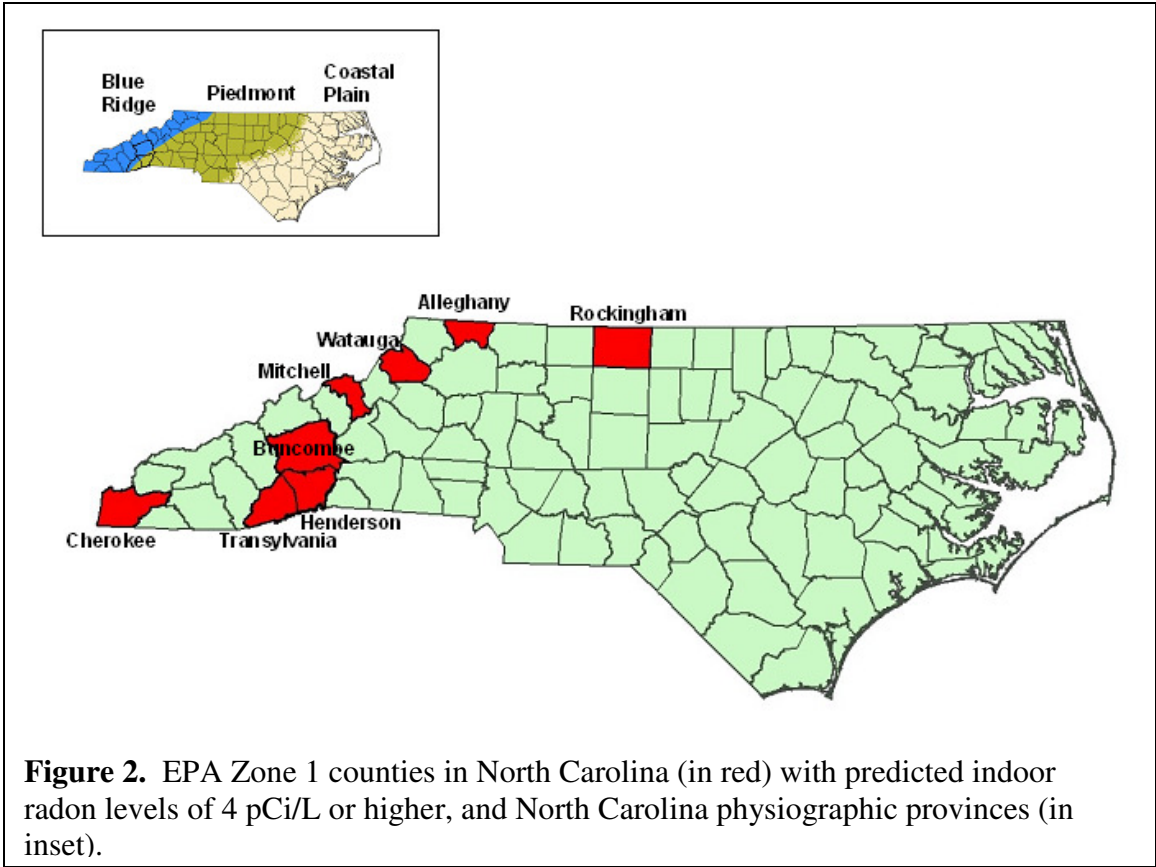
Figure 2. EPA Zone 1 counties in North Carolina (in red) with predicted indoor radon levels of 4 pCi/L or higher, and North Carolina physiographic provinces (in inset).
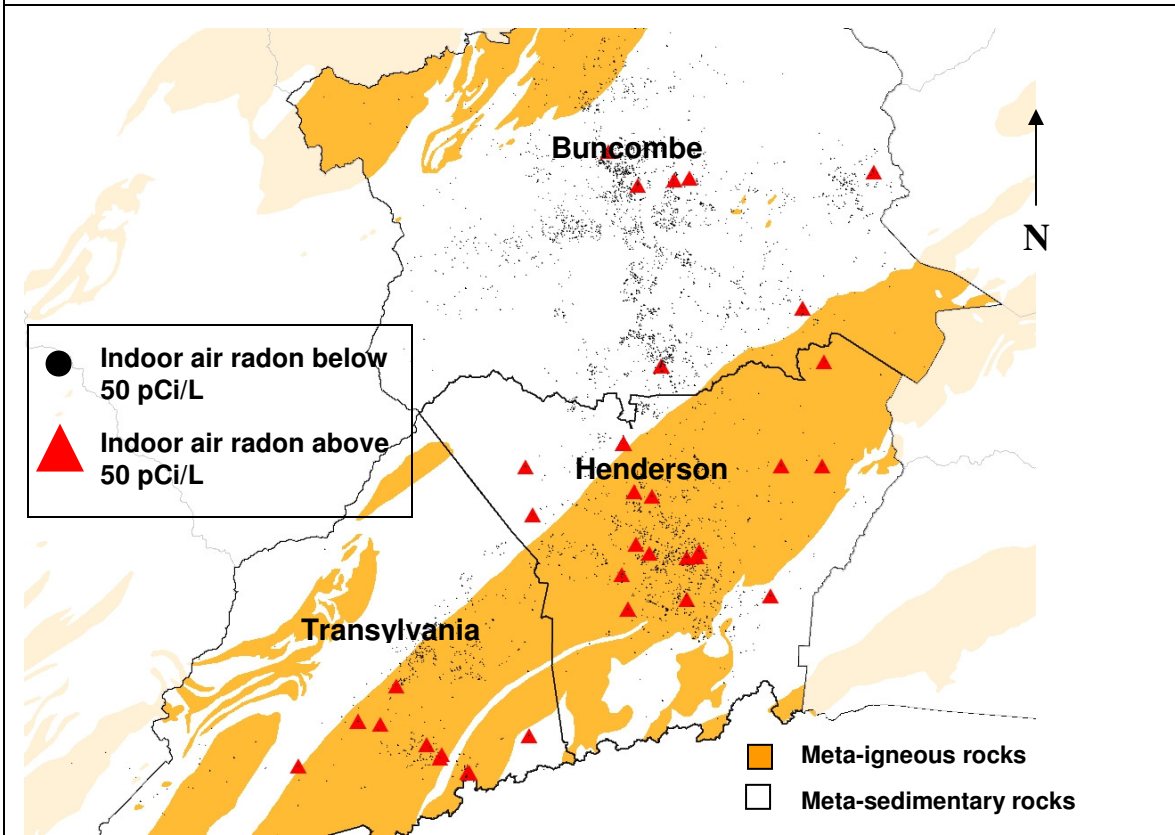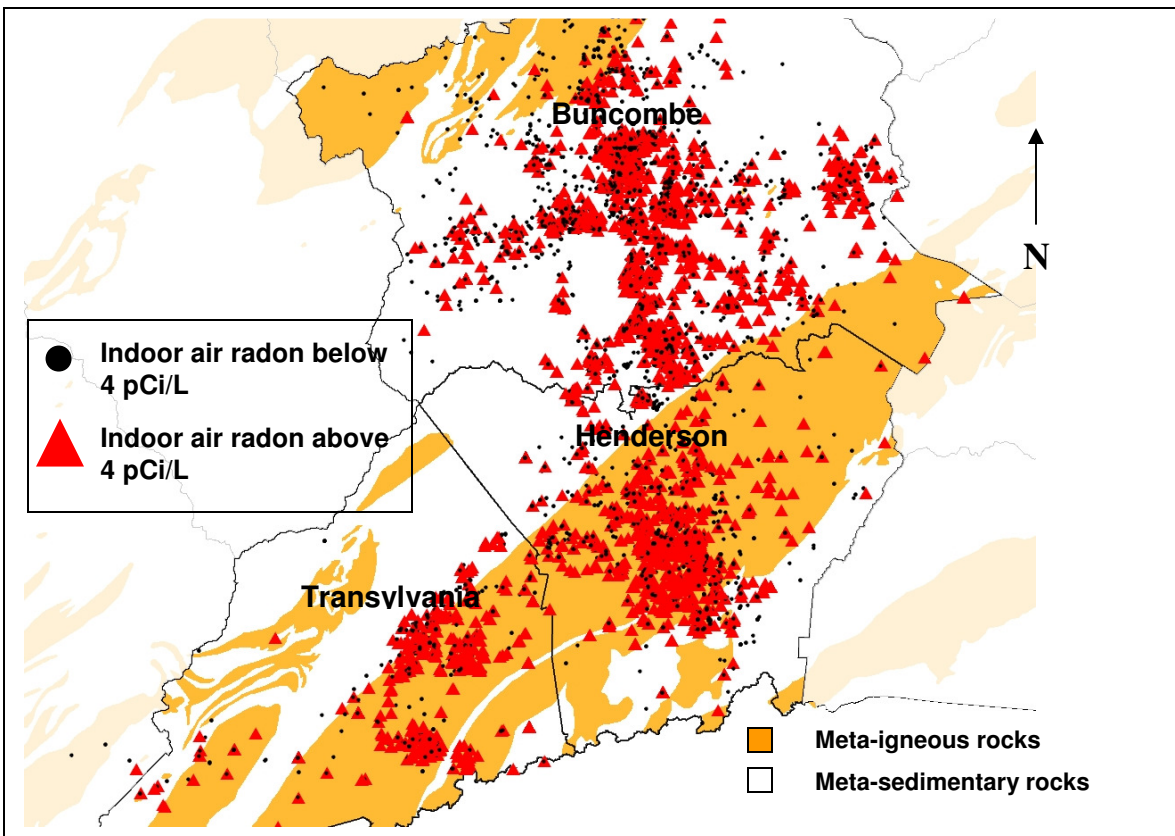
Figure 3. Indoor air radon in homes in Buncombe, Henderson, and Transylvania Counties, North Carolina (data obtained from Airchek, Inc., written communication, 2005).

Contribution of geology and geologic structures

Most uranium bearing minerals contain only trace amounts of uranium as an accessory to other major constituents, and rocks with mineable quantities of uranium are relatively rare. Uranium-bearing minerals include pitchblende, uraninite, carnotite, torbernite, tyuyamunite, autunite, uranophane, and brannerite. Other minerals contain only trace amounts of uranium and include davidite, samarskite, and euxenite. The mineral monazite occurs in North Carolina and is thorium bearing. Uranium rich minerals often occur in pegmatites, for example, or in deposits of sedimentary rocks (pitchblende). In intrusive and metamorphic terrains, uranium concentrations may vary widely depending on the composition and geologic history of the local rock. Often uranium will occur in oxide minerals that may resist chemical alteration and breakdown. Conversely, uranium may be found in secondary minerals coating fractures and were formed as precipitates from circulating ground water or convection of hydrothermal fluids.

Aeroradiometric mapping was completed by Duval and others (1989) using NURE flight line data, and high levels of radioactivity (greater than 2.5 equivalent uranium parts per million) were found to be associated with granites, granitic and meta-sedimentary gneisses, pegmatites, monazite-rich rocks and sediments, and major faults. The North Carolina Geological Survey compiled a geochemical atlas of the State showing areas of elevated uranium activity in stream sediment (Reid, 1993).

In the Blue Ridge Belt, uranium rich rock occurs in the Henderson and granitic gneisses and other meta-igneous rocks of Transylvania County, Henderson County, and others, the graphitic phyllite and schist of the Grandfather Mountain Window, the Wilson Creek Gneiss (along North Harper Creek in Avery County) (Feiss and others, 1989), the Spruce Pine pegmatite district (containing allanite and monazite), and the Alligator Back Formation north of the Bowens Creek Fault (a northern extension of the Brevard zone). Granitic bodies may contain several uranium bearing minerals including spene, zircon, uraninite, allanite, monazite, and other more obscure uranium and thorium minerals (Costain and others, 1986). In the Inner Piedmont, uranium rich rock is attributable largely to the mineral monazite found in high-grade metamorphic rocks and late-stage granitic intrusive rocks. Monazite is particularly abundant in the sillimanite-bearing schists and gneisses of this region.

The average concentration of uranium in soil is about 2 parts per million (ppm) (about 2 grams of uranium in 1000 kg of soil or rock). Rogers and Adams (1970) reported uranium concentrations for various types of rock (table 2). Acidic rocks with a high silicate content – such as granitic rocks - tend to have a higher than average uranium content. An average granite, a rock type common to Western North Carolina and the most common in the earth's continental crust, produces about 3.6 disintegrations per minute per gram (dpm/g) of uranium-238, 0.17 dpm/g of uranium-235, and 5.3 dpm/g of thorium-232.

Table 2. Ranges of typical uranium content in various rock types (Rogers and Adams, 1970).

Rock classification	Rock type	Uranium content, ppm*
Igneous	Granite, rhyolite, quartz monzonite	1 - 4
Igneous	Mafic, ultra mafic	0.001 – 1
Metamorphic	Various	Varies widely
Sedimentary	Quartz sandstone	0.45
Sedimentary	Shale	2.2

* parts per million

Geologic structural features also may contribute to elevated radionuclide concentrations in air and ground water. For example, brittle and ductile fault zones have been linked to elevated radionuclide concentrations (Gundersen, 1991). A terrestrial gamma-ray survey for the conterminous U.S. (Phillips and others, 1993) showed that equivalent uranium-238 concentrations were elevated along the northeast trending Brevard fault zone that cuts through Western North Carolina. Ductile shearing of rocks containing uranium rich minerals such as allanite, monazite, zircon, and titanite results in alterations of the microstructure, porosity, permeability, and chemical composition of the parent rock, and thereby increases the potential for higher radon concentrations in indoor air and ground water (Gundersen, 1991). And while the Brevard zone primarily exhibits ductile shearing, other areas of the zone show evidence of brittle deformation (Hatcher and others, 1989) that also may contribute to elevated concentrations of radionuclides in ground water.

Several physiogeochemical effects occur during the shearing process that may cause increased radionuclide concentrations. For example, as the grain diameters are reduced during shearing, the radionuclide atoms inside the grains are moved closer to the surface and are able to release a greater percentage of radioactive particles that would otherwise remain bound up within the grains (surface area to volume ratio increases). Further, radionuclides are redistributed into the foliation where they are more likely to be in close contact with weathering and transmissive zones, further increasing the rock's overall emanation potential. Ductile shearing also imparts a fabric to the rock that can lead to increased weathering and greater permeability, increasing the rock's transmissive properties. And finally, the shearing process can introduce uranium-rich fluids into the highly transmissive shear zone.

Faulting also causes distinctive physiogeochemical changes in the rock that result in an increase in weathering. Increased weathering is evidenced by the metal oxide "stains" common in fault and shear zones. These oxides (most notably iron and manganese) serve to sorb radionuclides that are present as charged ions or ion pairs such as uranium and radium, for example. Thus, decay elements of uranium and radium (radon, for example) are produced "on site" from the solid phase material and become available for transport in ground water by emission, desorption, or dissolution from the surface of the secondary mineral (Gundersen, 1991).
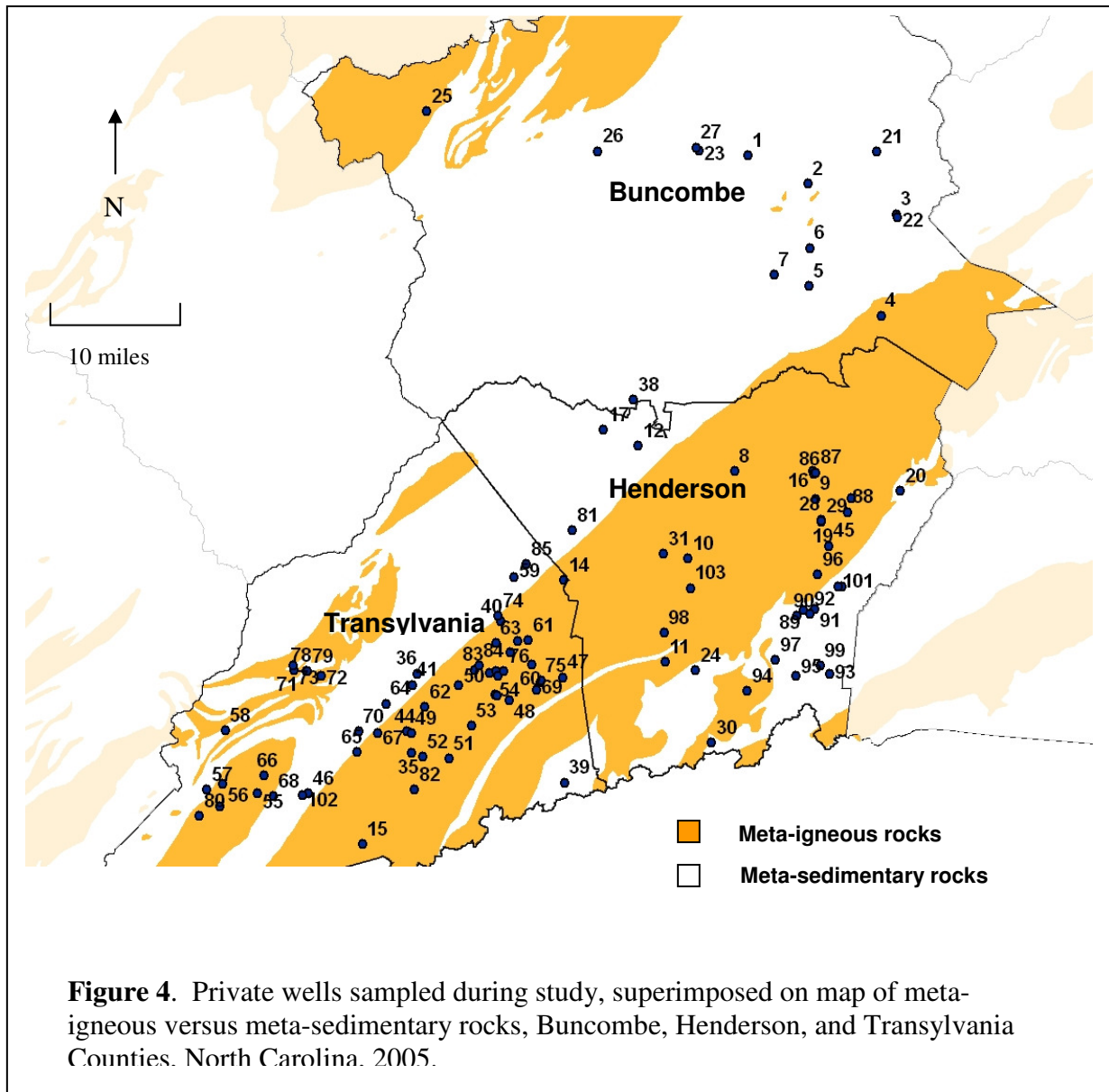
Data Collection and Analytical Methods

Ground-water samples were collected between May and August 2005, from 103 private wells within the study area (fig. 4). In addition, 67 indoor radon samples were collected from the homes associated with the sampled wells. The remaining 36 homes did not participate in the indoor-air radon sampling or did not obtain reliable results. Collecting samples in well-defined target areas helped to maximize limited resources by focusing on areas that were presumed to be among those with the greatest radionuclide concentrations. Newspaper advertisements and word of mouth were used to solicit volunteers for the study.

Each ground water sample was analyzed for total uranium, Ra-226, gross alpha-particle activity, radon, iron, manganese, lead, arsenic, alkalinity, bicarbonate, pH, DO, specific conductance, and temperature. Twenty-one samples also were analyzed for Ra-223 and Ra-224. Quality control replicate samples were collected and analyzed at nine wells. The sample collected from the abandoned public water supply well was not analyzed for total uranium, Ra-226, or gross alpha because historic data already existed for these constituents. Each well samples was identified by a sequential number from 1 to 103 (fig. 4).

Other than the small number of quality-control samples, each well was sampled on one occasion. Therefore, data collected in this study represent a "snap shot" of radionuclide concentrations at a point in time, and do not account for potential temporal variations due to long-term, seasonal, or pumping-related fluctuations. A single sample does not necessarily represent the overall quality of the ground water resource over a long period of time at that location, but it does provide an indication of the quality of the local ground water contributing water to the well for the time at which it was sampled.

Indoor air radon samples generally were obtained from a walkout basement (if present) or a first floor room with minimal ventilation. As such, the results represent what are believed to be worst-case levels. One indoor air sample was collected per site on one occasion, over a three-day period. Because of this, the sample did not account for changes that may occur due to long-term or seasonal fluctuations. Factors that may affect the observed concentration over time include height of the water table, timing and amount of recent rainfall, degree of indoor ventilation and fresh air circulation, variations in well operation and its proximity to the home, and other factors. Nevertheless, the sample represented a value obtained under normal conditions.

Rock type and structure were identified by on-site observation or by statewide or local scale geologic maps (North Carolina Geological Survey, 1985). Because of the geologic complexity of the region, in some cases the geologic setting of a particular home or well had to be inferred. Nevertheless, the designations used in this study were believed to be reasonable characterizations that allowed meaningful evaluations of geologic influence on radionuclide concentrations.

Figure 4. Private wells sampled during study, superimposed on map of meta-igneous versus meta-sedimentary rocks, Buncombe, Henderson, and Transylvania Counties, North Carolina, 2005.

Ground-water data collected in this study were augmented by historic public water supply data obtained during compliance monitoring of public well systems (written communication, M. Fillinger, North Carolina Department of Environmental Health, Public Water Supply, Jan 12, 2005). Information about rock type and structure in the vicinity of the public water supply systems was obtained from geologic maps or site-specific knowledge of the researchers. In many cases a given public water system is connected to several supply wells; a varying number of these wells were sampled for radionuclides. To maintain a consistent dataset, only one value – the maximum – and one location were used per public water supply system, regardless of the number of source wells that supply the system.

Sample-collection methods

A ground water sample was collected as an unfiltered, raw water sample as close to the wellhead as possible. The sample was collected after the pump had been operating for at least 20 minutes and after about 80 gallons or more of water had been removed. This helped to ensure that the sampled water was from the formation and not from a stagnant water column from within well bore. Ground water was placed in a 1-liter plastic container for the analysis of total uranium, gross alpha activity, and Ra-226. The sample date, time, and location were written on the sample container and on the chain of custody form. The sample was shipped by overnight mail to a certified contract laboratory in Oklahoma. Radon samples were collected using a special procedure designed to prevent aeration. Specifically, 40-milliliter glass radon vials were carefully submerged, filled, and sealed inside a 1.8 liter glass beaker that had been slowly filled with well water. The radon samples were iced and shipped to the certified laboratory by overnight mail in order to meet the 4-day holding time requirement. The metals samples (arsenic, lead, manganese, and iron) were preserved using ultra-pure nitric acid prior to shipment to the laboratory.

For Ra-224 and Ra-223 analyses, groundwater samples were collected and allowed to flow through manganese-impregnated acrylic fiber (Mn-fibers), which extracts radium from the solution (Moore, 1976). Typically about 40 liters of sample water was transferred at a rate of less than 2 L/minute through 10 grams of Mn-fibers packed in a 25-cm long filter cartridge. After the radium sample was collected, the water volume that was filtered was recorded, along with the date, time sampling ended, and any other pertinent information. The Mn-fiber was removed, sealed in a plastic bag, and shipped overnight to Stanford University for Ra-224 and Ra-223 measurements. At Stanford University, the Mn-fiber samples were placed in a photomultiplier tube–based counting system for the determination of Ra-223 and Ra-224 by delayed coincidence counting (Moore and Arnold 1996). An initial counting period measured the radon-219 and radon-220 produced by the decay of Ra-223 and Ra-224, respectively, which were adsorbed onto the Mn-fibers.

Field parameters such as DO, specific conductance, pH, ORP, and temperature, were measured in the field using a calibrated YSI 556 MPS multimeter. Information about well construction (depth, casing depth, yield, and others) was noted and recorded in the field. Survey-grade Global Positioning System (GPS) receivers were used to identify the locations of the private water supply wells sampled, and the resulting data were entered into Geographic Information System (GIS) data files. Duplicates were collected for about 10 percent of all radionuclide samples and were analyzed and found to vary by less than 15 percent.

Indoor air radon samples were collected using deployable activated carbon air-sample kits. The sampler was placed in the lowermost unventilated area of the home – typically a walkout basement if it existed - and left undisturbed for 72 hours. The sampler was then sealed and shipped overnight to the laboratory for analysis of the radon concentration.

Laboratory analytical methods

Radon in water was analyzed using a procedure based on Standard Method 7500-Rn (EPA, 1999b). In this method, radon is partitioned selectively into a mineral-oil scintillation cocktail immiscible with the water sample. The sample is dark-adapted, equilibrated, and then counted in a liquid scintillation counter using a region or window of the energy spectrum optimal for the specific alpha particles emitted from radon. Gross alpha-particle activity was analyzed using EPA method 900.0 (EPA, 1980). This method is a screening technique for monitoring water supplies for alpha activity according to the limits set forth by the Safe Drinking Water Act. Counting occurred between day 30 and 35 after the sample was collected, and, therefore, did not account for contributions of radon and Ra-224 (with half lives of 3.8 and 3.6 days, respectively) that were present in the sample at the time of collection. Radium-226 was analyzed using a modification of method SM7500 Ra (EPA, 1995). The method uses alpha spectroscopy. Total uranium was analyzed using method KPA ASTM 5174M (ASTM, 1994). The sample was digested with nitric acid and peroxide and measured by the laser-based kinetic phosphorescence analyzer (KPA).

STUDY AREA SETTING

The study area is comprised of three counties in Western North Carolina - Buncombe, Henderson, and Transylvania – and straddles the Blue Ridge and Inner Piedmont physiographic provinces (fig. 5). The topography of the Blue Ridge province was formed by uplift, erosion, and rock resistance, and is characterized by steep, rugged, incised, mountainous terrain, intermontane basins, and valleys. Part of the Appalachian Mountain system, the Southern Blue Ridge province has a large number of peaks, some with elevations of over 6000 ft above sea level (asl). The topography of the Inner Piedmont was formed through the same earth processes and is characterized by gently rolling, rounded hills, long low ridges, and shallow valleys, with elevations ranging from about 600 to 1500 ft asl.

Precipitation in the study area ranges from about 45 to 60 inches per year, but approaches 100 inches in localized areas. Ground water is particularly important to this region, and about half of the residents rely on it as their principal drinking supply. Yields from private wells typically range from about 1 to 50 gallons per minute (gpm), with averages of about 10 to 15 gpm (Daniel and Dahlen, 2002). Figure 6 shows a cross section of a typical well in the study area.
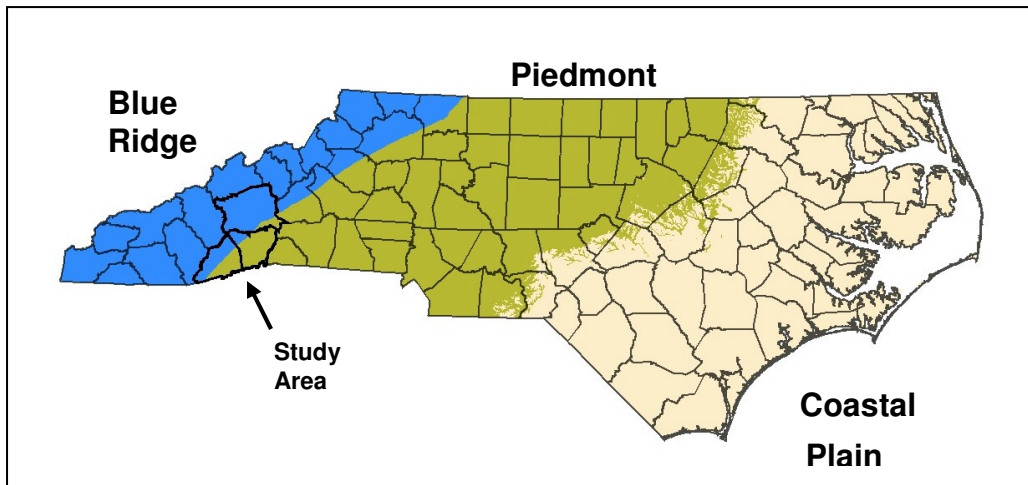
Figure 5. Physiographic provinces in North Carolina and three-county study area.
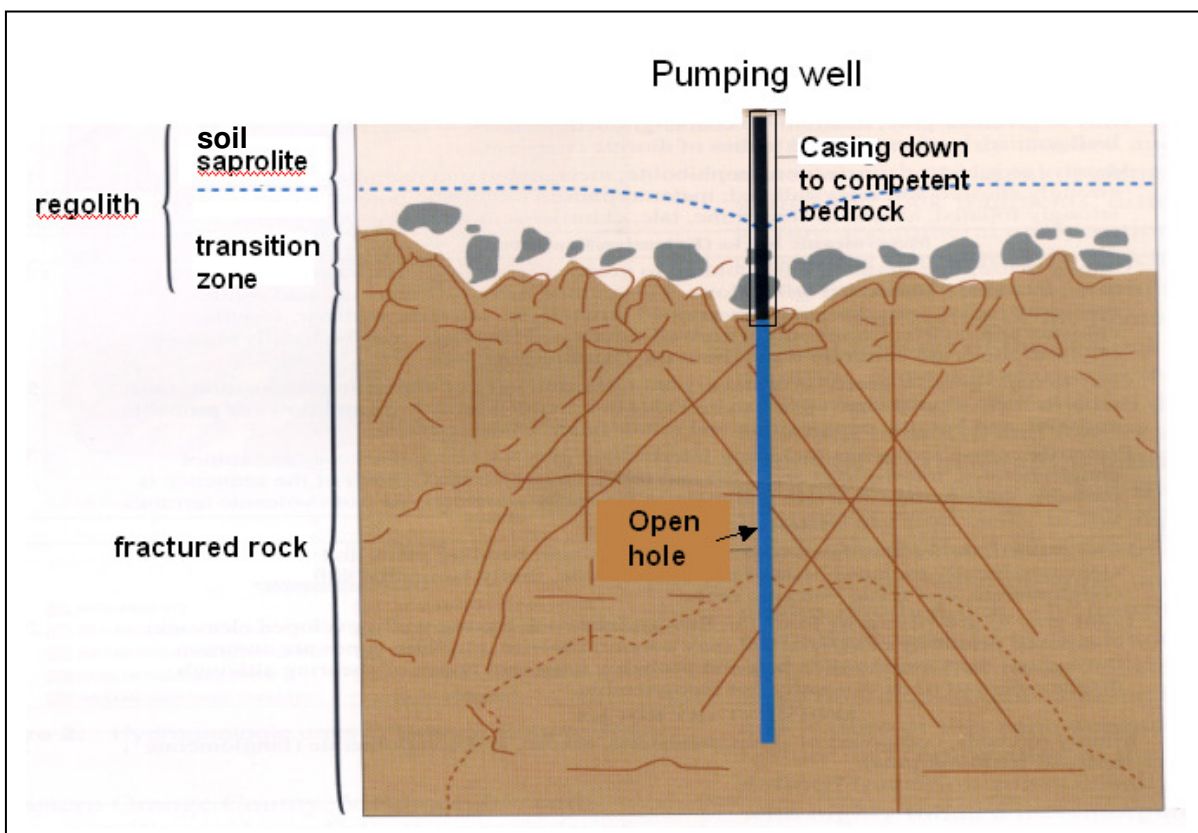
Figure 6. Schematic showing construction of typical private drinking water well in study area.

In the broadest sense, rocks in the study area can be grouped into either *meta-igneous* or *meta-sedimentary* rocks, as shown in fig. 7. Meta-igneous rocks are of igneous origin, and meta-sedimentary rocks are of metamorphosed sedimentary origin. Minor amounts of igneous metavolcanic rocks may occur within the rocks grouped as meta-sedimentary in nature. In the three-county study area, meta-igneous rocks consist largely of granitic gneisses that underlie a large swath of Henderson and southeastern Transylvania Counties. The meta-sedimentary rocks are metamorphosed sedimentary rocks of the Ashe Metamorphic Suite - Tallulah Falls Formation and they underlie a large portion of central Buncombe and northern Transylvania County.

Figure 7. Meta-igneous versus meta-sedimentary rock in study area, Buncombe, Henderson, and Transylvania Counties, North Carolina.
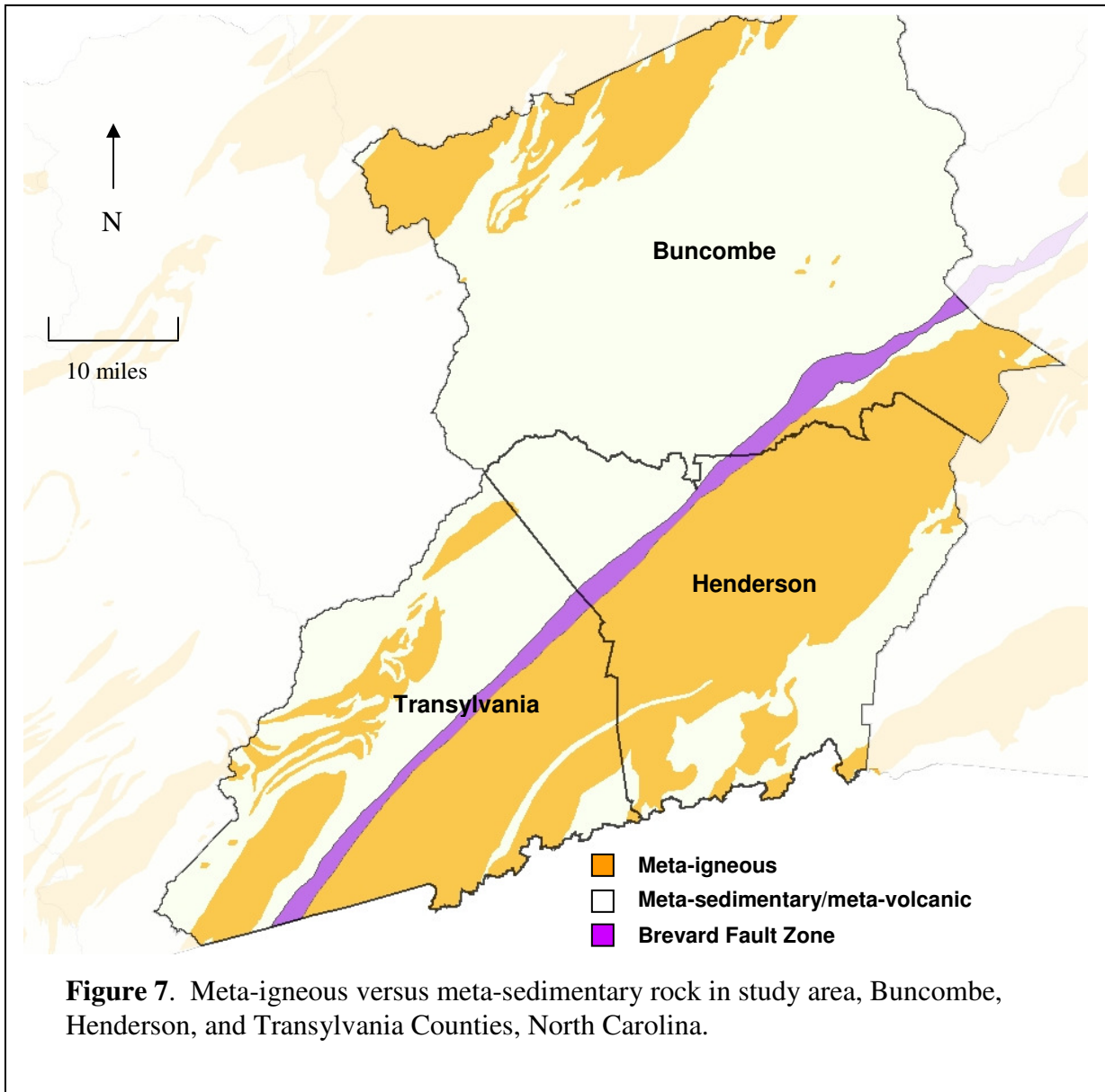
The meta-igneous and meta-sedimentary rocks in the study area may be further divided into individual formal and informal rock units. Meta-igneous rocks include Henderson Gneiss, Toxaway Gneiss, biotite granitic gneiss, migmatitic biotite-hornblende gneiss, granite gneiss, (meta)granodiorite, Caesars Head Granite, and amphibolite (fig. 8). Meta-sedimentary rocks include migmatitic biotite gneiss, garnet-mica schist, metagraywacke, biotite gneiss, muscovite-biotite gneiss, and rocks of the Brevard Fault Zone (fig. 9).

Figure 8. Meta-igneous rock in study area, sub-grouped by individual rock formation, Buncombe, Henderson, and Transylvania Counties, North Carolina.

Figure 9. Meta-sedimentary rock in study area, sub-grouped by individual rock formations, Buncombe, Henderson, and Transylvania Counties, North Carolina.
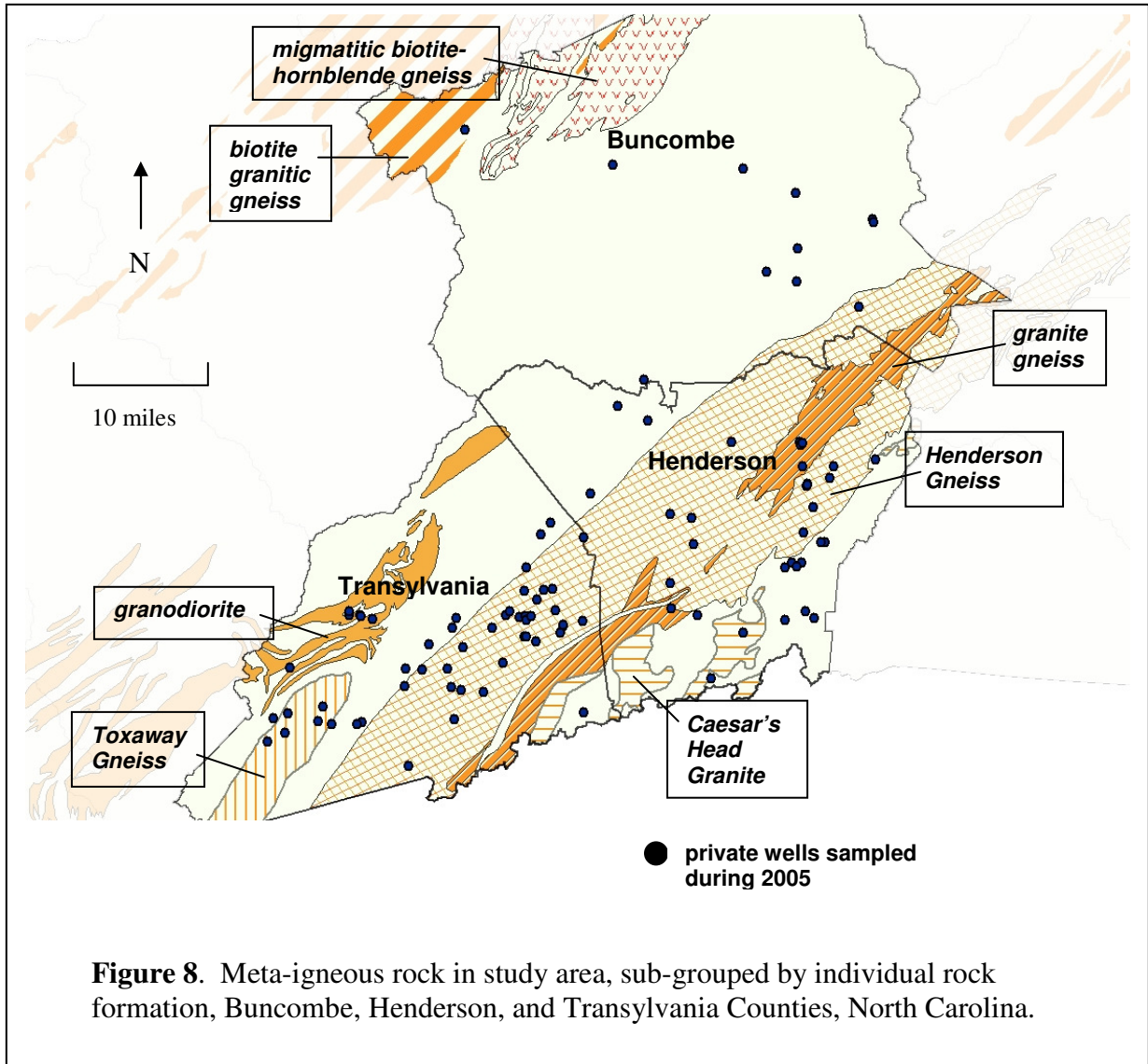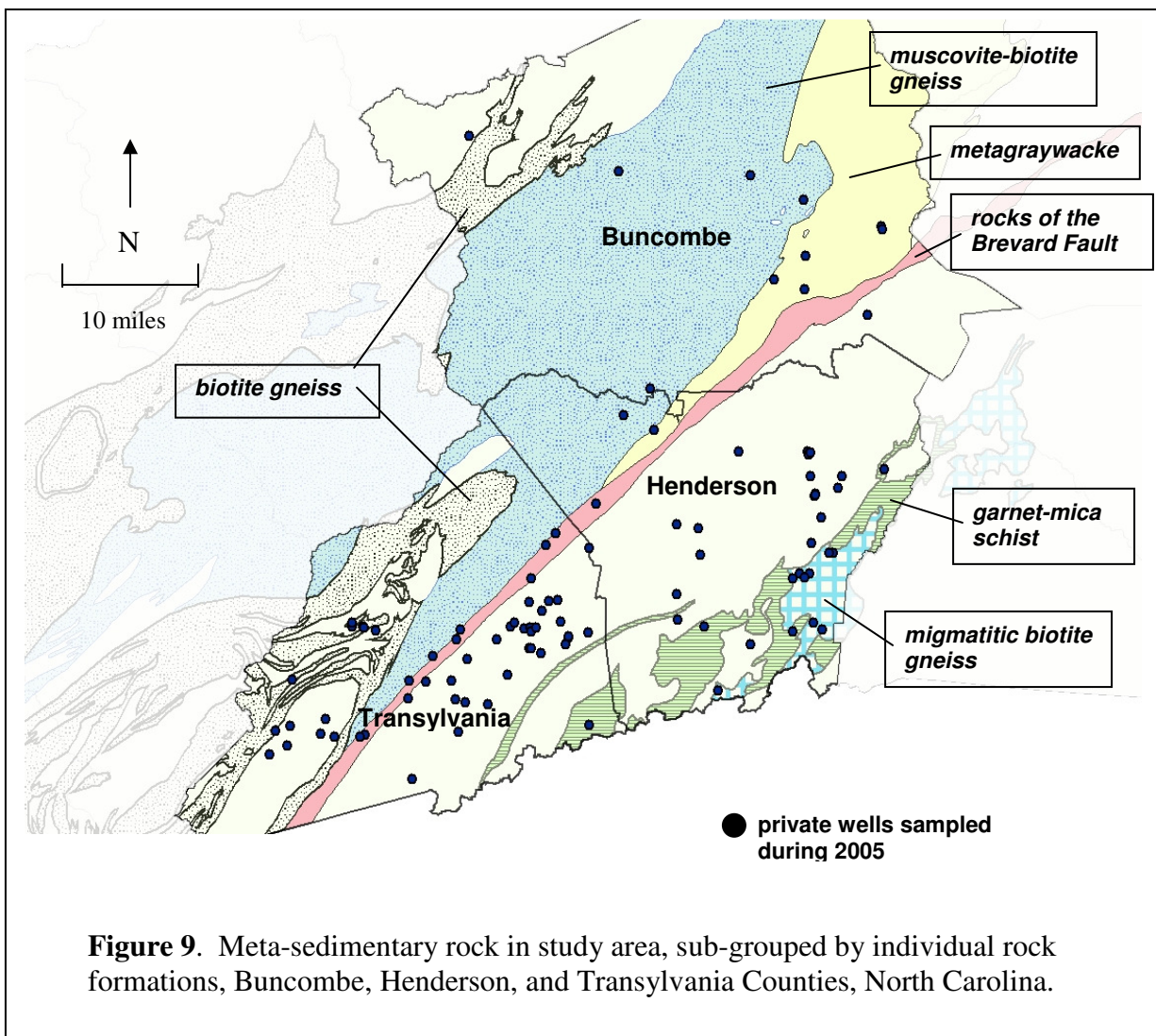
The 500 million year old rocks of the Henderson Gneiss are of igneous origin (Lemmon and Dunn, 1975) and crop out just south of the Brevard Fault Zone in all three counties in the study area. These rocks are variably mylonitized biotite granitic gneiss that contains medium to coarse-grained, rounded or elongated porphyroclasts (augens) of feldspar, in a finer-grained matrix of quartz, feldspar, biotite, muscovite, epidote, and minor amounts of titanite. The Henderson Gneiss is a biotite granitic augen gneiss. The more mylonitic version is finer grained and thinly laminated with microcline porphyroclast. The Henderson Gneiss varies in composition and texture, and distinguishable deformation gradients occur within and across the Brevard Fault Zone, from protomylonite to ultramylonite (Bobyarchick, 1999). The area around Rosman, North Carolina contains Henderson Gneiss along with brecciated phyllonite and ultramylonites.

The Toxaway Gniess in southwestern Transylvania county consists of the 1.15 billion year old granitic gneisses that were formed during the earliest episode of mountain building in the southern Appalachian range. These rocks occur as either layered biotite

granitic gneiss, a gneiss that exhibits distinct, alternating layers of dark colored biotite-rich and light colored quartz and feldspar-rich bands, or megacrystic biotite granitic gneiss, a gneiss that contains abundant, large eye-shaped porphyroclasts (augen) of feldspar. Rocks of the Toxaway Gneiss are likely of a felsic igneous intrusive origin and were metamorphosed during the Middle Proterozoic Grenville orogeny about one billion years ago (Carrigan and others, 2003). More recent Paleozoic metamorphism and mylonitization has altered and realigned the original constituent minerals.

The 950 to 1250 million year old biotite granitic gneiss located in northwestern Buncombe County is granitic to quartz monzonitic, massive to well foliated, and includes variably mylonitized orthogneiss and paragneiss and interlayered amphibolite. The 1214 million year old migmatitic biotite-hornblende gneiss located in northern Buncombe County consists of layered biotite-granite gneiss, biotite-hornblende gneiss, and amphibolite. The 438 million year old granite gneiss is a poorly foliated intrusive rock located in the southeastern part of the study area and contacts the Henderson Gneiss in all three counties. The 390 million year old (meta)granodiorite located in western Transylvania contains biotite, muscovite, and xenocrysts. The 409 million year old Caesars Head Granite located in southern Henderson and southeastern Transylvania Counites is massive to well foliated and contains biotite and muscovite. And locally occurring amphibolite is massive to well foliated, consists of interlayered intrusive and extrusive mafic rock, and may include meta-sedimentary rock.

Meta-sedimentary rocks of the 500 million year old Ashe Metamorphic Suite-Tallulah Falls Formation occur in central Buncombe and northern Transylvania Counties and are represented by mica schist, metagraywacke, and paragneiss (Hatcher, 1971). These rocks are interlayered with mafic metavolcanic rocks. Similar rocks also occur in southeastern Henderson County as Inner Piedmont metamorphic formations. They typically are migmatitic and interlayered and grade into other rock types. The mica schists are derived from metamorphosed siltstones and shales and are medium- to coarse-grained, lepidoblastic, and composed of quartz, feldspar, biototite, muscovite, and garnet. The metagraywackes are derived from metamorphosed muddy sandstones and are fine- to medium-grained, granoblastic, and composed of quartz, feldspar, biotite, muscovite, garnet, epidote, and apatite. The paragneisses include biotite-muscovite gneiss and biotite gneiss that extend across a large portion of central Buncombe and northern Transylvania Counties. Migmatitic biotite gneiss (a porphyroblastic gneiss) occurs in southeastern Henderson County.

Rocks of the Brevard fault zone lie in a northeast trending belt between the Ashe Metamorphic Suite-Tallulah Falls Formation and the Henderson Gneiss, and include early Paleozoic to late Proterozoic aged metagraywacke, marble, metasiltstone, phyllite, phyllonite, and graphitic breccia (Hatcher, 1969). Original deposition occurred between 500 and 600 million years ago. Mylonites and ultramylonites are prevalent within and along both sides of the Brevard Fault Zone (fig. 9). Some areas on the northwest edge of the Brevard Fault Zone show evidence of brittle deformation (Hatcher and others, 1989). These areas are being evaluated for their potential to result in higher concentrations of radionuclides due to increased permeability and foliation exposure.

A number of uranium-bearing minerals are found in Western North Carolina, primarily as minor constituents. Rock types in the region that typically contain elevated levels of uranium include meta-igneous rocks (pegmatites, diorites, granites, and granitic gneisses, for example) of the Blue Ridge Belt and the Inner Piedmont. The paragneisses formed by the metamorphism of sedimentary rocks may also contain elevated levels of uranium, but typically at somewhat lower levels than those of intrusive parent rocks.

Geochemical results obtained during the study are summarized in Table 3. The table also provides information on casing depth, a proxy used in this study to estimate the regolith thickness, and well depth. Taken as a whole, sampled ground water tended to be slightly acidic (median pH = 6.2), oxygenated (median DO = 7.4 mg/L), and minimally conductive (median SC = 65 uS/cm). The buffering capacity of the ground water was fairly low (median alk = 23 mg/L), and the levels of iron and manganese also were low (median Fe < 25 ug/L and median Mn < 5 ug/L). Raw oxidation-reduction potential (ORP) values were moderately high (median raw ORP = 308 mV).

Table 3. Descriptive statistics for field parameters and well characteristics measured in study wells in Buncombe, Henderson, and Transylvania Counties, North Carolina, 2005.

Parameter	No. of samples	Median value	Maximum value	Minimum value
pH	38	6.2	8.0	<4
Specific conductivity, in uS/cm		65	328	<10
Temperature, in degrees Celsius		14.8	>18	12.7
Dissolved oxygen, in mg/L		7.4	9.7	0.4
Oxidation reduction potential, in mV		308	758	-186
Lead, in ug/L		< 10 (DL)	24	< 10 (DL)
Arsenic, in ug/L		<5 (DL)	<5 (DL)	<5 (DL)
Iron, in ug/L	102	< 25 (DL)	15000	< 25 (DL)
Manganese, in ug/L	102	< 5 (DL)	100	< 5 (DL)
Alkalinity, in mg/L	103	23	120	< 1 (DL)
Casing depth, in feet	57	65	196	2
Well depth, in feet	81	305	905	27
Well yield, in gpm	72	5.5	100	0

[handwritten/annotation box overlapping No. of samples column: "38 water may become oxygenated inside the well bore due to water level fluctuations caused"]

uS/cm, microSiemens per centimeter

mg/L, milligrams per liter

mV, millivolts

ug/L, micrograms per liter

gpm, gallons per minute

Because of moderately high DO levels, low dissolved iron and manganese, and moderately high ORP levels, most ground water in the study area was considered to be oxidizing. It should be noted that in some cases otherwise *an*oxic ground (formation)