Dissolved Radon and Uranium, and Ground-Water Geochemistry in an Area near Hylas, Virginia

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Dissolved Radon and Uranium, and Ground-Water Geochemistry in an Area near Hylas, Virginia

By Mark R. Stanton,[1] Richard B. Wanty,[1] Errol P. Lawrence,[2] *and* Paul H. Briggs[3]

ABSTRACT

Radon gas (^{222}Rn) dissolved in domestic well water is a potential source of indoor radon. Recent evidence suggests that ground water in contact with rocks deformed by shear may contain higher concentrations of ^{222}Rn than ground water from adjacent unsheared rocks. Fifty-two ground-water samples from crystalline-rock aquifers in and near the Hylas shear zone of central Virginia were analyzed to investigate the relationship between water chemistry, dissolved ^{222}Rn and uranium (U), and aquifer type. Forty-four samples have concentrations of dissolved ^{222}Rn that exceed the recently proposed United States Environmental Protection Agency (EPA) maximum contaminant level (MCL) of 300 pCi/L (picocuries per liter). Eighteen ground-water samples from mylonite of the Hylas shear zone range from 360 to 32,000 pCi/L ^{222}Rn (average=5,800 pCi/L). Twelve ground-water samples from adjacent granitic intrusive rocks range from 400 to 81,000 pCi/L ^{222}Rn (average=14,900 pCi/L). Dissolved ^{222}Rn values are significantly lower in the 22 ground-water samples from metamorphic rocks located west of the shear zone and range from 80 to 2,300 pCi/L ^{222}Rn (average=550 pCi/L). In all samples, dissolved ^{222}Rn is unsupported by dissolved U, suggesting that excess ^{222}Rn is provided by emanation from aquifer rocks.

Mineral-solution equilibria calculations using the measured chemistry of the waters indicate that all waters are greatly undersaturated with respect to some common uranium minerals. Only 3 of the 49 water samples analyzed for U have concentrations greater than 1 part per billion (ppb). Adsorption of uranium onto mineral surfaces is probably the key process that limits dissolved U concentrations to such low levels. Ground waters from mylonite have higher average pH, total carbonate ($\Sigma CO_3{}^{2-}$), and dissolved U concentrations, suggesting the formation of uranyl-carbonate complexes ($UO_2{}^2+(CO_3)_n{}^{2-2n}$) as a means for solubilizing or mobilizing U from these rocks. Uranyl ion ($UO_2{}^{2+}$) or uranyl-carbonate complexes dissolved in ground water may be transported via fractures and adsorbed onto ferric oxides that coat grains and fracture surfaces, where U eventually decays to produce aqueous ^{222}Rn. Lower average pH, $\Sigma CO_3{}^{2-}$, and dissolved U concentrations in ground waters from granitic and metamorphic rocks suggest that U-CO_3 complexing occurs to a lesser extent in these aquifers and that adsorption and retention of U on mineral surfaces of these rocks is more effective than on mineral surfaces of the mylonite. Lower dissolved U may also be the result of less abundant labile U in primary minerals of the granitic and metamorphic rocks. The high concentrations of ^{222}Rn in ground waters from granite and mylonite are due to greater abundances of uranium and radium (Ra) at grain surfaces in contact with ground water. According to this model, the mobility and enrichment of ^{222}Rn in these ground waters is largely a function of the location of U and Ra near the mineral/water interface in the aquifer rocks.

INTRODUCTION

Ground water used for domestic supply may contribute radon gas (^{222}Rn) to indoor air (Hess and others, 1990; Lawrence and others, 1991; Nazaroff and others, 1988). Radon-rich ground water brought into enclosed living areas can release dissolved ^{222}Rn into indoor air when the water is used for household purposes. Degassing, combined with other processes such as soil gas leakage into basements, could lead to an increase in the airborne ^{222}Rn concentration in homes and buildings (Crawford-Brown, 1990).

The original accumulation of ^{222}Rn in domestic well water supplies and the subsequent contribution of ^{222}Rn to indoor air in homes through degassing depends on the distribution, mobility, and contrasting chemical behavior of ^{222}Rn, its immediate parent radium (^{226}Ra), and its ultimate parent uranium (^{238}U) (Brutsaert and others, 1981; Nazaroff and Nero, 1988). Because U can exist in several oxidation

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states (usually reduced U(IV) or oxidized U(VI)) and form complexes with different aqueous ions, U solubility is strongly influenced by changes in solution chemistry, Eh, and pH (Langmuir, 1978). Aqueous sulfate and carbonate complexes of radium (as Ra^{2+}) that form in natural waters are usually adsorbed onto mineral surfaces or coprecipitated with sulfate minerals, such as barite, or carbonate minerals, such as calcite (Ames and others, 1983). As a result, the abundance of dissolved Ra in natural waters rarely exceeds 10^{-12} M (Langmuir and Melchior, 1985). ^{222}Rn possesses a noble gas configuration and is chemically unreactive; ^{222}Rn mobility is therefore controlled primarily by physical factors, especially its contrasting solubility in water and air. Abundances of U, Ra, and Rn in ground water may depend on related factors such as U and Ra content of parent rock, accessibility of ground water to U- and Ra-bearing minerals, solubility of U and Ra in local ground water, and soil moisture regime (LeGrand, 1987).

The present study compares radionuclide concentrations (U and ^{222}Rn) and geochemistry of ground waters obtained from sheared mylonitic rocks, unsheared granitic and metamorphic rocks, and weathered saprolite overlying these rocks, to examine the effect of rock composition and texture on dissolved ^{222}Rn concentrations. Specific hydrologic properties of aquifer materials (for example, hydraulic conductivity and porosity) that might affect radionuclide abundances were not examined in this study. Increased levels of aqueous ^{222}Rn should be facilitated by enrichment of U and Ra at the mineral-water interface. Such enrichment is produced by weathering, alteration, and shear processes (brittle and/or ductile) that can redistribute U and Ra and perhaps increase permeability and surface area of host rocks. Differences in ground-water chemistry and radionuclide (U and ^{222}Rn) content in the study area do not correlate with well depth or well type, and water chemistry is controlled primarily by the interaction of ground water with the rock or saprolite. The results lend support to the hypothesis that ^{222}Rn emanation is enhanced in high-U rocks that have experienced extensive oxidative weathering and shearing.

GEOLOGY

The study area is located in central Virginia approximately 10 km northwest of the Richmond metropolitan area in Goochland, Hanover, and Henrico Counties (fig. 1). This region is part of the Piedmont Province of the Appalachian orogenic belt. Surficial aquifers in the study area include deeply weathered saprolite zones above fractured bedrock, and Quaternary sediments along stream drainages and other low-lying areas (Gates and Gundersen, 1992). Crystalline bedrock units include Precambrian metamorphic rocks and Paleozoic granites, pegmatites, and mylonites. Water is obtained from both the weathered (saprolitic) and unweathered (but apparently fracture permeable) crystalline rocks, although the water-bearing horizons vary with depth and geographic location. The most transmissive horizons are usually in the basal saprolite, an iron- and clay-rich assemblage produced by extensive chemical weathering of underlying crystalline bedrock. Atlantic Coastal Plain sediments (located east of the area of figure 1) and Triassic sedimentary rocks were not investigated. Geologic contacts are simplified on the geologic-sample locality map to accentuate the contacts of the granitic and metamorphic rocks with the mylonite (shear) zone. A more detailed description of the local geology and mineralogy is given in Gates and Gundersen (1992).

Twenty-two water samples were obtained from Middle Proterozoic metamorphic rocks of the Eastern gneiss complex that are present west of the Hylas shear zone (fig. 1). The Maidens Gneiss, which constitutes most of the mapped gneiss complex in the study area, is composed of several mineralogically distinct units presumed to have been derived from igneous and volcanic precursors (Goodwin, 1970); some units are interpreted as metamorphosed sediments (for example, metapelite). One well water sample was obtained from the Sabot Amphibolite in the western portion of the study area (fig. 1); this amphibolite represents metamorphosed lava flows and pyroclastic debris and is composed of hornblende, plagioclase, quartz, K-feldspar, pyroxene, sphene, apatite, and secondary calcite (Goodwin, 1970). Three additional water samples were taken from wells in an unnamed amphibolite in the Maidens Gneiss. The remaining 18 ground-water samples from metamorphic rocks were drawn from undifferentiated units of the Eastern gneiss complex. These undifferentiated units (not mapped separately in figure 1) include metapelite, hornblende-biotite gneiss, garnet-biotite gneiss, and granite gneiss (Gates and Gundersen, 1992). For purposes of this report, the amphibolites plus other metamorphic rocks are generally treated as a single rock type; in a few instances, the amphibolites are discussed separately from the other metamorphic units.

Eleven ground-water samples were taken from wells in the Petersburg Granite of Mississippian age that is present east of the Hylas shear zone and composed, on average, of 38 percent orthoclase, 35 percent plagioclase, 25 percent quartz, and 2 percent mafic minerals (Ellison and Masiello, 1979). Several small pegmatites (0.5-300 m wide) are present near the contact between the granite and mylonite; only one well within pegmatite was sampled in this study. On the basis of similar mineralogical composition and probable genetic relationships, the sample from the pegmatite (88VA-7) is grouped with samples from the granitic rocks.

The Hylas mylonite is a cataclastic unit composed primarily of quartz, potassium and plagioclase feldspars, and muscovite (sericite); 18 ground-water samples were collected from wells located within this lithologic unit. Mylonite and ultramylonite have been identified in the shear zone, but both are referred to here as mylonite since the primary difference is one of grain size rather than mineralogical composition. Shearing generally does not result in the growth of new or

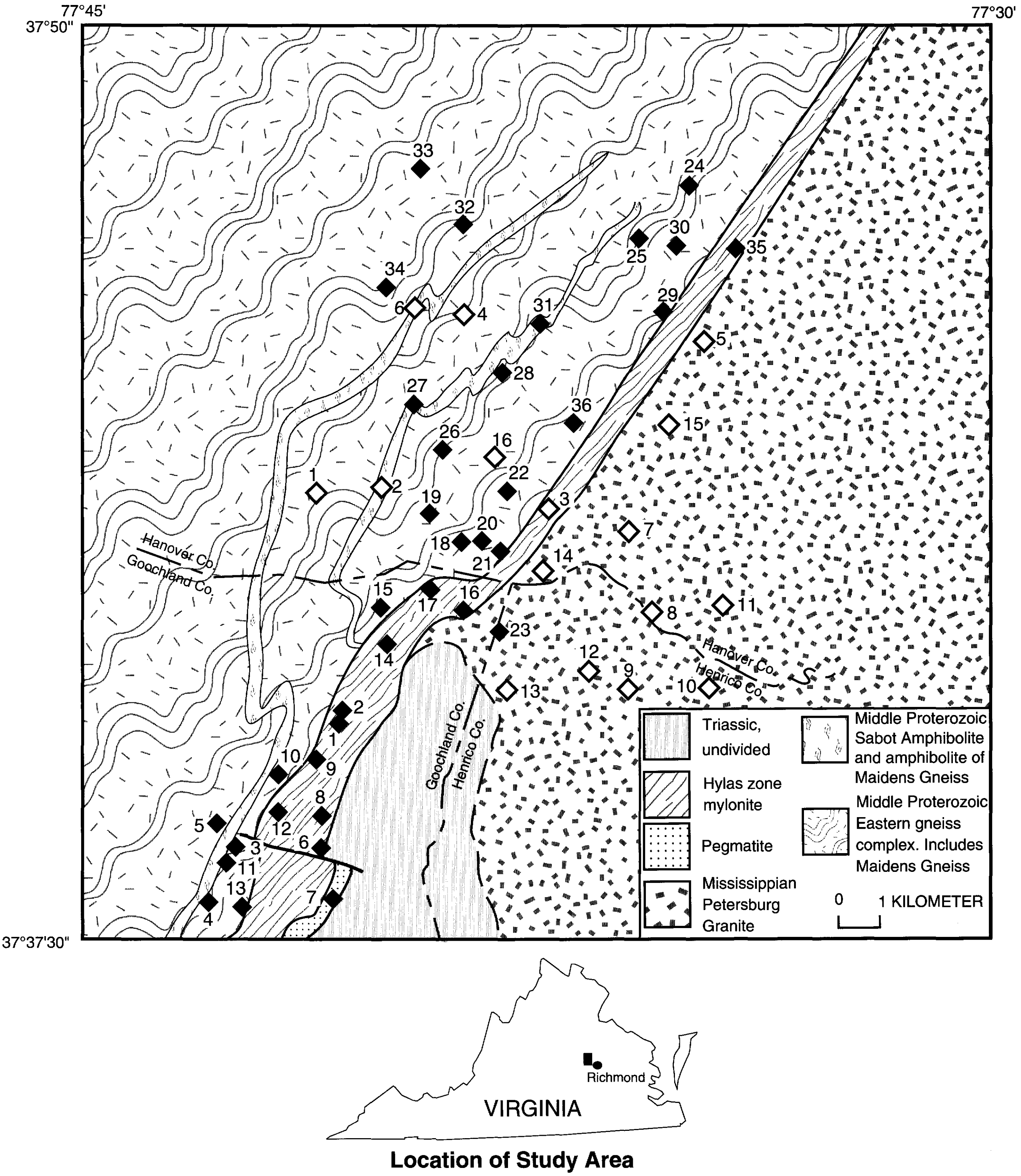


Figure 1. Generalized geologic map of the study area showing locations of ground-water samples. The geologic map base is from Gates and Gundersen (1992). Geology north of approximately 37°47 ′00″ is inferred from mapped units in the remainder of the study area. Sample numbers correspond to those in tables 1 and 2. Filled diamonds represent samples with prefix "88VA," collected in June 1988; open diamonds represent samples with prefix "90VA," collected in May 1990.

different minerals, rather only a reduction in grain size of existing minerals (Jackson, 1970). The mylonites were formed during several periods of brittle, brittle-ductile, and ductile shearing and faulting of adjacent rocks; the last episode of deformation in the Hylas zone was of the brittle type and produced cataclasite and gouge (Gates and Gundersen, 1992). The deformations could have produced a greater mineral surface area in the mylonites relative to adjacent rocks. Petrographic evidence indicates that most mylonite within the Hylas zone was derived from Petersburg Granite, pelitic units of the Maidens Gneiss (Gates and Gundersen, 1992), and volcanic rocks (Goodwin, 1970). Shear zone boundaries are characterized by heterogeneous rock assemblages and irregular contacts, and samples from these areas may encompass multiple lithologies.

Quaternary alluvial and fluvial deposits are largely confined to the margins of streams and rivers in the region. Saprolite ranges up to approximately 30 meters thick and is usually thickest in highly weathered, topographically low ground between drainages. To show bedrock geology more clearly, surficial deposits are not illustrated in figure 1.

FIELD AND LABORATORY METHODS

DESCRIPTION OF WELL TYPES

Water samples were obtained from both augered (or bored) and drilled wells. Available drilling records for the study area indicated augered wells generally penetrated the full thickness of the saprolite but did not extend into unweathered bedrock. Lithologic logs placed the depth of the saprolite (weathered) to bedrock (unweathered) transition at approximately 25 to 30 meters. Augered wells were consistently shallower than this maximum depth to saprolite; that is, all wells above the 30-meter depth are the augered type. Augered wells were lined with circular, 1-meter-diameter terra cotta tiles for their entire depth with pump placement commonly at the bottom of the well.

Drilled wells were as deep as 120 meters and therefore provided water from unweathered crystalline rocks. Drilling records were unavailable for the wells that were sampled, but records from other wells in the study area identified as many as 3 water-producing zones in crystalline rocks; the number of water-bearing zones intersected by a well depended on depth. Screening and the length of open intervals for drilled wells was variable. Wells were usually cased to approximately 10 meters, and generally, the water-bearing fracture zones were screened but the non-producing sections through bedrock were unscreened. Materials used for casing and screening were not identified on all records and grouting was not used in every well. Location of the pump in the drilled wells was not always known with certainty but was usually deeper than 30 meters (that is, below the saprolite zone). The nine wells (4 augered, 5 drilled) whose depths were unknown were not used in the evaluation of changes in water chemistry with depth.

SAMPLING AND ANALYTICAL METHODS

Fifty-one well-water samples and one spring-water sample were collected from an area of approximately 230 km^2 (fig. 1). Sampling was conducted during the first two weeks of June 1988, and the last two weeks of May 1990; both periods were characterized by stable, dry weather. Water was allowed to flow for sufficient time to empty stored water to obtain as fresh a sample as possible and to permit stabilization of temperature (T°C) and conductivity (SpC), which were monitored before sampling. Samples were withdrawn into a gastight syringe from a hose connected to a spigot as close to the wellhead as possible, but before the water entered any treatment system; turbulence and possible ^{222}Rn loss by degassing were minimized by using a slow flow rate through the hose during the entire sampling operation.

Dissolved oxygen (DO), alkalinity (as bicarbonate, HCO_3), and pH were determined at the time of sampling in addition to T°C and SpC; these results appear in table 1 (the electronic charges carried by dissolved ions are not shown in this report unless needed for clarity). Three aliquots were filtered through a 0.45 µm membrane filter for subsequent anion, cation, and uranium analyses. Microbes that might utilize and thus alter the concentrations of certain dissolved constituents, such as nitrate (NO_3), are removed by filtration; in addition, concentrated reagent-grade nitric acid (HNO_3) was added to the cation and uranium samples to prevent adsorption and precipitation during transportation and storage. Inductively-coupled plasma atomic emission spectroscopy (ICP-AES; Lichte and others, 1987) was used to determine concentrations of major and trace cations to within ±10%. Major anions—sulfate, nitrate, phosphate, chloride, and fluoride—were determined by ion chromatography (Fishman and Pyen, 1979) to within ±10%. TDS (total dissolved solids) was calculated for each sample as the sum of cations and anions (Hem, 1989). Individual sample cation + anion (C+A) balances agreed to within ±10%, but ten samples were outside of these limits; these samples generally had low total cations and anions, and low pH and HCO_3; lack of buffering capacity may have resulted in chemical changes that produced the larger analytical error. The mean for all C+A balances was -4.9%. Uranium was determined by laser-induced fluorescence with standard addition; replicate analyses agreed to within ±15%. The method determines total U only; U isotopes and oxidation states (U^{4+} and U^{6+}) are not differentiated. ^{222}Rn was determined by liquid scintillation within 48 hours of sampling on duplicate samples, and a correction factor was applied for the amount of ^{222}Rn that decayed between the time of sampling and counting.

Table 1. Results of chemical analyses for ground-water samples from the Hylas, Virginia, study area.

[Units are as follows: pH (dimensionless), Temperature (°C), Specific Conductivity (SpC, µS/cm), all others, (ppm). Phosphate was below detection in all samples (<0.1 ppm). ND, not detected. Well depths in meters; UNK, unknown. TDS was calculated as the sum of dissolved cations and anions (in ppm)]

SAMPLES OF GROUND WATER FROM GRANITES

Sample No.	pH	DO	Temp.	SpC	Ca	Mg	Na	TDS
88VA-7	5.3	1.3	15	74	11.	1.5	9.3	105
88VA-23	5.6	2.8	16	79	7.1	1.6	19.	119
90VA-5	7.3	8.4	15	50	17.	0.4	4.8	143
90VA-7	4.4	7.4	17	54	1.5	0.4	4.8	51
90VA-8	6.4	1.5	15	53	1.4	1.9	45.	226
90VA-9	5.6	2.9	14	51	9.3	0.8	7.3	101
90VA-10	4.8	6.4	15	48	6.2	0.19	2.1	47
90VA-11	5.6	9.3	17	50	3.3	1.0	7.9	72
90VA-12	5.7	6.0	17	52	13.	0.4	4.9	94
90VA-13	6.0	8.4	15	51	12.	1.9	7.2	121
90VA-14	5.3	3.3	15	47	8.9	1.0	5.3	85
90VA-15	6.6	0.9	17	49	23.	4.0	9.8	198
Mean	5.7	4.9	15.7	54.8	9.4	1.3	10.6	113.5
Std. Dev.	0.8	3.1	1.1	10.4	6.2	1.1	11.5	54.1

Sample No.	K	HCO_3	SO_4	Cl	NO_3	F	Si	Depth
88VA-7	1	46.	2.7	11.	3.0	0.2	18.	11
88VA-23	1	26	6.0	12.	22.	ND	25.	58
90VA-5	15.	95	2.9	2.3	ND	ND	6.1	11
90VA-7	1.7	25	ND	2.1	ND	ND	15.	63
90VA-8	3.2	130	12.	9.3	ND	2.4	18.	91
90VA-9	1.5	62	2.4	13.	0.6	ND	4.7	10
90VA-10	1.3	27	0.3	3.4	0.2	ND	6.0	9
90VA-11	2.2	21	ND	3.6	18.	ND	15.	UNK
90VA-12	2.4	59	0.4	2.3	0.2	ND	12.	10
90VA-13	ND	75	1.5	2	ND	ND	21.	91
90VA-14	1.4	48	1.8	4.4	ND	ND	14	9
90VA-15	3.8	130	4.7	2.4	ND	0.2	21.	93
Mean	3.1	62.	3.5	5.6	7.3	0.70	14.7	
Std. Dev.	4.0	39.	3.4	4.3	9.9	1.1	6.4	

The ^{222}Rn value in table 2 is the average of these two analyses. In nearly every case, the duplicates agreed to within 5%.

STATISTICAL PROCEDURES

Ground waters from the three rock types were compared using average values (arithmetic means) in a single-factor analysis of variance (ANOVA) when values for a geochemical parameter were available, or above the lower limit of detection (LLD), in at least 90 percent of the samples. The ground-water parameters subjected to statistical treatment include pH, DO, SpC, TDS, Hardness, ^{222}Rn, U, Ca, Mg, Na, K, HCO_3, SO_4, Cl, Mn, Si (reported as SiO_2), Ti, Sr, Ba, and Zn. (^{222}Rn was $\log_{10}$-transformed to more closely approximate a normal distribution). Appropriate values greater than zero and below the LLD (0<X<LLD) were substituted in the two cases (U, SO_4) where slightly less than 90 percent of the measurements were above the LLD. Because there were insufficient numbers of samples above the LLD for other trace elements, no statistical or correlative analyses were performed; therefore, most discussions that follow are concerned with major elements and ions, and the radionuclides U and ^{222}Rn. However, some elements or ions are discussed in terms of whether or not they were detected in a ground-water sample (NO_3, Li, Fe). The Scheffé F-test, a conservative estimator of sample variances in 3-group tests, was applied to determine if the means were significantly different

Table 1. Results of chemical analyses for ground-water samples from the Hylas, Virginia, study area.—Continued

SAMPLES OF GROUND WATER FROM MYLONITES

Sample No.	pH	DO	Temp.	SpC	Ca	Mg	Na	TDS
88VA-1	6.9	8.8	4	68	6.0	2.3	8.1	87
88VA-2	6.6	6.2	19	67	5.9	2.8	11.	99
88VA-4	6.9	2.1	15	55	6.3	0.8	7.1	78
88VA-6	6.1	7.6	19	38	3.4	0.4	4.1	62
88VA-8	7.2	0.3	16	74	42.	9.9	15.	285
88VA-9	5.6	1.5	19	90	33.	7.2	10.	225
88VA-10	5.7	8.8	16	24	2.3	0.5	1.9	34
88VA-12	6.5	0.3	17	84	26.	7.2	20	251
88VA-13	6.7	9.0	15	82	26.	2.1	8.4	175
88VA-14	6.5	3.0	15	82	30.	5.6	15.	235
88VA-16	5.6	5.3	15	81	8.6	2.3	10.	102
88VA-17	6.6	4.8	18	84	18.	3.2	15.	155
88VA-20	6.0	3.4	16	82	12.	2.7	18	148
88VA-21	6.6	0.3	18	81	47.	3.8	15.	264
88VA-29	6.0	0.4	18	87	16.	3.6	19.	175
88VA-35	7.0	0.4	15	56	37.	6.8	15	264
88VA-36	7.1	2.7	19	72	33.	5.8	24.	256
90VA-3	6.5	1.8	16	45	23.	4.9	23.	255
Mean	6.5	3.7	16.1	69.6	20.9	4.00	13.3	175
Std. Dev.	0.5	3.2	3.4	18.7	14.1	2.64	6.17	82.2

Sample No.	K	HCO_3	SO_4	Cl	NO_3	F	Si	Depth
88VA-1	2	25.	2.7	6.4	19.	ND	16.	UNK
88VA-2	1	42.	1.1	7.7	5.8	0.5	20.	38
88VA-4	2	31.	ND	9.9	5.8	ND	15.	14
88VA-6	2	33.	0.1	3.0	4.1	ND	12.	11
88VA-8	1	190	11.	7.7	ND	0.2	13.	69
88VA-9	2	130	5.3	8.4	1.5	0.2	23.	116
88VA-10	2	15.	0.2	2.5	2.9	ND	6.7	12
88VA-12	2	170	2.4	2.6	0.4	0.2	24	81
88VA-13	3	110	2.3	2.2	0.8	ND	19	12
88VA-14	2	150	3.3	6.4	ND	0.4	23.	37
88VA-16	2	29.	0.7	10.	22.	ND	18.	9
88VA-17	1	82.	11.	3.9	ND	0.4	19.	88
88VA-20	1	75.	0.7	11.	ND	0.1	28.	12
88VA-21	3	150	6.0	13.	1.6	0.1	21.	9
88VA-29	2	85.	8.6	13.	0.5	0.5	27.	70
88VA-35	3	170	5.4	2.4	ND	0.2	20.	UNK
88VA-36	1	130	20.	14.0	13.	0.2	11.	UNK
90VA-3	6	160	7.8	9.3	ND	0.3	21.	61
Mean	2.11	99.3	4.91	7.36	4.58	0.18	18.7	
Std. Dev.	1.18	59.6	5.17	3.91	6.93	0.18	5.69	

(Scheffé, 1933). In all statistical analyses, a 95 percent confidence level was employed. In addition, the nonparametric Kruskal-Wallis test, which is often applied to data sets containing mainly qualified (below the LLD) values (Till, 1974), corroborated the results and conclusions obtained from the parametric Scheffé method.

Table 1. Results of chemical analyses for ground-water samples from the Hylas, Virginia, study area.—Continued

SAMPLES OF GROUND WATER FROM METAMORPHIC ROCKS (* = AMPHIBOLITE)

Sample No.	pH	DO	Temp.	SpC	Ca	Mg	Na	TDS
88VA-3	7.1	7.4	18	56	6.4	3.4	5.3	89
88VA-5	6.7	0.5	17	120	19.	3.7	8.9	151
88VA-11	6.5	1.9	16	83	20.	6.8	10.	174
88VA-15	6.4	6.7	16	54	6.5	2.1	7.5	102
88VA-18	5.5	5.6	17	62	7.7	1.2	7.6	80
88VA-19	5.3	2.9	17	74	16.	9.7	18.	158
88VA-22	5.6	7.7	15	63	16.	0.9	1.3	82
88VA-24	5.9	6.3	19	92	7.1	3.9	8.2	115
88VA-25	6.4	0.2	16	82	8.3	4.6	19.	161
88VA-26	4.8	7.0	17	38	4.7	1.8	2.8	53
88VA-27*	5.5	8.6	18	32	2.3	1.3	2.8	38
88VA-28	5.7	9.0	16	85	14	4.7	5.4	120
88VA-30	5.8	9.4	18	63	7.9	3.0	4.9	94
88VA-31*	5.6	8.0	16	63	6.4	1.7	3.4	78
88VA-32	4.6	11.4	16	27	1.6	0.3	1.2	16
88VA-33	5.8	4.9	14	35	4.9	0.7	2.7	45
88VA-34	5.6	8.2	18	52	7.7	1.8	3.5	66
90VA-1	5.9	9.8	17	44	8.6	0.9	1.7	58
90VA-2*	6.7	4.3	16	43	15.	5.3	10.	166
90VA-4	3.8	9.2	16	48	1.6	2.6	5.0	52
90VA-6*	7.2	1.9	15	51	25	7.4	25.	246
90VA-16	6.6	8.2	16	54	31.	0.8	2.0	158
Mean	5.9	6.3	16.6	60.1	11.	3.1	7.1	110.7
Std. Dev.	0.8	3.2	1.2	22.3	7.7	2.5	6.3	62.9

Sample No.	K	HCO_3	SO_4	Cl	NO_3	F	Si	Depth
88VA-3	1	48.	ND	3.9	0.9	0.2	19.	100
88VA-5	2	90.	7.4	1.5	ND	0.1	19.	91
88VA-11	1	110	2.3	5.3	ND	0.2	20.	110
88VA-15	2	51.	0.6	2.6	4.6	ND	24.	UNK
88VA-18	1	31.	2.0	7.3	3.8	ND	18.	9
88VA-19	1	31.	3.8	58.	3.1	ND	18.	8
88VA-22	2	54.	1.1	1.1	0.6	ND	5.6	9
88VA-24	2	57.	12.	1.6	ND	0.4	23.	UNK
88VA-25	4	98.	3.6	1.6	0.1	0.2	22.	122
88VA-26	1	29.	0.2	1.6	1.9	ND	10.	11
88VA-27*	2	15.	ND	3.8	3.3	0.1	6.9	43
88VA-28	1	75.	0.2	3.2	0.2	ND	17.	20
88VA-30	1	46.	0.7	1.8	13.9	ND	15.	111
88VA-31*	1	41.	0.3	6.0	12.9	ND	5	12
88VA-32	1	7.2	0.4	1.4	ND	ND	2.6	UNK
88VA-33	1	25.	0.2	1.5	ND	ND	9.2	UNK
88VA-34	1	38.	0.5	1.6	0.7	ND	11.	UNK
90VA-1	1.1	32	ND	5.7	4.2	ND	3.8	9
90VA-2*	3.4	110	6.4	2.3	ND	ND	14.	61
90VA-4	2	4.6	1.2	5.4	30	ND	4.6	5
90VA-6*	3.7	120	30.4	21.	ND	ND	14.	30
90VA-16	3.3	110	1.3	3.4	ND	ND	6.4	10
Mean	1.75	55.5	3.57	6.46	4.22	0.07	13.0	
Std. Dev.	1.00	35.7	6.88	12.3	7.45	0.12	6.80	

Table 2. Analytical results for radionuclides and trace elements in ground water from the Hylas, Virginia, study area.

[Concentration units are as follows: Rn (pCi/L); U, B, Ba, Cr, Cu, Li, Mn, Sr, Ti, Zn (ppb); Al, Fe (ppm). NA, not analyzed; ND, not detected. Other elements sought but below the limit of detection (in ppb, shown in parentheses after element name) were Ag (2); Be (1), except 88VA-19, 2 ppb; Bi (10); Cd (1), except 90VA-9, 1 ppb; Co (3), except 88VA-4, 3 ppb, and 88VA-19, 4 ppb; Ga (5); Mo (10); Ni (5), except 88VA-19, 45 ppb, and 90VA-4, 5 ppb; Pb (10) except 88VA-26, 10 ppb; Sn (6); V (6); and Zr (1), except 88VA-16, 2 ppb]

SAMPLES OF GROUND WATER FROM GRANITES

Sample No.	Rn	U	Al	B	Ba	Cr	Cu
88VA-7	8900	0.3	<0.1	<10	18	<1	310
88VA-23	81000	4.4	<0.1	<10	37	<1	120
90VA-5	740	NA	0.23	<10	15	<1	17
90VA-7	16000	<0.05	<0.1	<10	62	<1	<10
90VA-8	5900	<0.05	<0.1	<10	3	<1	<10
90VA-9	1200	NA	0.11	<10	48	<1	<10
90VA-10	1600	<0.05	0.11	<10	44	<1	16
90VA-11	28000	0.06	<0.1	<10	82	<1	18
90VA-12	1600	<0.05	<0.1	<10	40	<1	25
90VA-13	30000	<0.05	<0.1	<10	12	<1	<10
90VA-14	390	<0.05	<0.1	<10	36	<1	13
90VA-15	2800	<0.05	<0.1	<10	19	<1	<10
Mean	14900	0.5	0.15		35.		74.
Std. Dev.	23400	1.4	0.07		23.		110

Sample No.	Fe	Li	Mn	Si	Sr	Ti	Zn
88VA-7	<0.05	12	21	18.	49.	<1	9
88VA-23	<0.05	16	13	25.	55.	4	7
90VA-5	<0.05	14	1.7	6.1	160	6.3	9
90VA-7	<0.05	<4	3.1	15.	35.	6.1	7
90VA-8	1.05	51	40.	18.	11	4.9	1000
90VA-9	3.3	<4	120	4.7	69	4.9	74
90VA-10	0.08	<4	4.5	6.0	8.3	6.1	16
90VA-11	0.08	<4	2.4	15.	59.	4.8	11
90VA-12	0.08	<4	8.7	12.	57.	6.2	6
90VA-13	<0.05	16	14	21.	40.	4.9	11
90VA-14	0.08	5	12.	14	30.	4.8	9
90VA-15	0.32	46	140	21.	120	4.8	<3
Mean	0.71	23.	32.	15.	58.	5.3	110
Std. Dev.	1.19	18.	48.	6.4	43.	0.77	310

RESULTS

GROUND-WATER QUALITY

Ground water in the Hylas study area is generally of good quality for domestic consumption. The U.S. Environmental Protection Agency (EPA) standard for drinking water is exceeded only by dissolved Fe (8 samples) and Mn (15 samples); for other dissolved components for which standards exist or are proposed, the concentrations are below the recommended value (U.S. Environmental Protection Agency, 1976). (^{222}Rn, however, exceeds the proposed standard in nearly all samples; this is discussed below). The maximum total dissolved solids (TDS) is 285 ppm (parts per million) and the maximum total hardness (Ca + Mg) is 50 ppm (table 1). The pH range in ground water from mylonites is 5.6-7.2, pH in the granite-hosted ground water ranges from 4.4 to 7.3, and pH in ground water from metamorphic rocks ranges from 3.8 to 7.2. The pH range for drinking water recommended by EPA is 6.5-8.5. Corrosion of pipes and staining of porcelain fixtures may result from low (acidic) pH; both were observed in some water delivery systems.

MAJOR IONS AND ELEMENTS

The concentrations of major dissolved ions and elements are summarized in table 1. The water samples are dominated by Ca-Mg/HCO_3 types. As seen in the summary

Table 2. Analytical results for radionuclides and trace elements in ground water from the Hylas, Virginia, study area.—Continued

SAMPLES OF GROUND WATER FROM MYLONITES

Sample No.	Rn	U	Al	B	Ba	Cr	Cu
88VA-1	12000	0.24	<0.1	<10	50	<1	<10
88VA-2	32000	0.58	<0.1	<10	10	<1	50
88VA-4	3700	0.34	<0.1	<10	110	<1	10
88VA-6	4700	0.2	<0.1	<10	47	<1	30
88VA-8	2400	2.9	<0.1	<10	7	<1	<10
88VA-9	600	0.47	<0.1	<10	4	<1	<10
88VA-10	460	0.14	<0.1	<10	36	2	40
88VA-12	2400	0.21	<0.1	<10	28	<1	<10
88VA-13	850	0.42	<0.1	<10	72	1	20
88VA-14	3400	0.07	<0.1	<10	13	<1	<10
88VA-16	11300	<0.05	<0.1	<10	93	<1	<10
88VA-17	1900	0.34	<0.1	<10	3	<1	<10
88VA-20	360	0.13	<0.1	<10	53	<1	<10
88VA-21	1600	0.28	<0.1	<10	75	<1	<10
88VA-29	20000	0.98	<0.1	<10	6	<1	50
88VA-35	2500	2.29	<0.1	10	77	<1	<10
88VA-36	750	0.63	<0.1	10	14	<1	<10
90VA-3	3900	0.05	<0.1	<10	7	<1	<10
Mean	5800	0.6			39.		
Std. Dev.	8300	0.8			35.		

Sample No.	Fe	Li	Mn	Si	Sr	Ti	Zn
88VA-1	0.59	6	120	16.	83.	<1	20
88VA-2	<0.05	18	5	20.	66	4	18
88VA-4	0.08	<4	67	15.	83.	3	91
88VA-6	<0.05	<4	5	12.	25.	<1	13
88VA-8	0.09	16	120	13.	95.	4	14
88VA-9	<0.05	56	160	23.	94.	<1	19
88VA-10	<0.05	<4	24	6.7	12.	6	11
88VA-12	1.38	39	140	24	120	7	9
88VA-13	<0.05	12	1	19	170	7	21
88VA-14	2.34	60	370	23.	120	6	32
88VA-16	<0.05	7	3	18.	140	6	15
88VA-17	<0.05	7	4	19.	39.	7	25
88VA-20	<0.05	10	2	28.	72.	4	5
88VA-21	<0.05	28	5	21.	240	7	7
88VA-29	0.06	62	55	27.	54	5	6
88VA-35	0.2	37	100	20.	300	7	230
88VA-36	<0.05	6	43	11.	90.	6	2200
90VA-3	0.1	46	230	21.	80.	3.6	220
Mean	0.61	27.	80.	19.	105	5.5	170
Std. Dev.	0.83	21.	99.	5.7	73.	1.4	520

Piper diagram (fig. 2), the fields encompassing ground-water compositions of each rock type overlap significantly. The lobes that extend from the main group of water compositions on the Piper plot are due to six outlying samples—two from metamorphic rock-hosted ground water and four from granite-hosted ground water. The sample with the second-highest

Table 2. Analytical results for radionuclides and trace elements in ground water from the Hylas, Virginia, study area.—Continued

SAMPLES OF GROUND WATER FROM METAMORPHIC ROCKS (* = AMPHIBOLITE)

Sample No.	Rn	U	Al	B	Ba	Cr	Cu
88VA-3	1400	0.2	<0.1	<10	7	2	<10
88VA-5	1000	0.3	<0.1	<10	7	<1	<10
88VA-11	2300	0.3	<0.1	<10	3	<1	<10
88VA-15	800	0.1	<0.1	<10	35	1	<10
88VA-18	170	0.1	<0.1	<10	51	1	100
88VA-19	320	0.2	<0.1	<10	130	12	10
88VA-22	200	0.1	<0.1	<10	46	1	<10
88VA-24	320	0.3	<0.1	<10	36	<1	<10
88VA-25	510	<0.05	<0.1	30	8	<1	<10
88VA-26	270	0.2	<0.1	<10	30	2	140
88VA-27*	450	0.1	<0.1	<10	30	<1	50
88VA-28	230	0.1	<0.1	<10	26	2	70
88VA-30	1300	0.1	<0.1	<10	9	1	<10
88VA-31*	200	0.1	<0.1	<10	46	<1	20
88VA-32	190	0.1	<0.1	<10	6	<1	<10
88VA-33	470	<0.05	<0.1	<10	26	1	10
88VA-34	740	0.1	<0.1	<10	18	1	10
90VA-1	350	<0.05	0.14	<10	59	<1	19
90VA-2*	390	<0.05	<0.1	<10	52	<1	<10
90VA-4	80	NA	<0.1	<10	160	<1	854
90VA-6*	300	<0.05	<0.1	12	7	<1	<10
90VA-16	90	0.05	0.16	<10	46	<1	18
Mean	550	0.13			38.		120
Std. Dev.	530	0.08			39.		250

Sample No.	Fe	Li	Mn	Si	Sr	Ti	Zn
88VA-3	<0.05	<4	<1	19.	91.	3	7
88VA-5	<0.05	<4	59	19.	51.	6	3
88VA-11	<0.05	<4	11	20.	71.	4	770
88VA-15	<0.05	<4	2	24.	43.	6	6
88VA-18	<0.05	<4	2	18.	44.	6	7
88VA-19	<0.05	5	31	18.	85.	6	7
88VA-22	<0.05	<4	7	5.6	33.	6	<3
88VA-24	3.0	<4	69	23.	70.	3	6
88VA-25	0.8	<4	54	22.	47.	4	140
88VA-26	<0.05	<4	2	10.	21.	7	12
88VA-27*	<0.05	<4	3	6.9	25.	3	61
88VA-28	<0.05	<4	1	17.	58.	5	<3
88VA-30	<0.05	<4	2	15.	41	4	13
88VA-31*	<0.05	<4	12	5.0	22.	<1	100
88VA-32	<0.05	<4	3	2.6	6.3	3	4
88VA-33	0.08	<4	6	9.2	30.	3	360
88VA-34	<0.05	<4	1	11.	37.	6	6
90VA-1	<0.05	<4	15	3.8	73.	11.	20
90VA-2*	0.06	6	57	14.	46.	6.4	<3
90VA-4	0.16	<4	43	4.6	22.	4.8	44
90VA-6*	0.12	5	94	14.	79.	3.6	<3
90VA-16	<0.05	4	9.6	6.4	67.	2	4
Mean	0.71		23.	13.	48.	4.9	88.
Std. Dev.	1.2		28.	6.8	23.	2.0	190

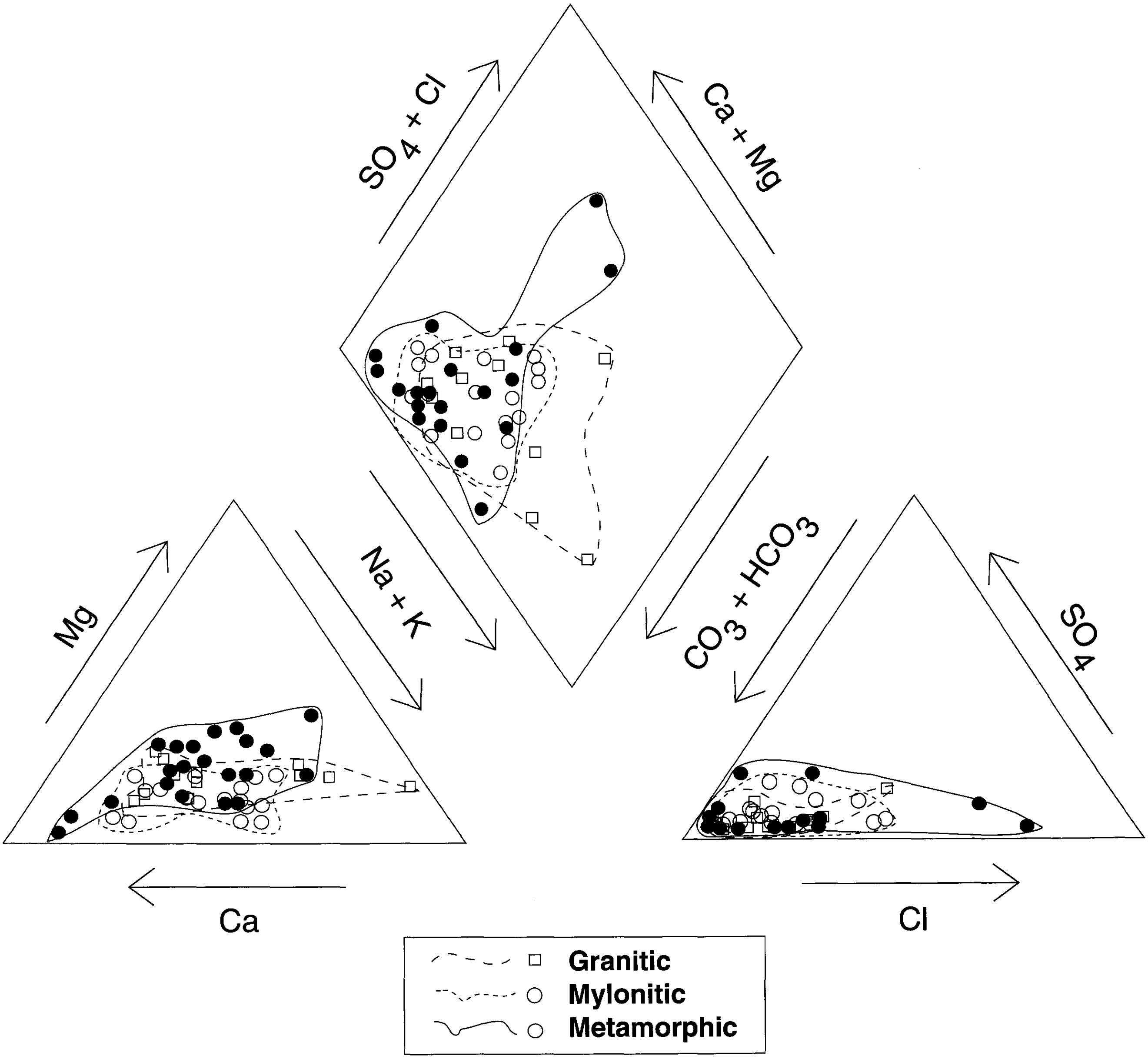


Figure 2. Trilinear (Piper) diagram for ground-water samples from Petersburg Granite, shear zone mylonite, and metamorphic rocks (including amphibolites within the Maidens Gneiss) of the Hylas study area. Lines show approximate fields of water composition for the three rock types.

chloride content from the granite-hosted waters (88VA-23; Cl=12 ppm) is located near a rock quarry that has been active for several decades. It is possible that the prolonged effects of quarrying have led to an alteration, although not necessarily a deterioration, of water quality in the immediate area. Several of the other "outlying" samples, including the one near the quarry, have high NO_3 concentrations, suggesting leakage from septic systems or input from fertilizers.

Ground-water chemistry throughout the study area suggests a limited degree of water-rock interaction. Some wells may sample more than one lithology, but the similarity of water composition from all wells suggests that minerals controlling the chemical composition of the waters (quartz, feldspar) are present in all rock types. Results of chemical modelling using the program PHREEQE (Parkhurst and others, 1980) show that the only major rock-forming mineral to approach saturation in these waters is quartz. Because most aluminum values were below detection, the saturation state was not calculated for common aluminosilicates that might be expected to form (clays, feldspars). Generally, the major mineral classes for which saturation states were calculable were carbonates, sulfates, oxides, and oxyhydroxides.

Ground water in the study area is dilute (defined as having TDS of less than 500 ppm), indicating that it has not had a long history of contact with the rocks. A high recharge rate may also be responsible for the low TDS. Because TDS is strongly controlled by the calcium and magnesium concentrations ($r^2 = 0.76$ for TDS vs Ca+Mg), this indicates that the water has not evolved chemically to any great degree. Ca-Mg/HCO_3 (fresh) waters are dominant in the earliest stages of ground-water evolution but are altered over time to waters having much higher TDS values and containing primarily Na, K, Cl, and SO_4 (Davis and DeWiest, 1966).

TRACE ELEMENTS

Data for dissolved trace elements are summarized in table 2. Abundances of most dissolved trace elements vary only slightly with host lithology, and as noted, statistical analyses were not performed when qualified values were present in more than 10 percent of the samples. Trace elements below detection in every sample include Ag, Bi, Ga, Mo, Sn, and V. Comparative statistics were not computed for the following nine trace elements because of insufficient numbers of samples above detection: Be, Cd, Pb, and Zr were above detection in only one sample each (four separate samples); Co and Ni were found in only 2 samples; Al and B were present in detectable amounts in only 4 samples; Cr was detected in no granite-hosted samples and in only 2 mylonite-hosted samples.

DISSOLVED URANIUM AND RADON ABUNDANCES

Results of radionuclide analyses are also summarized in table 2. Average ^{222}Rn values for ground waters from the Petersburg Granite and mylonite of the Hylas shear zone are 14,900 and 5,800 pCi/L (picocuries per liter), respectively (table 2; fig. 3). ^{222}Rn ranges from 360 to 32,000 pCi/L in water samples from mylonite and from 400 to 81,000 pCi/L in water samples from granite. Thus, all ground waters from granitic and mylonitic samples have ^{222}Rn values above the recently proposed EPA maximum contaminant level (MCL) for drinking water of 300 pCi/L (U.S. Environmental Protection Agency, 1991). Significantly lower ^{222}Rn values are associated with the Maidens or Eastern gneiss complex. The average value for ^{222}Rn in ground-water samples from the metamorphic rocks is 550 pCi/L, with a maximum concentration of 2,300 pCi/L. The 8 water samples with ^{222}Rn concentrations below the proposed MCL of 300 pCi/L are all from metamorphic rocks. The cumulative frequency plot of figure 3 illustrates that the range of ^{222}Rn concentrations in water samples from mylonitic and granitic units are similar but much higher compared to the metamorphic units. The median ^{222}Rn value (50th percentile in figure 3) in water samples from mylonite is 2,500 pCi/L and 4,400 pCi/L for water samples from granite, but in waters from metamorphic units, the median ^{222}Rn value is only 330 pCi/L.

Aqueous U concentrations from all samples are generally above the lower limit of detection (0.05 ppb) and as high as 4 ppb (table 2). Virtually all of the water samples contain measurable dissolved oxygen and can therefore be considered to be oxidizing. The dominant form of dissolved U under these conditions is U(VI), as the uranyl ion, UO_2^{2+}. Uranyl forms a series of strong complexes with carbonate, represented by the general formula $UO_2(CO_3)_n^{2-2n}$. As shown in figure 4, ground waters from mylonitic rocks generally have higher ΣCO_3 compared to samples from granitic and metamorphic rocks. The formation of uranyl-carbonate complexes stabilizes U in solution and inhibits UO_2 adsorption onto mineral surfaces (Wanty and others, 1991a).

Average U concentrations are higher in ground water from mylonite (0.6 ppb) and granite (0.5 ppb) compared to metamorphic rocks (0.1 ppb), similar to the results for ^{222}Rn. However, no correlation exists between dissolved U and dissolved ^{222}Rn activities in the water samples ($r^2 = 0.024$, fig. 5). A primary drinking water standard for dissolved U of 20 ppb U (30 pCi/L) has been recently proposed by the EPA (U.S. Environmental Protection Agency, 1991); all ground waters that were sampled are well below the 20 ppb level. In addition to the proposed standards for aqueous ^{222}Rn and U, the EPA has proposed an MCL for ^{226}Ra in drinking water

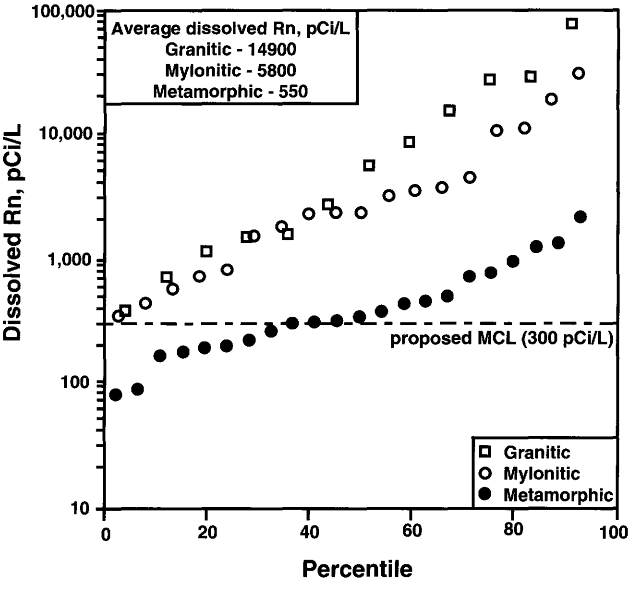


Figure 3. Cumulative frequency plot of ^{222}Rn concentrations in ground-water samples from granitic, mylonitic, and metamorphic rocks in the Hylas area. Average and median values (50th percentile) respectively, are 14,900 and 4,400 pCi/L from granitic rocks, 5,800 and 2,500 pCi/L from mylonitic rocks, and 550 and 330 pCi/L from metamorphic rocks. Dashed horizontal line indicates the U.S. Environmental Protection Agency's proposed maximum contaminant level (MCL) of 300 pCi/L as dissolved ^{222}Rn.

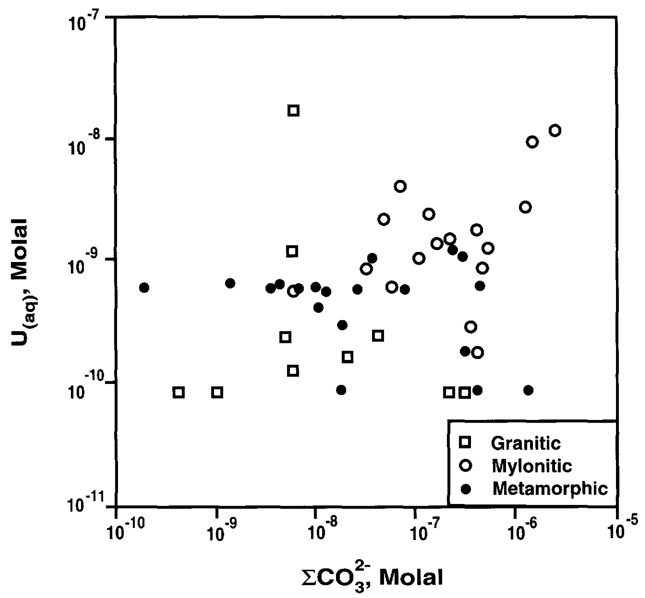


Figure 4. Total dissolved uranium (measured) versus concentration of ΣCO_3^{2-} (calculated by PHREEQE) in ground water samples from granitic, mylonitic, and metamorphic rocks in the Hylas area.

of 20 pCi/L (U.S. Environmental Protection Agency, 1991). Although ^{226}Ra concentrations were not measured in the Hylas water samples, most values reported for ^{226}Ra in ground water are on the order of 10^{-2} to 10^{1} pCi/L (Wanty and Gundersen, 1991, and references therein). Below pH values of approximately 5.5, aqueous ^{226}Ra concentrations may be higher than the upper limit for the range listed above (Szabo and Zapecza, 1987; Zapecza and Szabo, 1987; Cecil and others, 1987), but as discussed later, it is unlikely that the standard for aqueous ^{226}Ra would be exceeded in most samples from the Hylas area.

DISCUSSION

COMPARISON OF GROUND-WATER CHEMISTRY TO WELL TYPE OR DEPTH

Samples from wells within both saprolite and crystalline rocks generally showed variable and broad ranges of dissolved constituents; no systematic relationships of ground-water chemistry and radionuclide content dependent on well depth or well type were observed. For example, TDS varies widely regardless of well depth or type. In ground waters from crystalline rocks, ^{222}Rn in metamorphic units does show an increase with well depth, but only below the saprolite zone (r^2=0.64); no depth-related trends in ^{222}Rn concentrations (or other parameters), either within or beneath the saprolite, were observed in water samples from mylonite (r^2=0.02) or granite (r^2=0.03). Nonetheless, some differences in ground-water chemistry were observed; the remainder of this report examines some possible reasons for these differences in ground waters from the 3 major rock types in the Hylas area.

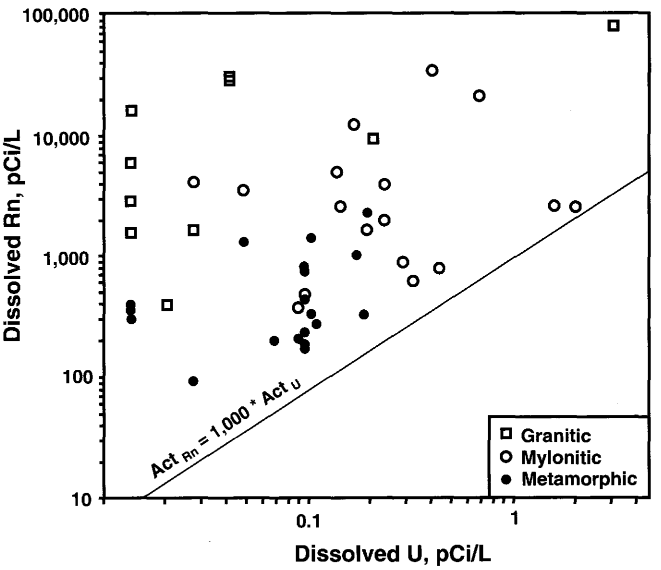


Figure 5. Radioactivities (in pCi/L) of dissolved U versus dissolved ^{222}Rn in ground water. The line drawn across the lower right corner of the diagram represents radioactivity of ^{222}Rn that is approximately 1,000 times that of uranium (assuming secular equilibrium ^{238}U and ^{234}U; U activity calculated using the conversion 1 ppb U = 0.68 pCi). All samples plot above this line, indicating that at least 99.9% of the dissolved ^{222}Rn is unsuppoprted by dissolved U.

COMPARISON OF GROUND-WATER CHEMISTRY OF SAPROLITE AND BEDROCK

When all 52 ground-water samples are examined solely on the basis of whether they originate from saprolite or bedrock (but without regard to the type of bedrock), only average DO and Cl were significantly higher in the saprolite than in the bedrock. No significance can be attached to Cl because of the conservative nature of this anion. The higher DO in saprolite undoubtedly reflects its close proximity to the atmosphere, where oxygenated meteoric waters can readily infiltrate the saprolite and continually replenish O_2 that may be consumed by near-surface weathering reactions. Dissolved O_2 in ground water in the crystalline units tends to decrease with increasing depth, although DO was detected in every sample. Lower DO in the deeper crystalline units may result from multiple causes: (1) O_2 does not reach these depths because of slow recharge or recharge with poorly-oxygenated meteoric waters, (2) O_2 is rapidly consumed by weathering reactions as oxygenated recharge water percolates through the saprolite, or (3) O_2 carried in ground water reaches lower depths but is quickly used up by weathering reactions within the crystalline rocks. It is likely that all three processes are operating in the ground-water system, but the relative magnitude or importance of each process is unknown.

In comparing saprolite to its underlying parent bedrock (and distinguishing among bedrock types), some geochemical differences in ground waters are more noticeable, particularly for waters from the mylonitic and metamorphic rocks. It is assumed that the saprolite was derived from the crystalline rocks directly beneath the saprolite. Because the area is tectonically stable and has low relief, it is less likely that geological processes have caused large vertical or horizontal displacements of the surficial layers away from the parent bedrock.

Granite-hosted ground waters showed the fewest number of geochemical differences; only SpC, ^{222}Rn, Si (as SiO_2), Mg, and NO_3 were significantly higher in the unweathered granitic rock compared to the weathered saprolite. The higher concentration of ^{222}Rn in bedrock ground waters, even though $U_{(aq)}$ shows no differences in ground water from rock or saprolite, suggests that U is more strongly retained by mineral surfaces in the bedrock. Alternatively, these results may merely be an indication of the higher abundance of U in the bedrock; as noted later, however, U within minerals would have to be ideally sited at the rock-water interface to support the observed ^{222}Rn abundances. Similarly, higher Si and Mg may merely reflect the higher abundances of these elements in granitic rocks, or that crystalline rock weathering is proceeding to a greater degree than in the saprolite. Which of these 2 possibilities accounts for the observed Si and Mg concentrations could not be determined with the available data. Higher NO_3 in waters from the deeper crystalline rock is difficult to explain, but it may be that some surface NO_3 does indeed infiltrate to deeper ground waters, whereas the bulk of NO_3 is consumed by biological processes in the shallow saprolite.

Ca, Mg, Na, HCO_3, SO_4, TDS, U, and Hardness (=Ca+Mg) were significantly higher in waters from the mylonitic bedrock than in the saprolite derived from this rock. These parameters, especially the higher TDS and major cations, and the lower DO of the crystalline rocks, suggest that the mylonitic rocks may be undergoing weathering with release of cations to a greater degree than the granitic rocks. NO_3 and Ba contents were significantly lower in the crystalline rock, and all other parameters showed no differences between saprolite and bedrock. The high U may be indicative of mobile dissolved U, which is not held as tightly on mineral surfaces as in the other two rock types.

Samples from metamorphic-hosted ground waters exhibited the greatest number of differences in saprolitic versus bedrock ground waters. The pH, SpC, ^{222}Rn, K, HCO_3, TDS, Mn, Si, Sr, Na, SO_4, and Hardness are all higher in the bedrock; only DO is higher in the saprolite. Again, the dissolved elements could result from ferromagnesian and silicate mineral weathering (K, Mn, Si, Sr, Na), while the higher SpC, TDS, HCO_3, SO_4, and Hardness simply reflect increased abundances of dissolved species from the weathering process. $U_{(aq)}$ is probably adsorbed more strongly to mineral surfaces within the metamorphic rocks than in the saprolite derived from these rocks, as indicated by the observed ^{222}Rn being higher in ground waters from bedrock, while $U_{(aq)}$ shows no differences in waters from either bedrock or saprolite.

STATISTICAL COMPARISON OF WATER CHEMISTRIES

MAJOR IONS AND ELEMENTS

Bicarbonate (HCO_3) is the dominant anion in most ground-water samples; its significance in the observed U and ^{222}Rn abundances is discussed in a later section. Average sulfate (SO_4) concentrations are similar for ground-water samples from granitic and metamorphic samples (3.5 ppm) and slightly higher in the mylonitic samples (4.9 ppm), but these SO_4 values are indistinguishable at the 95% confidence level. Sulfate can be produced by oxidative weathering of sulfide minerals, such as pyrite (FeS_2), that have been noted in some of the aquifer rocks (Ellison and Masiello, 1979). Although no relationship of dissolved Fe and SO_4 was observed ($r^2 = 0.01$), weathering of pyrite and mafic minerals is a potential source of dissolved and solid forms of iron in the aquifers. Average chloride (Cl) in ground waters from the 3 rock types does not exceed 8 ppm; the maximum abundance of Cl was 58 ppm and only one other sample had Cl higher than 20 ppm. Phosphate (PO_4) and fluoride (F) form complexes with U and could potentially enhance the abundance of soluble U (Langmuir, 1978); however, PO_4 is below the detection limit (<0.1 ppm) in all samples and F is below the detection limit (<0.1 ppm) in 31 samples; no statistics were computed for PO_4 or F. The Environmental Protection Agency's MCL of 45 ppm as nitrate (NO_3) was not exceeded in any sample. NO_3 was not detected in 20 samples, and only 8 samples have NO_3 concentrations greater than 10 ppm. Samples with the highest NO_3 values are from areas where fertilizers are used, such as golf courses or agricultural fields.

Major dissolved species in ground water are Si (SiO_2), Ca, Mg, and Na, but the abundance of dissolved K is generally low (1-2 ppm) despite the fact that alkali feldspar is a component of most aquifer rocks; this lack of dissolved K probably reflects the formation of K-bearing weathering products, such as clay minerals, or the resistance of the feldspars to weathering (Busenberg and Clemency, 1976). Ca, Mg, and Na are major constituents of aquifer minerals, such as hornblende, pyroxene, and plagioclase feldspars, and their dominance in the aqueous phase is consistent with the weathering of crystalline silicate rocks in a humid environment (Garrels and MacKenzie, 1967). Dissolved Ca, Na, and

K are not statistically different among the three ground-water types; only dissolved Mg showed some variation. Mg was lower in the granite-hosted waters (1.2 ppm) but the same in metamorphic- and mylonitic-hosted waters (3.7 ppm). On a mass basis, the sum of the major cations (Ca, Mg, Na, and K) comprises up to 26 percent of TDS in some samples while the major anion HCO_3 constitutes as much as 70 percent of TDS in other samples.

Average dissolved Si values are 19 ppm from mylonite, 15 ppm from granite, and 13 ppm from metamorphics; the metamorphic units are significantly lower in Si compared to the mylonite. The higher Si concentrations from mylonite samples suggests more extensive weathering or alteration of silicate minerals, but the extent of weathering or alteration and types of minerals involved is unknown. Si may also be controlled or limited by mineral precipitation reactions, for example, the formation of clays or other secondary silicates (Garrels and MacKenzie, 1967). Ion ratios that have been used as indicators of weathering reactions (for example, Si/TDS or Na/total cations; see Hem, 1989, and Garrels and MacKenzie, 1967) did not provide any insight into the types of minerals undergoing weathering or processes that might be involved.

TRACE ELEMENTS

Quantitative comparisons were made for the following trace elements that were above detection in 90 percent of the ground-water samples: Ba, Mn, Sr, Ti, and Zn. However, the mean values of Ba, Ti, and Zn are not significantly different among ground waters from the three rock types. It should be noted that the Zn and Cu detected in some samples could arise from solubilization of these metals by acidic waters from galvanized or copper pipe that is commonly used in well-water delivery systems.

Average dissolved Mn concentrations are 80.3 ppb from mylonite, 31.8 ppb from granite, and 23 ppb from metamorphics. The metamorphic units are significantly lower in Mn compared to the mylonite. Mn often behaves chemically-similar to Fe in that both are generally more soluble in reduced form and less soluble in oxidized form, but the correlation of these two elements was weak ($r^2 = 0.27$). A weak negative correlation of higher dissolved Mn versus lower dissolved O^2 was observed in all water samples ($r^2 = 0.21$); this trend was slightly stronger in waters from metamorphic ($r^2 = 0.36$) and granitic ($r^2 = 0.36$) rocks. The increase in dissolved Mn concentrations with higher dissolved O_2 values seems inconsistent with the known geochemistry of Mn and could be due to mixing of waters from different aquifers. Nonetheless, the higher Mn concentrations from mylonite-hosted water samples suggest more extensive weathering or alteration of Fe- and Mn-bearing silicate minerals in this unit, but as noted with silica, the possible weathering mechanisms are not known.

Sr is significantly higher in water samples from both mylonite (105 ppb) and granite (58 ppb) compared to the metamorphics (48 ppb). The most likely explanation for the observed values is that Sr is more abundant in granitic rocks and less abundant in basic (ultramafic) crystalline rocks (Turekian and Wedepohl, 1961); weathering and dissolution of plagioclase feldspars, apatite, or calcite, where Sr readily substitutes for Ca, could lead to higher dissolved Sr.

Although Li was detected in only 4 water samples from metamorphic rocks, it was detected in 7 granite-hosted and 15 mylonite-hosted water samples. The presence of detectable Li in waters from granites and mylonites, like Sr, probably reflects a greater original abundance of this element in micaceous minerals from granitic or volcanic rocks (Hem, 1989) that were precursors to mylonite.

The distribution of the 20 water samples that contain measurable dissolved iron is approximately equal among all three rock types. Average Fe concentrations in filtered water samples from mylonite (0.27 ppb), granite (0.42 ppb), and metamorphics (0.2 ppb) are not significantly different. Six freshly collected water samples from the mylonitic rocks and three water samples from the metamorphic rocks contained suspended rust-colored particles, assumed to be amorphous iron oxyhydroxides (and hereafter referred to as ferric oxides or iron oxides); water samples from granitic rocks contained no visible iron oxide. The presence of suspended iron oxides does not correlate with dissolved oxygen ($r^2 = 0.04$) or dissolved iron concentrations ($r^2 = 0.1$). An increase in dissolved iron with decreasing pH was observed in those samples with iron concentrations above the detection limit. Consistent with the known geochemistry of iron, a rise in pH and aeration during sampling would favor oxidation of dissolved Fe(II) to form insoluble Fe(III) oxides. Alternatively, the wells may sample several of the water-bearing horizons (as noted in some drilling logs) with different redox potentials and dissolved Fe and O_2 concentrations. Mixing of these waters may produce changes in water chemistry that lead to in situ iron oxide precipitation (Hem, 1989). Although the presence of some iron oxides may be due to corrosion (formation of soluble Fe^{2+}) of steel well casings, iron oxides were also observed in samples from the augered wells lined with terra cotta tiles. A newly drilled flowing well (in operation 1 week; 120 meters deep) within the Maidens Gneiss had deposited a 1-2 mm thick layer of fresh iron oxide precipitate on the ground surface, suggesting that mobile Fe^{2+} is present in the waters tapped by this well. In the active quarries in the area, abundant iron oxides were observed on fresh rock surfaces and throughout the overlying saprolite. Additionally, autoradioluxographs of shear zone mylonites show that abundant ferric oxides are located along shear planes and that radioactive elements are concentrated within these ferric oxides (L.C. Gundersen, oral commun., 1992). These observations suggest, but do not necessarily confirm, that iron oxides may be suspended in ground waters or are precipitating in these rocks. The presence of abundant iron oxide sorbents could be important in limiting the concentrations of dissolved trace elements and radionuclides.

DISSOLVED URANIUM AND RADON

Although the major and minor element chemistry of ground water in the study area does not differ markedly from one rock type to another, analysis of variance suggests that there are differences with respect to key geochemical parameters that may influence ^{222}Rn and U abundances and mobilities in the ground waters. These parameters (pH, HCO_3, DO, and TDS) indicate that ground waters from the mylonitic, granitic, and metamorphic rocks are sufficiently different to account for some of the variation in the U and ^{222}Rn abundances and distributions.

Ground water from mylonite within the Hylas zone is characterized by a narrower range of pH (5.6-7.2), higher average pH (6.4), and higher HCO_3 (average=99.3 ppm). These values are significantly different (95% confidence level) compared to values from the Petersburg Granite and metamorphic units (table 1).

The correlation of (calculated) total carbonate (ΣCO^3) with dissolved U in mylonite ground waters seems quite strong (r^2= 0.7, fig. 4) but should be viewed with caution due to the few data points at higher values. The average U value from the mylonite ground waters is not significantly different from the granite-hosted group of samples. However, both are higher in U (0.5 ppb) than ground waters from the metamorphic rocks (0.1 ppb). Most samples analyzed for dissolved U from Petersburg Granite were below detection, and the high average value is primarily caused by sample 88VA-23 (4.4 ppb). As mentioned earlier, this high U sample is located next to an active quarry near the contact of granite and mylonite and may be influenced by anthropogenic activities (fracturing by explosives) to a larger extent than other samples. This sample also has the highest measured ^{222}Rn value (81,000 pCi/L) and is anomalous in many other ways (high Cl and NO_3, see earlier discussion of Piper diagram). If 88VA-23 is excluded from statistical calculations, the average U from granite-hosted water samples is only 0.1 ppb, the same as water samples from the metamorphic rocks. (Similarly, average ^{222}Rn from granite-hosted water drops to 8,800 pCi/L, still higher than the mylonite but almost one-half the value when 88VA-23 is included). However, because all but three uranium values are below detection in the granitic samples, statistical calculations involving U from these samples may not be reliable. In contrast, nearly all mylonite samples show U values well above detection (table 2). The lower pH and lower abundance of aqueous U (but nonetheless high ^{222}Rn values) in the granite-hosted water samples suggest that U and Ra may be adsorbed onto mineral surfaces in these rocks to a greater extent than in the mylonite.

The metamorphic samples are uniformly low in aqueous U and HCO_3, suggesting that low whole-rock U abundance, decreased CO_3 complexation, or adsorption act to produce low aqueous U values. In summary, data for pH, ΣCO_3, and U in mylonite ground water suggest that there is greater potential for CO_3 complexing to stabilize U in solution relative to sorptive mineral surfaces.

Individual water samples from the mylonites generally contain lower DO and higher TDS than samples from the metamorphic and granitic units. The average DO content of samples from mylonite (3.7 ppm) is not statistically different from the granite-hosted samples (4.9 ppm) but is lower than the metamorphic samples (6.3 ppm). Average TDS from mylonite samples (175 ppm) is significantly higher than samples from granites (113 ppm) or metamorphic units (105 ppm). The differences in DO and TDS suggest that the mylonite reacts with ground water to a greater extent or at a faster rate than the other rock units, perhaps because of greater permeability and/or greater surface area available to the ground water. O_2 in ground water is efficiently consumed during oxidative weathering of fine-grained mafic minerals. These reactions promote oxidation of iron and release of other cations and silica. An example of such a reaction is the weathering of pyroxene:

$$\{1\}\quad 2CaFeSi_2O_6 + 1/2O_2 + 10H_2O + 4CO_2 \rightarrow Fe_2O_3 + 4H_4SiO_4 + 2Ca^{2+} + 4CO_3^-$$

Other reactions (such as organic carbon oxidation) may also consume O_2, but the extent of any of these O_2-consuming reactions is not known.

Exclusion of the anomalous granitic water sample 88VA-23 from the statistical calculations produced a significant decrease in average dissolved U in granites (discussed above) and equalized the average dissolved HCO_3 in mylonite and granite. These changes did not greatly alter the conclusions derived from the analysis of variance. Summary statistical comparisons for major ground-water parameters from the three 3 rock types is presented in table 3.

PROCESSES AFFECTING URANIUM CONCENTRATIONS IN GROUND WATER

WHOLE ROCK URANIUM ABUNDANCE AND DISTRIBUTION

Figure 6 compares the average abundance of whole rock eU to the average concentration of ground-water U from four rock types. The minimum amount of U present is 2.2 ppm in amphibolite, the rock type most likely have the lowest U abundance. The eU analyses (Gates and Gundersen, 1992) show only slightly higher average U concentrations for metamorphic rocks of increasingly granitic composition, as with granite gneiss (2.8 ppm); all metamorphic rocks are lower in total U (as eU) than the granitic or mylonitic rocks. A likely explanation for these differences in

Table 3. Statistical comparison of pH, HCO_3, TDS, DO, and dissolved U and ^{222}Rn (log-transformed) in ground water samples from granitic, mylonitic, and metamorphic rocks in the Hylas area, Virginia.

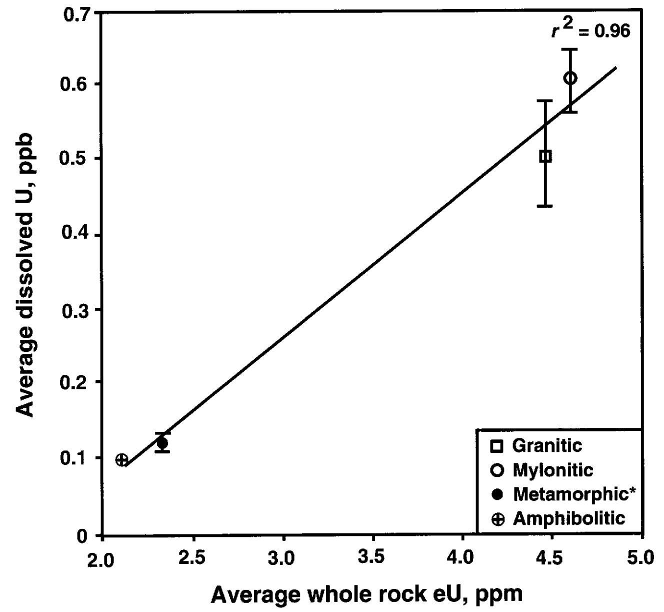
[Mean values compared using Scheffe F-test for one-factor analysis of variance. In general, water samples from mylonitic rocks are significantly different from metamorphic and granitic water samples at the 95% confidence level. See text for detailed discussion of differences in ground water chemistry. (♦) = Significant at 95% confidence level; sample 88VA-23 included. (*) = Significant at the 95% confidence level; sample 88VA-23 not included. (#) = Not significant at the 95% confidence level; sample 88VA-23 not included]

	pH	HCO_3	DO	TDS	U	log ^{222}Rn
MYLONITIC vs METAMORPHIC	♦	♦	♦	♦	♦	♦
MYLONITIC vs GRANITIC	♦	♦,#		♦	*	
METAMORPHIC vs GRANITIC					♦,#	♦

U abundances is that mylonitization is a kinetic metamorphic process that does not involve greatly elevated temperatures or recrystallization (Jackson, 1970); U is not highly mobile under these conditions, so the shear zone mylonites probably did not suffer a significant loss of primary U (U present in precursor rocks) during deformation (Gundersen, 1989). On the other hand, at least some of the Maidens Gneiss units formed under medium-grade (amphibolite facies) metamorphic conditions (Goodwin, 1970). With increasing metamorphic grade, U is less compatible in new mineral lattices and may escape with volatiles (H_2O + CO_2) along pressure release conduits, such as shear zone boundaries; U may ultimately be lost from the precursor rocks (Heier and Adams, 1965; Yermolayev, 1971). Thus, U either is no longer present or is present at reduced levels in minerals of the Maidens Gneiss rocks. U in all rocks is not limited to the parts per billion (ppb) levels found in the ground waters, and the strong correlation of dissolved U with whole rock eU (r^2 = 0.96) in figure 6 initially suggests that the concentration of U in ground water might be controlled by the original abundance of U in the rocks. If an assumption is made that all dissolved U was derived from crystalline-rock U, the percentage of "labile" U from the rocks, calculated as

$$\frac{[\text{average dissolved } U_{(ppb)}]}{[\text{average whole rock } eU_{(ppb)}]} * 100$$

is only 0.005 to 0.01 for all four rock types. Therefore, only a small fraction of total U in the rock is available to the ground water at the rock-water interface. Because dissolved U varies over large ranges for each rock type (fig. 4; table 2), it would be an oversimplification to use whole-rock U abundances as a primary predictor of dissolved U abundances.



(*Metamorphic rocks do not include four rocks of amphibolite composition)

Figure 6. Average dissolved U in ground water versus average whole rock eU (equivalent uranium) in four major rock types in the Hylas area. Error bars are standard deviations for dissolved U analyses. Amphibolites are plotted separately to show the range of whole rock eU and dissolved U for rock compositions ranging from ultamafic to granitic. The group denoted as "metamorphic" does not include the four rocks of amphibolite composition.

The parent radionuclides are available from the aqueous or solid (crystalline and adsorbed) phases within the aquifer units. In figure 7, the graph of dissolved ^{222}Rn versus whole rock U (eU) for the four rock types shows that dissolved ^{222}Rn is not derived wholly from U within the crystalline rocks. Figure 7 also indicates that approximately 60 percent (r^2 = 0.61) of the variation in observed average ^{222}Rn abundances is attributable to variation of parent radionuclides in the whole rock. Meanwhile, figure 5 shows that dissolved ^{222}Rn is also unsupported by dissolved U because U radioactivity is much less than that of its daughter,

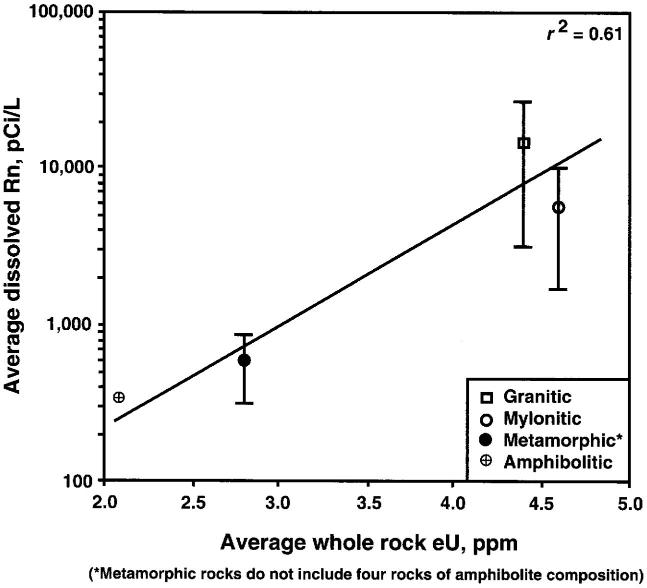


Figure 7. Average dissolved ^{222}Rn in ground water versus average whole rock eU (equivalent uranium) in four major rock types in the Hylas area. Error bars are standard deviations for dissolved ^{222}Rn analyses. Amphibolites are plotted separately to show the range of whole rock eU and dissolved ^{222}Rn for rock compositions ranging from ultamafic to granitic. The group denoted as "metamorphic" does not include the four rocks of amphibolite composition.

^{222}Rn. (An element produced by radioactive decay is said to be unsupported by its parent element if the radioactivity of the parent is less than that of its daughter). Thus, figures 5 and 7 demonstrate that another source of the parent radionuclide(s) in the aquifer must be present. This additional source is the U or Ra (which ultimately decay to produce dissolved ^{222}Rn) adsorbed onto the solid mineral surfaces in the aquifers. Variations of ^{222}Rn in ground water are due to differing contributions of the three sources to the total U or Ra abundances.

WATER CHEMISTRY

Ground-water analyses in tables 1 and 2 were used to calculate saturation indexes (SI) for some of the more common uranium minerals; the SI values are compiled in table 4. The SI of a mineral is defined as the base-10 logarithm of the ion activity product (IAP) of dissolved mineral constituents divided by the equilibrium constant (solubility product, K_{sp}) for the mineral:

$$SI = \log_{10}\frac{IAP}{K_{sp}}$$

A positive SI indicates that mineral precipitation from the solution may occur (supersaturation), while a negative SI indicates mineral dissolution may occur (undersaturation) if the mineral is present. All of these oxygen-bearing groundwater samples were highly undersaturated with respect to the major oxidized U(VI) minerals, usually by many orders of magnitude. Measurable uranium concentrations in ground water from the three rock types are generally limited to the range of 0.1 to 1.0 ppb. These concentrations are far below those predicted by the solubility of insoluble U(VI) minerals, such as schoepite ($UO_2(OH)_2 \cdot H_2O$) or rutherfordine (UO_2CO_3). The observed U concentrations are not limited by the solubility of discrete U minerals; rather, the data suggest that some other process, such as adsorption, plays a role in limiting U concentrations.

Ferric oxide minerals are the most likely sorbents for U because of their observed abundance in the host rocks, high specific surface area, and recognized ability to adsorb a variety of dissolved constituents including U (Hsi, 1981; Wathen, 1987). Ferric oxides are able to adsorb U about 10^5-10^6 times more efficiently than most aluminosilicates (Langmuir, 1978; Wanty and others, 1991b), and amorphous ferric oxides are more effective adsorbents than crystalline forms like hematite. Adsorption of U onto ferric oxides is most complete over the pH range 5-8.5, and increasing CO_3 concentration leads to desorption of the U species; Ca and Mg have little influence on the adsorption process (Hsi and Langmuir, 1985). Ferric oxides are also effective adsorbents for ^{226}Ra, the immediate parent of ^{222}Rn (Ames and others, 1983), although below pH 5.5, ferric oxides begin to dissolve and thus become less effective adsorbents (Szabo and Zapecza, 1987). Nonetheless, a small mass of ferric oxide can be a particularly important adsorbent when it is present as fracture or grain coatings in direct contact with coexisting water. Chemical weathering of ferromagnesian minerals in a humid climate leaches Ca and Mg, but Fe generally remains as an oxide (equation 1); thus, the rocks and residual saprolite are enriched in iron oxides and oxyhydroxides. Shear zones in other regions of the Eastern United States are recognized by their characteristic red staining caused by abundant Fe-oxides and Fe-oxyhydroxides (Gundersen, 1989). Further, ferric oxides collected on the 0.45µm filter suggest that soluble forms of iron and perhaps iron oxides are present in the local aquifers.

PROCESSES AFFECTING RADON CONCENTRATIONS IN GROUND WATER

SECULAR EQUILIBRIUM

^{222}Rn is produced by the α-decay of ^{226}Ra; both are daughters in the ^{238}U decay series. With the passage of time, a condition known as secular equilibrium may be attained in a closed system. Secular equilibrium is reached when the rate of decay of all nuclides in a decay series is equal. Attainment of secular equilibrium occurs independently of the system's tendency to attain chemical equilibrium. In fact, deviations from secular equilibrium may yield valuable clues to active chemical processes, as will be demonstrated. To determine whether secular equilibrium is attained in a system, the radioactivity of all nuclides must be measured and compared. Uranium is easily measured in terms of mass and can be converted to radioactivity units assuming that ^{235}U activity is small and that ^{234}U and ^{238}U are in secular equilibrium. The first assumption has been shown to be generally correct but the second may be a poor one, as ^{234}U:^{238}U α-activity ratios over 10:1 have been observed (Szabo, 1982). However, most water samples have ^{234}U:^{238}U activity ratios of 1 to 2 (Osmond and Cowart, 1976). Even considering the uncertainty in the ^{234}U:^{238}U ratio, the dissolved U α-activities in solution are at least three orders of magnitude less than that required to sustain the observed activities of dissolved ^{222}Rn (fig. 5) and thus indicate a dramatic departure from secular equilibrium between U and ^{222}Rn.

TRANSFER OF RADON FROM AQUIFER ROCKS TO GROUND WATER

The α-decay of ^{226}Ra causes a recoil of the daughter ^{222}Rn according to the laws of conservation of momentum. This α-recoil effect has been described recently by Semkow (1990, and references therein) and is thought to be the principal mechanism for transferring ^{222}Rn from mineral surfaces to coexisting pore water. It is unlikely that ^{222}Rn produced deep within mineral grains could diffuse to mineral surfaces within the short (3.8 days) half-life of ^{222}Rn. Therefore, ^{222}Rn in solution is probably derived from the parent ^{226}Ra (and ^{238}U) located very near the water-rock interface (Torgersen and others, 1990). After entering the aqueous phase, ^{222}Rn is probably not far removed from its original source (U and Ra in the rocks) because of its fast decay rate relative to probable ground-water flow rates.

^{226}Ra concentrations were not measured in this study, but as noted, most reported values for ground water are in the range of 10^{-2} to 10^{1} pCi/L, well below the range of ^{222}Rn concentrations observed in the Hylas ground waters (10^{2}-10^{4} pCi/L). Therefore, ^{222}Rn concentrations in the water are almost certainly unsupported by dissolved ^{226}Ra. Rather, ^{222}Rn is transferred to the solution via α-decay and recoil from ^{226}Ra located in the solid phase. This condition requires that some portion of the parent Ra (and perhaps associated U precursor) be located within an α-recoil distance of the water-rock interface, a distance on the order of hundredths of micrometers (Torgersen and others, 1990).

If efficient transfer of ^{222}Rn from solid to solution occurs at grain surfaces, then processes that increase the specific surface area of the aquifer rock should lead to greater concentrations of dissolved ^{222}Rn, and this has been observed (Torgersen and others, 1990; Wanty and others, 1991b; Wanty and others, 1992). This argument generally applies to all rocks in the study area, and especially to the mylonitized rocks, which probably have a greater specific surface area and a larger number of fractures per unit area than the other rocks (Bobyarchick and Glover, 1979; Gates and Gundersen, 1992), and have ^{222}Rn-enriched ground water.

^{222}Rn values show that water samples from mylonitic and granitic rocks are similar with respect to average dissolved ^{222}Rn but that metamorphic rocks produce significantly lower average dissolved ^{222}Rn; this is perhaps one of the more important observations in the Hylas area. An initial screening for areas of high dissolved ^{222}Rn in the Hylas region should be based on the geographic distribution of underlying rock types. In a companion study of the Hylas area (Gates and Gundersen, 1992), similar positive relationships of soil gas and indoor ^{222}Rn values to rock type were found. With the proposed ^{222}Rn drinking water standard in mind, screening measurements in the Hylas area should be directed initially to wells tapping mylonitic and granitic rocks.

CONCLUSIONS

CONTROLS ON URANIUM AND RADON DISSOLVED IN GROUND WATER

ROCK CHEMISTRY AND MINERALOGY

The concentrations of solid phase U (eU) indicate that the abundance of U (and Ra) located at the rock-water interface differs among the mylonitic, granitic, and metamorphic rocks. The observed abundances of dissolved U are probably controlled, in part, by labile U in these aquifers. High abundances of U in ground water from mylonite suggests that U is not strongly adsorbed onto mineral surfaces or that U-bearing minerals are more susceptible to release of U; thus, more U is present in solution. In contrast, low U in ground water from granite suggests that U is held more strongly to mineral surfaces or that U-bearing minerals are less susceptible to release of U; thus, less U is present in solution. In contrast to both mylonite and granite, the metamorphic units are uniformly low in solid-phase U and dissolved U, indicating that low U (and Ra) abundances limit dissolved ^{222}Rn to a much greater extent than in the mylonite or granite.

WATER CHEMISTRY

Abundances of most dissolved species in ground-water samples in the Hylas area do not change dramatically despite the fact that aquifer lithologies vary in composition from granitic to ultramafic. This observation, combined with the fact that ground water generally contains low total dissolved

Table 4. Results of saturation index for selected common uranium minerals.

[The computer program PHREEQE (Parkhurst and others, 1980) was used along with thermodynamic data for aqueous uranium species and minerals from Langmuir (1978). "ND" means that uranium was not detected in the sample and no SI values were calculated. For samples with U below the detection limit (<0.05 ppb), 0.05 ppb was used to calculate a maximum SI. Uraninite + (UO_2); coffinite = ($USiO_4$); schoepite = ($UO_3(OH)_2{*}2H_2O$); rutherfordine = (UO_2CO_3)]

SAMPLES OF GROUND WATER FROM GRANITES

Sample No.	Uraninite	Coffinite	Schoepite	Rutherfordine
88VA-7	-2.5	-2.8	-5.9	-4.6
88VA-23	-1.4	-1.6	-4.1	-3.4
90VA-5	ND	ND	ND	ND
90VA-7	-27	-27	-7.7	-5.8
90VA-8	-7.7	-8	-7.1	-6.5
90VA-9	ND	ND	ND	ND
90VA-10	-17	-18	-7.3	-5.7
90VA-11	-12	-13	-5.9	-5.3
90VA-12	-12	-13	-6.4	-5.5
90VA-13	-16	-16	-6.2	-5.3
90VA-14	-14	-15	-6.8	-5.6
90VA-15	-7.5	-7.8	-6.7	-6.3

SAMPLES OF GROUND WATER FROM MYLONITES

Sample Number	Uraninite	Coffinite	Schoepite	Rutherfordine
88VA-1	-3	-3.2	-4.6	-5
88VA-2	-3.9	-4.3	-4.4	-4.5
88VA-4	-5.1	-5.5	-4.3	-4.8
88VA-6	-3.6	-4.2	-5	-4.8
88VA-8	-4.3	-4.8	-4.8	-4.9
88VA-9	-3.3	-3.6	-5.8	-4.4
88VA-10	-2.8	-3.6	-5.2	-4.9
88VA-12	-5.8	-6.1	-5.8	-5.3
88VA-13	-4.6	-4.9	-5.2	-4.9
88VA-14	-5.8	-6	-6.3	-5.7
88VA-16	ND	ND	ND	ND
88VA-17	-4.5	-4.8	-5.1	-4.9
88VA-20	-3.9	-4	-5.8	-5
88VA-21	-5.1	-5.4	-5.6	-5.2
88VA-29	-9.5	-9.8	-4.9	-4.2
88VA-35	-4.4	-4.7	-4.8	-4.7
88VA-36	-5.6	-6.2	-5.1	-5.2
90VA-3	-8.8	-9.1	-6.6	-6

solids, is consistent with relatively rapid recharge attendant in a moist climate. Apparently, dissolution of rock-forming minerals in the different lithologic units that serve as aquifers has not proceeded to a great enough extent to impart a distinctive chemical signature to the various aquifer waters. As noted, the only major rock-forming mineral to approach saturation is quartz.

In contrast to major dissolved species, dissolved U and particularly dissolved ^{222}Rn, show a greater variation with aquifer lithology. In some areas, the concentrations of dissolved ^{222}Rn may exceed the newly proposed MCL of 300 pCi/L as ^{222}Rn, especially in ground waters from granitic and mylonitic rocks. Although ^{222}Rn concentrations in ground-water samples from the granite and mylonite encompass a fairly wide range and are not readily distinguishable, the ^{222}Rn concentrations in ground water from these two aquifers are much higher than in ground water obtained from the low-U metamorphic rocks.

Table 4. Results of saturation index for selected common uranium minerals.—Continued

SAMPLES OF GROUND WATER FROM METAMORPHIC ROCKS (* = AMPHIBOLITE)

Sample No.	Uraninite	Coffinite	Schoepite	Rutherfordine
88VA-3	-5.3	-5.7	-4.9	-5.4
88VA-5	-4.8	-5.2	-5.3	-5.1
88VA-11	-4.5	-4.8	-5.4	-5
88VA-15	-4.1	-4.3	-5.3	-5.1
88VA-18	-2.9	-3.2	-5.8	-4.9
88VA-19	-2.7	-3.1	-5.9	-4.9
88VA-22	-3.1	-4	-6	-4.9
88VA-24	-6.8	-7.1	-5.3	-4.7
88VA-25	ND	ND	ND	ND
88VA-26	-2.2	-2.8	-6.4	-4.9
88VA-27*	-8.7	-9.5	-5.4	-4.9
88VA-28	-3.5	-3.9	-6	-4.9
88VA-30	-3.7	-4.2	-6	-5.2
88VA-31*	-9.2	-10.	-5.9	-5.0
88VA-32	-1.6	-2.8	-6.3	-5.2
88VA-33	ND	ND	ND	ND
88VA-34	-3.1	-3.7	-5.8	-4.9
90VA-1	-16	-17	-6.2	-5.7
90VA-2*	-8.2	-8.7	-6.5	-6.3
90VA-4	ND	ND	ND	ND
90VA-6*	-5.8	-6.3	-6.5	-6.8
90VA-16	-17	-18	-6.2	-5.9

U and Ra solubility and adsorption at the rock-water interface are strongly influenced by ground-water chemistry, particularly pH, Eh (DO), and the concentration of CO_3^{2-} ions that form soluble complexes with U. The lower pH and lower alkalinity (as ΣCO_3) of ground water from metamorphic and granitic rocks favors adsorption of U (as UO_2) on mineral surfaces. Thus, the variation of dissolved U within and between different aquifers is also influenced by water chemistry that determines the importance of adsorptive uptake on mineral surfaces. Increased solubility and adsorption of U and Ra at the rock-water interface enhances the emanation of ^{222}Rn into ground water.

FRACTURES AND SURFACE AREA

Differences between mylonitic and granitic ground waters with respect to pH, ΣCO_3, U, TDS, and DO suggest that dissolved U may increase as a result of shearing and chemical weathering processes. Shearing and weathering of the mylonitic rocks increase the surface area that is available for reaction with ground water. Fracturing of anthropogenic or geological origin acts to increase the efficiency of rock-water interaction and the efficiency of ^{222}Rn emanation to pore water and needs to be investigated in greater detail.

SUMMARY

In general, ground water in the vicinity of Hylas, Virginia, is of good quality for domestic consumption and major and trace element chemistry does not vary greatly with host lithology. However, greater abundances of U and certain ions and trace elements in ground waters from mylonite indicate that weathering and probable increased surface area due to shearing enhances U release into ground water (and perhaps U- and Ra-siting at the rock-water interface), and thus ^{222}Rn emanation into ground water. Adsorption probably limits dissolved U (but not ^{222}Rn) to the lower levels observed in water samples from granitic rocks. Both dissolved U and ^{222}Rn in ground waters from metamorphic rocks appear to be limited by the low abundance of U. In areas where crystalline rocks are dominant, determination of lithologies would be a logical first step prior to examining ground-water supplies thought to have elevated levels of dissolved ^{222}Rn.

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