

**Evaluating the Use of Sr, Pb, and U Isotopes for Fingerprinting Surface
and Groundwaters in Southern New Hampshire**

by

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Abstract

Increased pressure on fresh water reserves in southern New Hampshire requires new methods for monitoring exploitation. In a pilot study, surface water and groundwater samples from southern New Hampshire were analyzed for Sr, Pb, and U isotopes to test if they could be used to distinguish between different reservoirs (surface waters, bedrock wells, and sand and gravel wells). Detecting and monitoring communication between different aquifers is an essential part of water management, and mixing between the reservoirs due to overproduction of the wells could adversely impact drinking water quality. The results show that a mix of $^{234}\text{U}/^{238}\text{U}$, $^{206}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios was the most useful in distinguishing between the reservoirs. This method has great potential for the long-term monitoring of water wells to prevent overproduction.

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1 Introduction

The growing population of southern New Hampshire places increasing pressure on the availability of clean water. As the demand for fresh water is met by increased pumping of groundwater, it becomes crucial for both industry and municipalities to know the source of their well water and the interactions between different reservoirs. The purpose of this study is to test the feasibility of using multiple radiogenic isotopes to fingerprint major groundwater and surface water reservoirs in North Hampton, New Hampshire. My approach is to establish a baseline of Sr, Pb, and U concentrations and their isotopic ratios for water in the area and to use the data to investigate mixing between ground and surface waters.

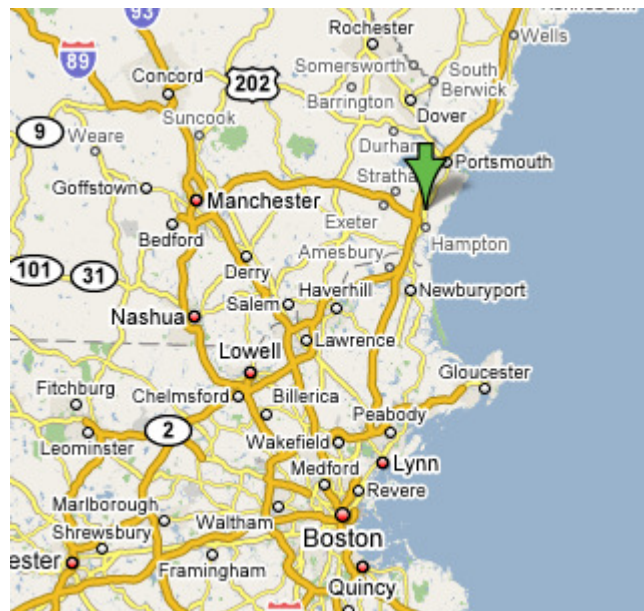


Figure 1: A map showing the location of North Hampton. See Figure 6 for a detailed map of the region.

A series of water wells drilled into both bedrock and sand and gravel in the vicinity of the Little River (Fig. 6) provides fresh water for the municipality of North Hampton (Fig. 1). Overpumping of the aquifers could lead to lower quality drinking

water if, for example, the wells begin drawing water from surface waters. While well water is monitored to ensure compliance with EPA drinking water standards, surface waters are more vulnerable to anthropogenic or natural sources of pollution. Using isotopes as tracers has the potential for determining the relative contributions of well and surface water to the drinking supply. There are relatively few studies of this kind, and this study explores the strengths and weaknesses of a multi-isotope approach for the waters of southern New Hampshire.

The isotopic composition of groundwater is controlled by the time-integrated history of interaction between the water and its host materials. Groundwater can flow through fracture-dominated porosity in igneous and metamorphic rocks, or through void spaces in sand and gravel aquifers. In each case, the chemistry of the water varies as a result of its history. The isotopic composition of surface water is controlled by precipitation run-off, and groundwater during low flow periods.

There are few studies where the data obtained from characterizing isotopic composition is then used to evaluate how groundwater from different sources and surface water may be connected (e.g. Hunt et al., 1998; Négrel et al., 2004; Woods et al., 2000). Although this is only a pilot study, it is hoped that the results will be useful for future applications.

2 Background: Isotopic Tracers

2.1 Sr

The four naturally-occurring isotopes of Sr are ^{84}Sr , ^{86}Sr , ^{87}Sr , and ^{88}Sr . ^{87}Sr is a daughter product from the decay of ^{87}Rb , with a half-life of 48.8 Ga (Steiger and Jäger, 1977). ^{88}Sr (the most abundant Sr isotope) and ^{86}Sr are both stable. ^{84}Sr (the least abundant isotope) is often enriched and used as a spike in isotopic dilution analysis.

In rocks and minerals, the factors that control the isotopic composition of Sr and how it changes with time are $^{87}\text{Rb}/^{86}\text{Sr}$ (the parent-daughter ratio) and the initial Sr concentration. The natural variations of $^{87}\text{Sr}/^{86}\text{Sr}$ found in rocks, minerals, and water are useful tools for understanding their history.

For dating igneous rocks, the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the parental magma is assumed to be homogenous throughout the magma. As the magma crystallizes, Rb is fractionated by different minerals, and each mineral develops a distinct Rb/Sr ratio. Rb^+ substitutes for K^+ in minerals, while Sr^{2+} substitutes for both Ca^{2+} and K^+ (Faure and Powell, 1972). Minerals with relatively high Rb/Sr ratios will result in higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratios over time than those with low initial Rb/Sr ratios; examples include micas and K-feldspar. In contrast, for minerals with high Sr-content and low Rb/Sr (plagioclase feldspar, calcite), the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios typically change relatively little with time.

Limestones (composed of CaCO_3) have a low parent/daughter ratio and high initial Sr content. Sr can substitute for Ca, but Rb cannot, so the $^{87}\text{Sr}/^{86}\text{Sr}$ in limestones remains virtually constant over time. As a result, limestones can be used as indicators of how the Sr isotopic composition of seawater from which the carbonate precipitates has changed over Earth history (Faure, 1991).

The ^{87}Sr content of a mineral at any time is described by the equation

$$^{87}\text{Sr} = ^{87}\text{Sr}_0 + ^{87}\text{Rb}(e^{\lambda t} - 1)$$

where $^{87}\text{Sr}_0$ is the amount of ^{87}Sr initially present when the mineral formed, and t is the time (in years) that has elapsed since then. The decay constant λ is calculated by $\ln 2 / t_{1/2}$ (where $t_{1/2}$ is the half-life of ^{87}Rb) and is equal to $1.42 \times 10^{-11} \text{ yr}^{-1}$ (Steiger and Jäger, 1977). Since measuring isotopic ratios is easier than measuring the absolute number of atoms of each isotope, the above equation is divided by ^{86}Sr , an isotope whose abundance in nature does not change with time.

$$\left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}} \right) = \left(\frac{^{87}\text{Sr}}{^{86}\text{Sr}} \right)_0 + \frac{^{87}\text{Rb}}{^{86}\text{Sr}} (e^{\lambda t} - 1)$$

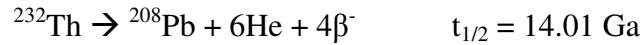
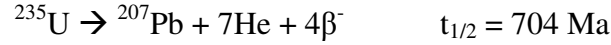
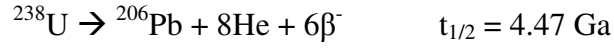
This is an equation for a straight line. If different samples from an igneous rock have the same initial $^{87}\text{Sr}/^{86}\text{Sr}$ but different Rb/Sr ratios, after time t the samples will plot on a line (called an isochron) with a slope of $e^{\lambda t} - 1$. The y-intercept of the isochron is the initial $^{87}\text{Sr}/^{86}\text{Sr}$ of the magma.

Isochrons are only valid for closed systems—rocks whose change in ^{87}Sr comes solely from the decay of ^{87}Rb originally present in the rock. Rocks whose ^{87}Sr content has been altered in other ways (such as weathering) are no longer closed systems.

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio is used for both radiometric dating and tracing solute sources in weathering studies. The Sr ratio of the water (whether surface water or groundwater) is a mixture of the Sr ratios from the rocks that it weathers, and thus different aquifers often have distinct ratios.

2.2 Pb and U

Pb has four naturally-occurring isotopes: ^{204}Pb , ^{206}Pb , ^{207}Pb , and ^{208}Pb . Only ^{204}Pb is essentially non-radiogenic. The other isotopes are produced by U or Th in the following ways:



(Faure, 1986; Steiger and Jäger, 1977). In each case, U and Th form a series of intermediate daughter products before Pb, but the half lives of the intermediates are insignificant compared to those of ^{238}U , ^{235}U , and ^{232}Th .

The ratios of Pb isotopes are expressed with respect to the concentration of ^{204}Pb , which does not change with time. If t is the amount of time that has elapsed since the rock system crystallized from magma at time 0, then the decay equations can be written as

$$\left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}} \right) = \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}} \right)_0 + \left(\frac{^{238}\text{U}}{^{204}\text{Pb}} \right) (e^{\lambda_1 t} - 1)$$

$$\left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}} \right) = \left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}} \right)_0 + \left(\frac{^{235}\text{U}}{^{204}\text{Pb}} \right) (e^{\lambda_2 t} - 1)$$

$$\left(\frac{^{208}\text{Pb}}{^{204}\text{Pb}} \right) = \left(\frac{^{208}\text{Pb}}{^{204}\text{Pb}} \right)_0 + \left(\frac{^{232}\text{Th}}{^{204}\text{Pb}} \right) (e^{\lambda_3 t} - 1)$$

where λ_1 , λ_2 , and λ_3 are the decay constants of ^{238}U , ^{235}U , and ^{232}Th , respectively (e.g. Dickin, 2005). These equations also describe isochrons.

U and Th often substitute for each other in minerals; both elements have similar ionic radii and exist as U^{+4} and Th^{+4} . Under oxidizing conditions, U is mobile and soluble

as UO_2^{2+} , the uranyl ion. U is insoluble under reducing conditions. Th and U are often fractionated from one another under oxidizing conditions, leading to elevated U/Th in water.

U and Th are relatively incompatible elements, concentrated in the last liquids to crystallize from a magma. As a result, U and Th are generally enriched in the continental crust compared to the upper mantle. Pb found in sources with elevated time-integrated U and Th content is called radiogenic lead. Zircons, for example, incorporate U and exclude Pb in their formation and thus accumulate pure radiogenic Pb over time. In contrast, non-radiogenic or common Pb precipitates in ore, forming minerals such as galena (PbS) with very low U/Pb and Th/Pb ratios (Faure, 1986). The Pb isotopic concentration of these minerals do not change appreciably with time.

The currently accepted value for the $^{238}\text{U}/^{235}\text{U}$ ratio of natural uranium is 137.88 (Steiger and Jäger, 1977). ^{238}U decays much more slowly than ^{235}U , thus ^{207}Pb increased at a faster rate than ^{206}Pb early in Earth history and the trend reversed in the last 1.0 billion years. This can best be visualized on a plot of $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 2), measured on mantle derived rocks and ores. The least radiogenic, known Pb with the lowest $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ is a troilite (FeS) from the Canyon Diablo meteorite. Its $^{207}\text{Pb}/^{206}\text{Pb}$ ratio has remained essentially unchanged with time due to an extremely low U/Pb ratio. The troilite's Pb isotopic composition is assumed to be the best estimate of the Earth's Pb isotopic composition when the Earth was formed. Along with other meteorites, the troilite was used to determine the age of the Earth. It was assumed that the meteorites were closed systems and formed at the same time. The range of $^{207}\text{Pb}/^{206}\text{Pb}$ ratios from the meteorites plots on a straight line called the geochron. $^{207}\text{Pb}/^{206}\text{Pb}$ ratios

from ocean sediments also fall on the geochron, thus the Earth and the meteorites share the same U-Pb system (Patterson, 1956). The slope of the geochron is the age of the Earth—the current accepted value is 4.567 Ga (Amelin, Ghosh, and Rotenberg, 2005).

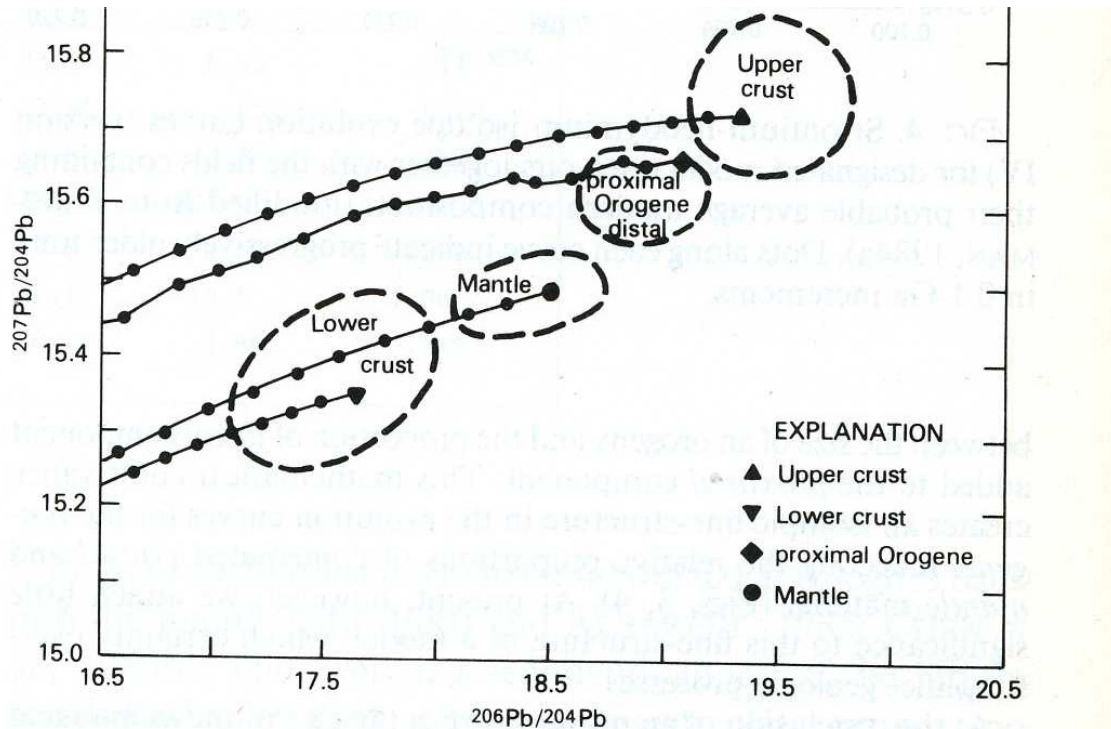


Figure 2: A plot of the current $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ for different systems on Earth. Each dot represents a 0.1 billion year increment, going backwards in time (Zartman and Haines, 1988).

The Pb isotopic composition of different systems is controlled by the initial U/Pb ratios and the time it has remained a closed system. For example, the upper crust, enriched in U, has a higher Pb growth curve than that of the bulk Earth. Industrial Pb has signature isotopic ratios that are controlled by the origin of the Pb ores, so anthropogenic Pb can be traced back to its source.

2.3 ^{234}U

^{234}U is a radiogenic isotope produced by ^{238}U (Fig. 3). The $^{234}\text{U}/^{238}\text{U}$ ratio is a useful tool for studying groundwater. Different aquifers have distinct ratios that are controlled by interactions between the water and the host rock (Ivanovich and Alexander, 1987).

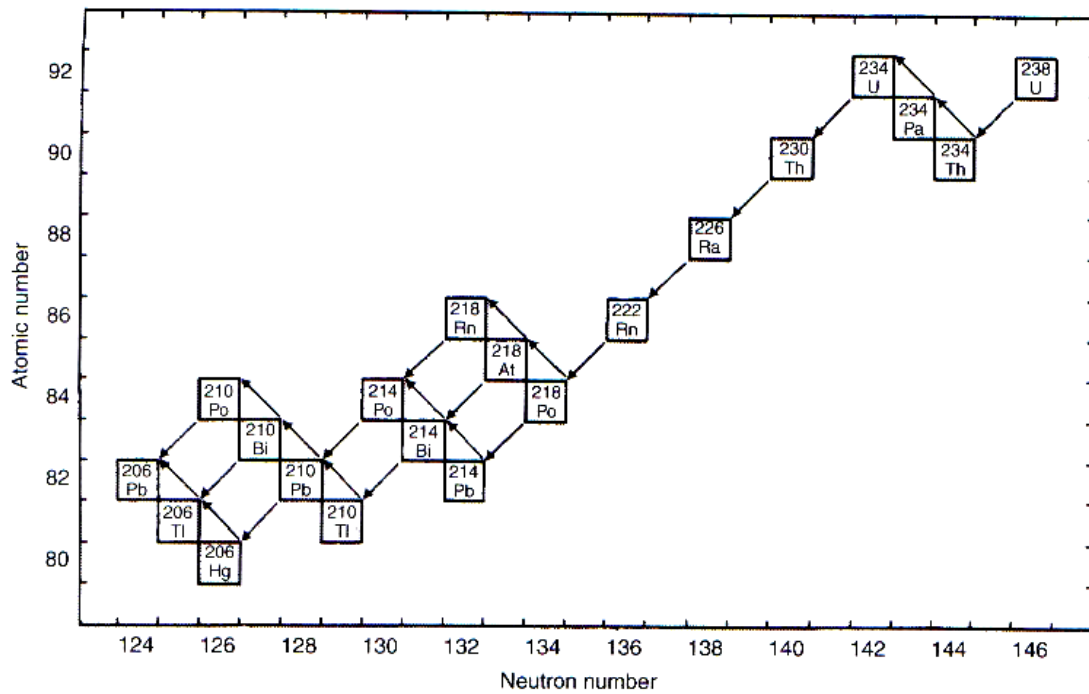


Figure 3: A diagram showing the position of ^{234}U within the ^{238}U decay chain (Faure and Mensing, 2005).

Due to its short half life ($t_{1/2} = 2.47 \times 10^5$ years), ^{234}U is present today only in small quantities. Equilibrium between ^{234}U and ^{238}U is measured by the activity ratio, equal to the average number of radioactively decaying atoms per unit time. Secular equilibrium is reached when the $^{234}\text{U}/^{238}\text{U}$ activity ratio is equal to one. Closed systems reach secular equilibrium after about 1 million years, but due to the input of ^{234}U over time, groundwater is rarely in secular equilibrium (Cherdyntsev et al., 1955; DePaolo et al.,

2006). The $^{234}\text{U}/^{238}\text{U}$ ratio of individual aquifers is controlled by the balance between alpha-recoil of ^{238}U decay and U dissolution in weathering. The high-energy release of alpha particles in ^{234}Th production causes the recoiled atom to damage the crystal lattice. ^{234}U is preferentially leached into the water through these damaged sites, increasing the $^{234}\text{U}/^{238}\text{U}$ ratio. (DePaolo et al., 2006; Maher et al., 2006). Under oxidizing conditions where U is in solution as the uranyl ion, the $^{234}\text{U}/^{238}\text{U}$ ratio of groundwater will also increase with time. Under reducing conditions, however, U is highly insoluble and the $^{234}\text{U}/^{238}\text{U}$ ratio will decrease with time (Fleischer, 1982; Kendall and McDonnell, 1998).

3 Geologic Setting

The bedrock geology of southeastern New Hampshire is dominated by metamorphosed sedimentary and volcanic rocks that are around 300-400 million years old (Fig. 4) (Lyons et al., 1997).

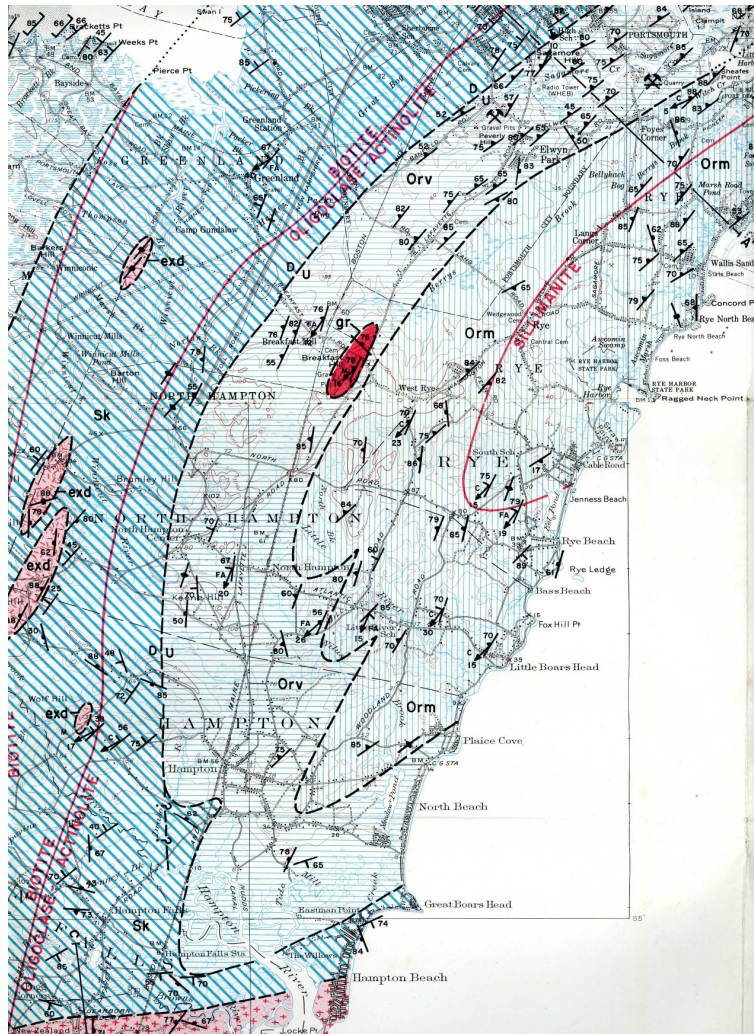


Figure 4: A map of bedrock geology in the region where the water samples were collected (Novotny, 1969).

All of the groundwater samples were taken from areas underlain by the Rye and Kittery Formations, which strike parallel to one another, north-to-south along the

seacoast. The Rye Formation (estimated to be Ordovician in age) borders the coast; it is roughly six kilometers wide and extends from Hampton Falls in the south to New Castle in the north. There are two subdivisions within the Rye Formation: the lower member is metasedimentary, and composed primarily of light-colored schists, quartzites, and gneisses. The upper metavolcanic member is dominated by dark-colored schists, gneisses, and amphibolites. The Kittery Formation lies to the west of the Rye Formation. A steep fault is the contact between the two formations; the Kittery Formation has moved stratigraphically down relative to the Rye Formation. Estimated to be Silurian in age, the Kittery Formation contains a mixture of dark-colored banded quartzite, interbedded with slate, phyllite, and fine-grained schist (Billings, 1956; Novotny, 1969). The surficial geology of the area is a mix of unstratified glacial till, stratified drift, and fine-grained marine deposits (Flanagan et al., 1998).

4 Methods

4.1 Sample Collection

Surface and well water samples were collected using bottles pre-cleaned with HNO_3 . Over time, metal cations in the water will adsorb onto the plastic bottle surfaces; to solve this problem following standard procedure, 2-5 mL concentrated HNO_3 were added to each bottle shortly after sampling (e.g. Environmental Protection Agency, 2007; Moody, 1982).

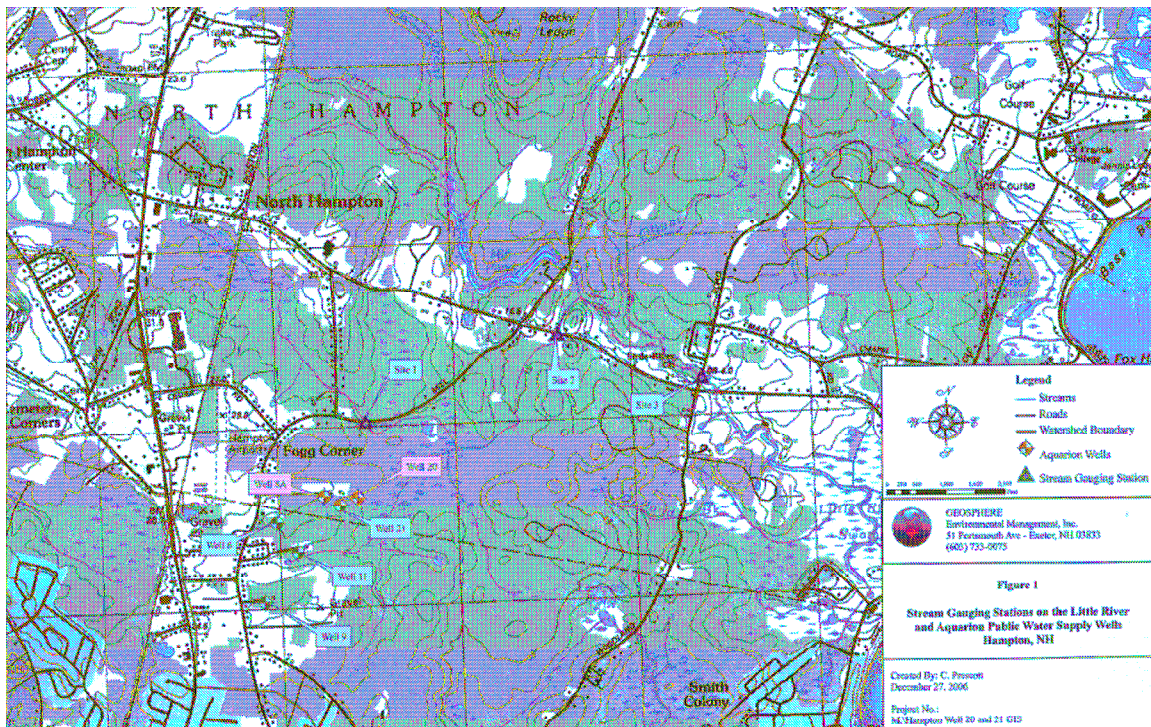


Figure 5: A map of surface water sample sites (Sites 1-3) (Prescott, 2006).

The Little River runs southeast through the Rye Formation and drains into the Atlantic Ocean. Surface water samples (SW) were collected from sites 1-3: SW-1 and SW-2 are from tributaries of the Little River (SW-2 is further downstream), and SW-3 is

from the Little River (Fig. 5). Site 1 is located in the metavolcanic member of the Rye Formation, while sites 2-3 are in the metasedimentary member.

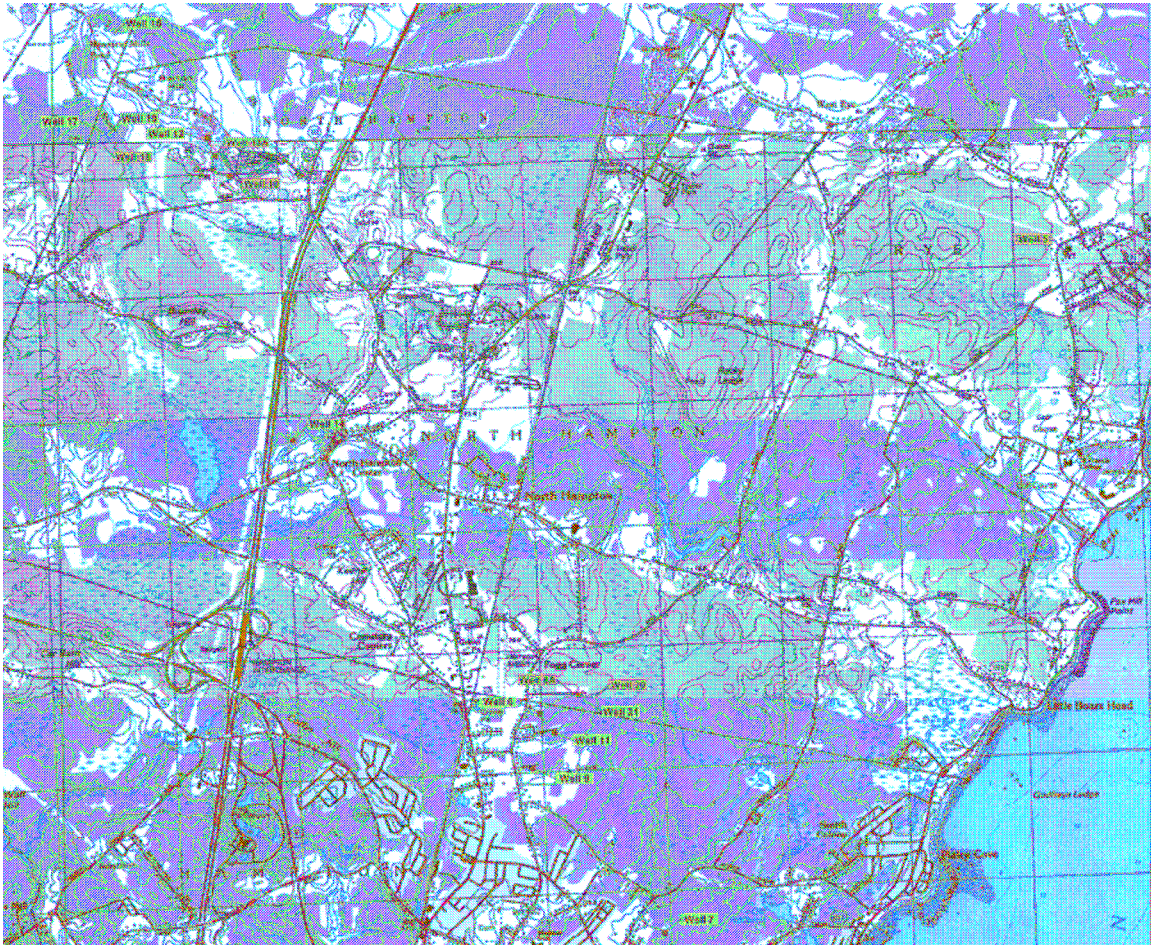


Figure 6: A map of all groundwater sample sites. Well 13 has two branches (13A and 13B) that share the same surface location; only Well 13A is labeled on the map. Scale: 1 inch = 2,750 ft (Mattimore, 2003)

Sixteen wells used for drinking water were also sampled (Fig. 6). Wells 13A, 13B, 17, 18, 19, 20, and 21 are located in bedrock. Of the bedrock wells, wells 13A, 13B, 17, 18, and 19 are in the Kittery Formation; wells 20 and 21 are in the metavolcanic member of the Rye Formation. The remaining wells are sand and gravel wells.

Samples were analyzed for Sr, Pb, and U isotopic composition (IC), as well as Sr and Pb concentrations using isotope dilution (ID). To find the amount of Sr present in a

sample, the sample is mixed with a prepared Sr "spike" or tracer solution that has been enriched in ^{84}Sr . The concentration of Sr in the sample can be calculated from the isotopic composition of the sample and the spike, and the concentration of the spike. The Pb concentration was determined using a tracer enriched in ^{208}Pb .

4.2 Sample Preparation

Sr IC

About 12-15 mL of water were measured into a 15-mL Teflon beaker. To decompose the organic matter, 1 mL concentrated HNO_3 was added to each sample. The samples were then dried on a hot plate. Subsequent ion exchange separation is done in HNO_3 , so once the samples were dry, 0.5 mL 3.5 M HNO_3 was added to each. The samples were capped and stored at room temperature in preparation for column separation.

Sr ID

For each sample, 20-30 μL of ^{84}Sr spike and 1 mL of the sample were added to a 15-mL Teflon beaker. Half a mL concentrated HNO_3 was added to dissolve the organic matter, and the sample was capped and refluxed on a hot plate for at least an hour. The sample was dried down, and 0.5 mL 3.5 M HNO_3 was added. The samples were capped and stored at room temperature for column separation.

Pb IC

Water from the samples was measured into beakers and dried on a hot plate. Different sample sizes were tested (15 mL, 30 mL, 110-120 mL), and the best results were obtained with the 15 mL samples. Larger sample sizes often resulted in very poor

ionization during analysis. It is possible that higher concentrations of sulfate in the larger samples precipitated during evaporation, effectively isolating the Pb from the solution prior to column separations. None of the 110-120 mL samples was successfully analyzed, and many of the 30 mL samples yielded results with poor precision. Surface water samples required an extra step: to decompose the remaining organic matter, 0.2 mL concentrated HNO_3 and 0.1 mL H_2O_2 were added to the beaker and the sample placed on a hot plate for 15-20 minutes. Two more aliquots of H_2O_2 were added, and the sample dried down.

Unlike the Sr samples, Pb samples must be converted to chloride form for the column procedure. To do this, 2 mL 6 M HCl were added to the dried samples. The beakers were capped and placed on the hot plate for at least an hour. The samples were dried down. Half a mL 1.1 M HBr was added to each beaker. The samples were capped and stored at room temperature until column separations were performed.

NH_4OH was added to some samples before evaporation. If there is enough Ca, Mg, or Fe in the samples, ammonium hydroxide causes Pb to co-precipitate with $\text{Ca}(\text{OH})_2$, $\text{Mg}(\text{OH})_2$, and $\text{Fe}(\text{OH})_3$. By centrifuging the samples, sulfates in the liquid are separated from Pb in the solid form. The precipitates contain all of the Pb, and are soluble in HCl. The samples were then processed as usual with enhanced recovery of Pb.

Pb ID

About 0.02-0.04g of ^{208}Pb spike was added to a 15-mL Teflon beaker and the weight of the spike recorded. 1 mL sample water was added to each beaker and the weight of the water recorded. Half a mL concentrated HNO_3 was added to each beaker

and the samples dried down on a hot plate. The rest of Pb ID preparation followed the same steps as Pb IC.

U IC

About 110-120 mL of water were measured into 120-mL Teflon beakers and dried down with 3-5 mL concentrated HNO_3 . The samples were transferred into 15-mL Teflon beakers once the samples evaporated to less than 15-mL. If the samples had evaporated completely, 3-5 mL concentrated HNO_3 were used for the transfer. The samples were dried down on a hot plate.

The remaining steps are the same as those used for Pb IC preparation. Like Pb, U must be converted to chloride form for column separation.

4.3 Ion Exchange Separations

The purpose of column ion exchange is to isolate the ion of interest from the rest of the sample. This is performed by first loading the sample onto a column containing an ion exchange resin (Marhol and Svehla, 1982). In the case of Sr, which sorbs onto the resin under acidic conditions, acid is used to rinse the column. This has the effect of washing out most other elements. Under low acidity or neutral conditions, Sr desorbs from the resin, so water was used to elute the Sr. Similarly, Pb sorbs to the resin in weak acid (unlike most other elements) and desorbs in strong acid. Each specific column ion exchange procedure is based on the sorption and desorption characteristics of the element of interest.

Sr separation was performed using Eichrom Sr-spec resin. Samples were loaded onto 50 μL columns containing pre-cleaned Sr resin. The columns were rinsed with 3.5 M HNO_3 to remove unwanted elements, and the Sr was eluted with MQ water.

For Pb, the separation was done on pre-cleaned Eichrom AG1-X8, 200-400 mesh Cl^- form resin using 120- μL columns. The columns were washed with 1.1 M HBr and 2 M HCl. Pb was eluted with 6 M HCl.

Column separation of U used 120-mL columns containing pre-cleaned Eichrom AG1-X8, 200-400 mesh Cl^- form resin. The acid from the Pb sample loading and subsequent HBr wash steps was collected and loaded onto U columns. After drying down, the samples were taken up in 3 M HCl and processed again using U columns and pre-cleaned AG1-X8 resin. The columns were rinsed with 3 M HCl and U eluted with 0.1 M HCl.

4.4 Mass spectrometry

After the samples were purified through column ion exchange, they were loaded onto rhenium filaments for analysis using thermal-ionization mass spectrometry (TIMS). The samples are purified before analysis as the TIMS method separates ions based on their mass-to-charge ratio, and different elements with similar masses can create isobaric interferences. For example, ^{87}Sr and ^{87}Rb have almost identical masses, thus it becomes difficult to distinguish between the two isotopes in the mass spectrometer. Isobaric interference is avoided due to the selective nature of the Sr resin, which excludes Rb but retains Sr in the elution.

In TIMS, the filament is heated with a current to evaporate and ionize the sample. The ions are accelerated by 8,000 volts of potential through a magnetic field toward a series of detectors, and heavier masses are deflected less than lighter masses.

Mass fractionation occurs as a result of volatilization and ionization. As the samples are heated, the lighter isotopes are released more easily due to their weaker bonds. To adjust for the effects of fractionation, the samples' isotopic ratios are compared to the isotopic composition of standards. The standards are measured frequently in the lab, and have isotopic compositions that are well-known and agreed upon. One can use the difference between the measured and accepted values to correct for fractionation.

5 Data

Data tables of all isotopic analyses (Tables 2-5) and a discussion of data quality (Fig. 14-16, Table 6) are included in the appendix. Table 1 lists the analyzed samples separated by collection time and sample type.

Table 1: A summary of isotopic analyses for the samples.

Date Collected	Sample types	Analyses
6-25-2007	3 surface water samples 6 bedrock wells 10 sand and gravel wells	19 Sr IC (and 4 replicates), 19 Sr ID (and 4 replicates), 17 Pb IC (and 3 replicates), 19 Pb ID (and 3 replicates), 17 U IC (and 1 replicate)
8-30-2007	2 surface water samples	2 Sr IC, 2 Sr ID, 2 Pb IC (and 1 replicate), 2 Pb ID
2-28-2008	3 surface water samples	3 Sr IC, 3 Sr ID, 1 Pb IC, 3 Pb ID
2-29-2008	7 bedrock wells 10 sand and gravel wells	17 Sr IC, 17 Sr ID, 14 Pb IC, 17 Pb ID
3-5-2008	4 surface water samples	4 Sr IC, 4 Sr ID, 4 Pb IC, 4 Pb ID

Due to difficulties encountered in the Pb extraction process, some samples yielded results with lower than expected precision. As described in the methods section, insoluble sulfate precipitates may contain much of the Pb. Data points with unacceptably large uncertainties were not included in the graphs, and many samples were re-run to generate better data. For multiple analyses of the same sample, only the data point with better precision was graphed.

6 Discussion

6.1 Sr Isotopes

The Sr content of all samples ranges from 0.0220-4.7710 ppm (Fig. 7). Most surface water samples have lower Sr concentrations than the majority of sand and gravel wells. With the exception of one sample with an abnormally low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.709269, the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of surface water samples range between 0.712466- 0.71395.

There is a high degree of overlap between the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of surface water and bedrock well samples, which suggests mixing between the two reservoirs. The overlap can also be explained if surface waters are dominated by bedrock weathering. Similar trends have been observed for the Connecticut River watershed: Douglas et al. (2002) conclude that $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.7110-0.7180) of the Connecticut River's tributaries reflect the underlying bedrock geology. Surface water $^{87}\text{Sr}/^{86}\text{Sr}$ values in this study fall within the larger range obtained by Douglas et al. Likewise, a study by Hogan and Blum (2003) at the Hubbard Brook Experimental Forest in central New Hampshire found that the Sr ratios of stream water are sensitive to variable compositions of the underlying bedrock. The range of values from Hogan and Blum (0.72034-0.72049) are higher than the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios reported here and must reflect differences in bedrock composition between the two sites.

In this study, sand and gravel wells show a wider range of $^{87}\text{Sr}/^{86}\text{Sr}$ values (0.711960-0.715132), although most of the ratios are above 0.714000. In contrast, the majority of surface water samples have lower ratios, between 0.7120-0.7130. The highly variable Sr ratios obtained for sand and gravel well samples indicates that neither precipitation nor surface waters are the main sources of recharge for these wells—the

$^{87}\text{Sr}/^{86}\text{Sr}$ ratio of precipitation in coastal New Hampshire is dominated by the composition of aerosols over the ocean, which would be near 0.70916. The elevated $^{87}\text{Sr}/^{86}\text{Sr}$ ratios can be explained in at least two ways. It is possible that the sand and gravel wells are pumping older water with more radiogenic Sr that reflect the weathering of rocks following deglaciation around 12,000 years ago (Flanagan et al., 1998). Another possibility is that the groundwater has been in contact with clays and weathered minerals derived from glacially-transported debris (such as till and clays) whose Sr isotopic composition is different from that of the local bedrock. This theory can be investigated by carbon-dating the groundwater or analyzing till materials.

The sample with the low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio was taken from site 3 at the end of summer (8/30/2007). Currently, the accepted $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the oceans is 0.70916, similar to that of this surface water sample (Palmer and Edmond, 1989). The value obtained for site 3 (0.709269) suggests that the Sr ratio is dominated by seawater and must reflect mixing between the Little River and the ocean during times of low flow. Interestingly, another surface water sample taken from site 2 (a short distance away from site 3) on the same day (and essentially at the same time)* has the highest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of all surface water samples (0.71395). This can be explained by the differences in location and elevation between the two sites. Site 3 is one meter above sea level (while site 2 is about 5 meters above sea level) and much closer to the coast, hence site 3 is more vulnerable to tidal mixing. Another factor is the lack of continuous stream flow at site 2 on the day of collection;* the stagnant water at site 2 may have been fed in part by groundwater.

* Abby Thompson, personal communication.

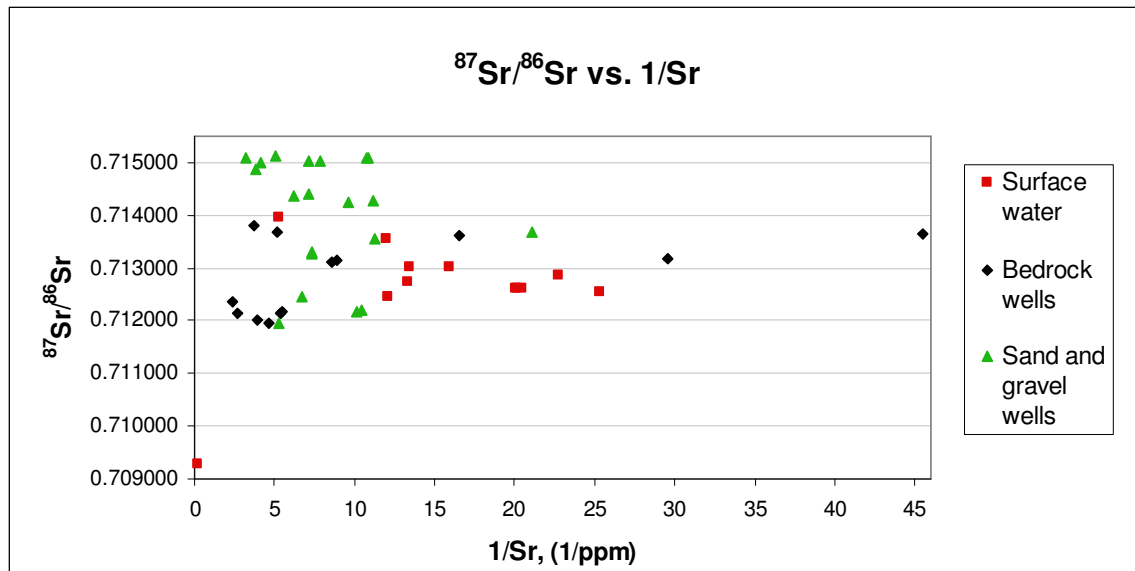


Figure 7: A plot of $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $1/\text{Sr}$, separated by sample type. Sand and gravel wells have a wider range of $^{87}\text{Sr}/^{86}\text{Sr}$ values, while the Sr ratios of bedrock wells and surface waters overlap with each other. One surface water sample with a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.709269 indicates mixing with seawater.

6.2 $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$

For this study, surface water samples are clearly separated from sand and gravel wells in $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space (Fig. 8). Most of the variability lies in the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio; the $^{206}\text{Pb}/^{204}\text{Pb}$ values of surface waters and sand and gravel wells do not overlap. Sand and gravel wells have a wide range of Pb isotopic values while bedrock wells and surface water samples have more similar Pb ratios. As was the case in $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $1/\text{Sr}$ space, $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space supports the idea that sand and gravel wells pump water that is distinct from surface and bedrock well waters.

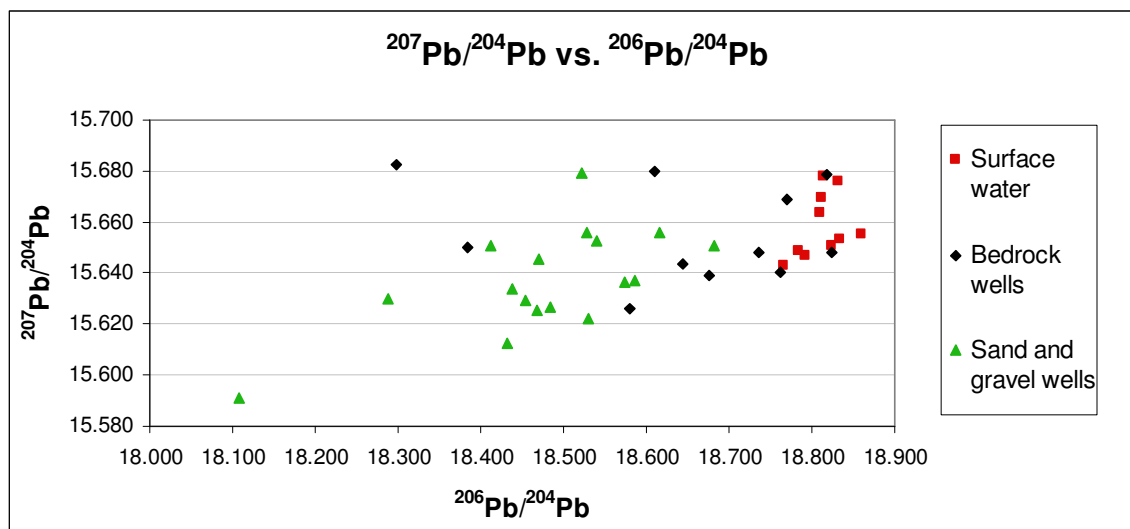


Figure 8: A plot of $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ shows that surface water samples do not overlap with sand and gravel well samples.

6.3 $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$

All of the surface water samples have higher $^{208}\text{Pb}/^{204}\text{Pb}$ ratios (38.386-38.526) than sand and gravel wells (37.862-38.345). While the range of ratios for bedrock wells overlap with the values of the other sample types, surface waters and sand and gravel well samples can be differentiated in $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space (Fig. 9).

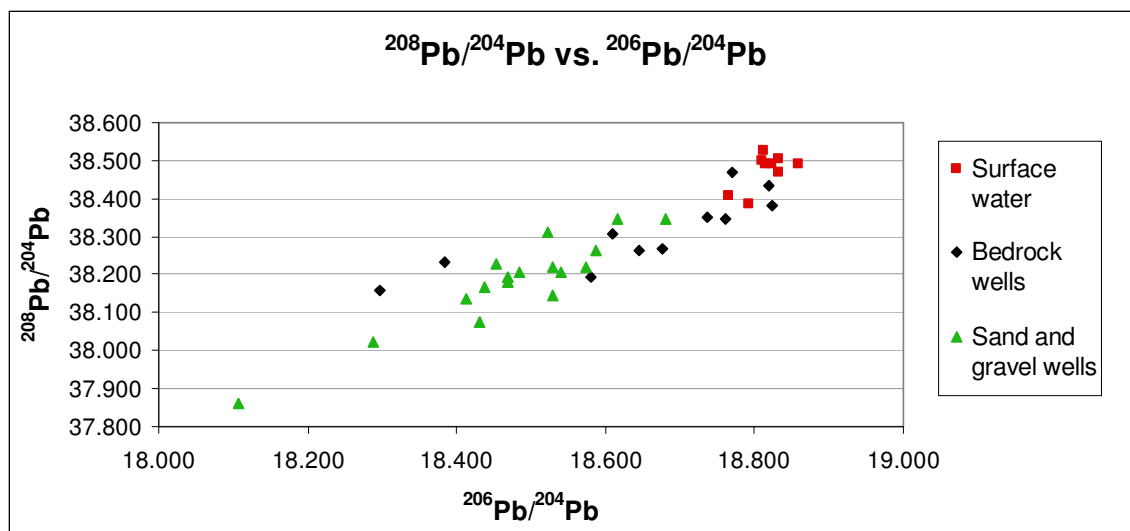


Figure 9: A plot of $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, separated by sample type. Surface waters are clearly separated from sand and gravel wells.

6.4 $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$

Surface waters, most bedrock wells, and sand and gravel wells are clearly separated in $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space (Fig. 10), with most of the variability in the $^{206}\text{Pb}/^{204}\text{Pb}$ ratio. Once again, this plot shows that the isotopic signature of the sand and gravel well waters is distinct from that of the other two reservoirs, which overlap more with each other.

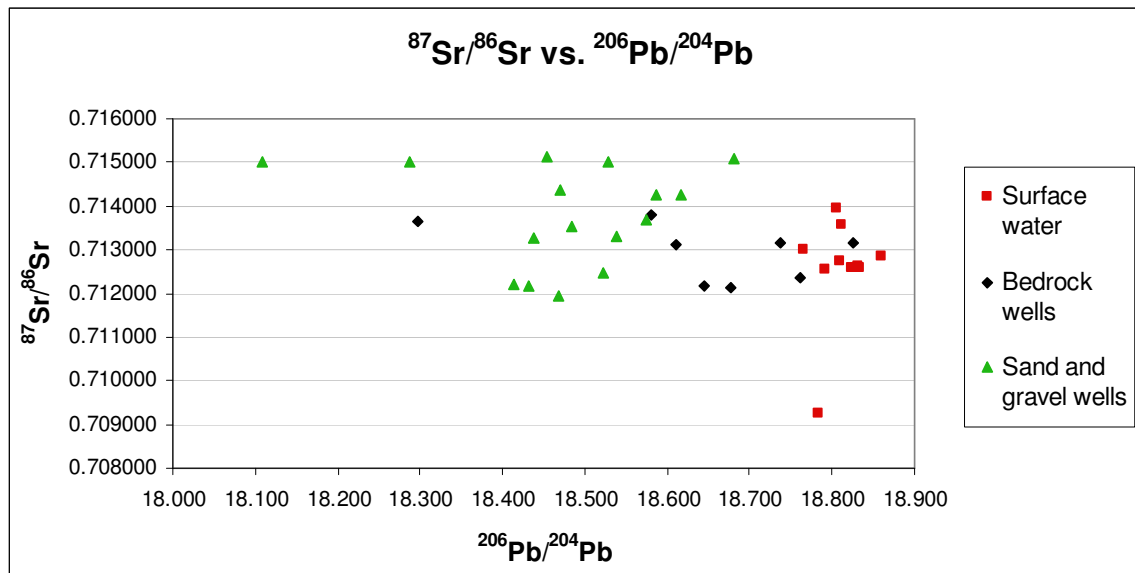


Figure 10: With few exceptions, samples from the three reservoirs can be distinguished from one another in this plot of $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$.

6.5 $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$

Well water samples have a wide range of Pb concentrations, which overlap with the values for surface water (Fig. 11). In general, surface water samples have higher Pb concentrations (0.5732-11.8 ppb) than well waters (0.1244-10.04 ppb). The concentration of Pb in the wells is below the EPA maximum limit of 15 ppb in drinking water (Environmental Protection Agency, 2008).

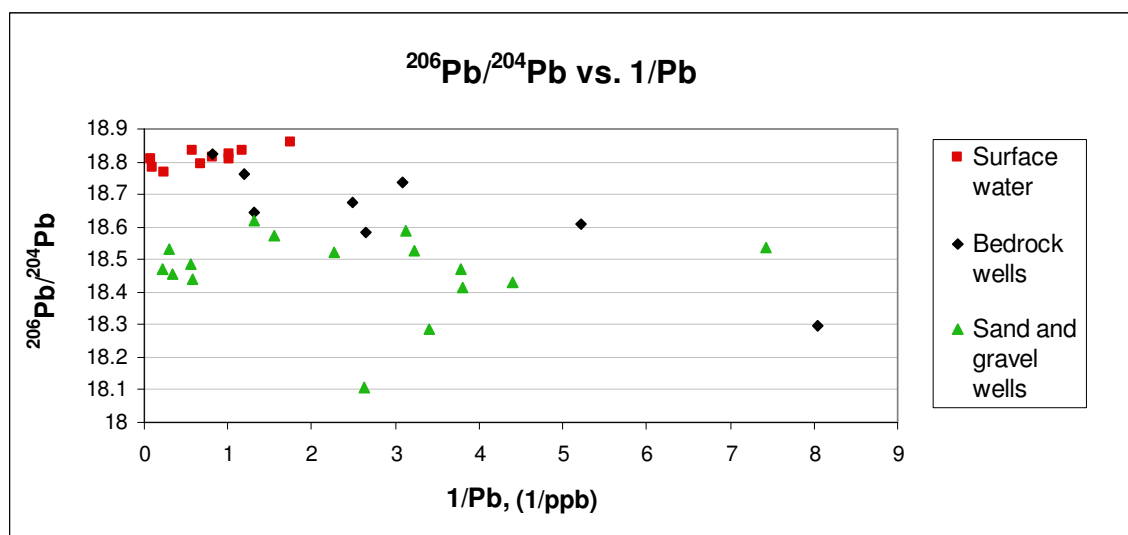


Figure 11: A plot of $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$, separated by sample type. The data suggest mixing between surface waters and bedrock wells. Sand and gravel wells are clearly distinguishable from surface water samples.

A near-linear trend in $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$ space for surface waters and bedrock wells shows evidence of mixing between the reservoirs. Figure 12 shows a plot of ideal mixing behavior. The bedrock end-member has a Pb concentration of 0.1244 ppb and a $^{206}\text{Pb}/^{204}\text{Pb}$ ratio of 18.297. Surface water samples are likely to be mixtures of different components (e.g. precipitation), so the surface water end-member was found by the intersection of the two trendlines drawn for the reservoirs (2.44 ppb Pb, $^{206}\text{Pb}/^{204}\text{Pb} = 18.8$). This ideal mixing line can be used to calculate the relative contributions of both

components to any sample whose isotopic composition falls on the line drawn between these two end-members. Again, mixing between surface waters and bedrock wells supports the idea that water from sand and gravel wells is in a different system and that there is minimum communication between these wells and the other two reservoirs.

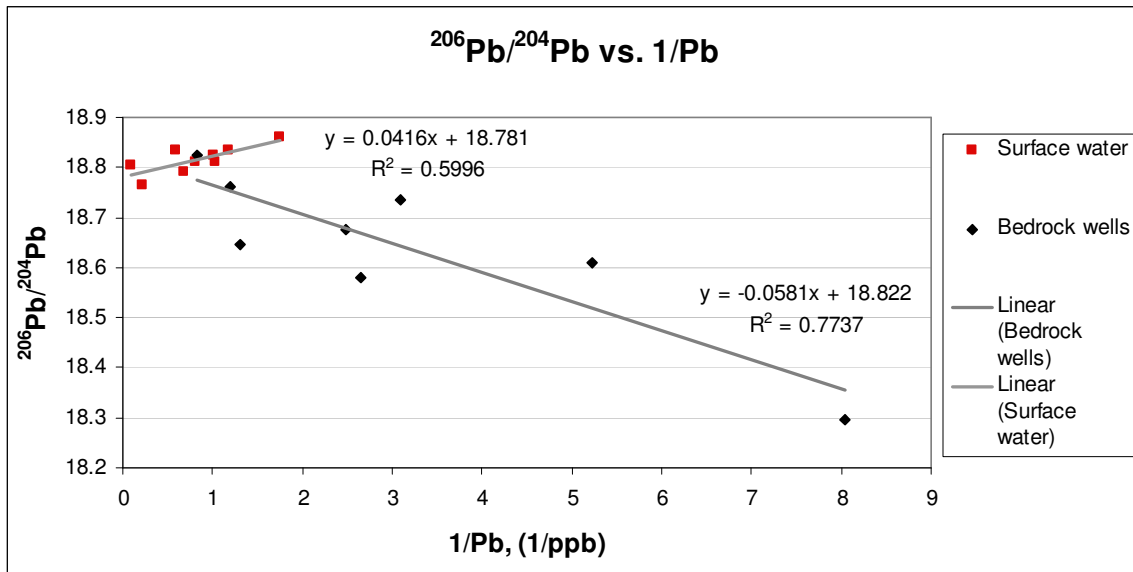


Figure 12: A plot of ideal mixing behavior between surface waters and bedrock wells in $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$ space. The low-flow surface water samples have been removed from the graph.

6.6 $^{234}\text{U}/^{238}\text{U}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$

On a plot of $^{234}\text{U}/^{238}\text{U}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 13), bedrock wells show elevated $^{234}\text{U}/^{238}\text{U}$ ratios compared to surface water and sand and gravel wells. This suggests that water in the bedrock wells has been leaching ^{234}U from U-bearing minerals. Bedrock wells also appear to have higher $^{206}\text{Pb}/^{204}\text{Pb}$ than the majority of sand and gravel wells. In $^{234}\text{U}/^{238}\text{U}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space, all three reservoirs are separated from one another.

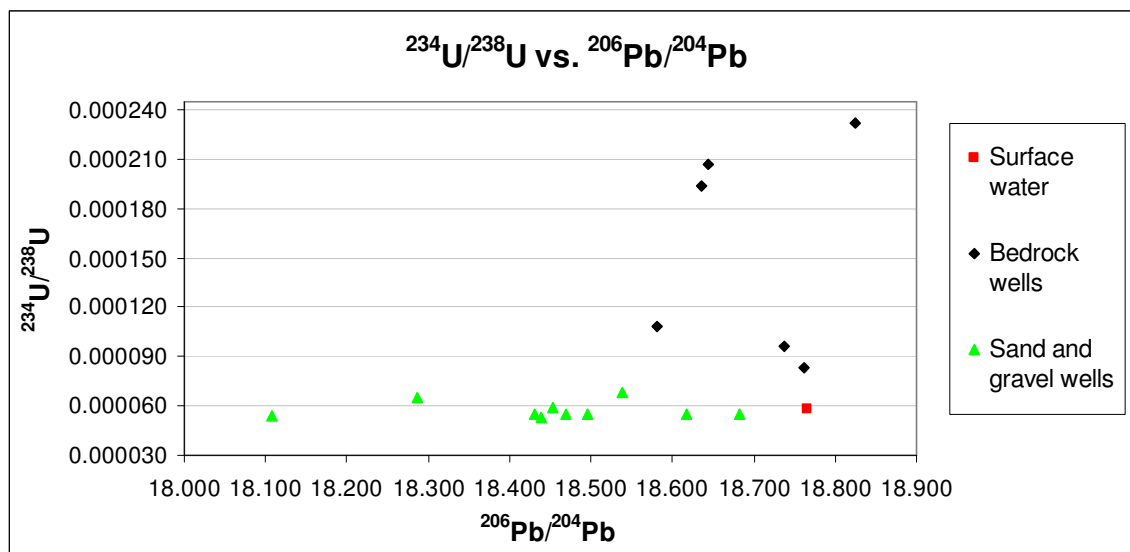


Figure 13: A plot of $^{234}\text{U}/^{238}\text{U}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, separated by sample type. Sand and gravel wells have lower $^{234}\text{U}/^{238}\text{U}$ ratios (around 0.000050) than bedrock wells (0.000084-0.00023).

7 Conclusions

Analyses of surface and well samples from southern New Hampshire suggest that different reservoirs can be distinguished from one another based on data from a multi-isotope (Sr, Pb, and U) approach. Surface waters are clearly separated from sand and gravel wells in $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$ space. Isotopic measurements show that bedrock wells have elevated $^{234}\text{U}/^{238}\text{U}$ ratios compared to other samples; most bedrock wells can also be distinguished from the other two sample types in $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ space. In general, a combination of $^{234}\text{U}/^{238}\text{U}$, $^{206}\text{Pb}/^{204}\text{Pb}$, and $^{208}\text{Pb}/^{204}\text{Pb}$ ratios is the most useful for distinguishing between all three reservoirs.

The largest range of isotopic ratios was obtained for sand and gravel wells, which likely reflects the heterogeneous nature of the aquifers as well as sources of recharge. High $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for sand and gravel wells indicate that neither precipitation nor surface waters are the main source of recharge. It is also possible that sand and gravel wells are pumping older water that reflects weathering from deglaciation around 12,000 years ago, or equilibration with glacially-transported debris. Bedrock wells and surface water samples overlap with each other in $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $1/\text{Sr}$, $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$, and $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$ spaces, while sand and gravel wells generally remain isotopically distinct.

The plot of $^{206}\text{Pb}/^{204}\text{Pb}$ vs. $1/\text{Pb}$ is consistent with mixing between bedrock wells and surface waters. Mixing between these two reservoirs supports the idea that surface water and bedrock wells can be distinguished from sand and gravel wells. The lack of mixing between surface water and sand and gravel wells can be explained if these

aquifers are better sealed and less open to percolation from surface flow than bedrock wells. Information on the wells' pumping history would provide key data to further support or refute this mixing theory. The data presented here could also be used to detect for the overpumping of sand and gravel aquifers that are drawing in surface waters.

8 Future Studies

Future studies will better constrain the isotopic signatures of different reservoirs, especially their temporal trends as affected by climatic variability and water production. For this study, only two samples per well were taken, spaced 8 months apart, and the four surface water samples were not taken at regular intervals. More comprehensive time-series data from single wells may allow one to monitor the effects of precipitation and changes in surface water flow. Information on when and how the wells were pumped will provide additional insight for mixing between surface and groundwater reservoirs. Mixing can be further constrained by analyzing U concentrations. The theory that there is older water in the sand and gravel wells can be tested by carbon-dating the groundwater and analyzing for the signature of weapons testing . Once a baseline is established, the isotopic signatures of the waters can be used as a sensitive monitor of resource exploitation, and as a tool in planning for conservation.

Appendix

Data tables

Table 2: Sr isotope ratio and concentration data, separated by sample type (data table continues on next two pages).

Date collected	MIT #	Well #	HC? (1)	$^{87}\text{Sr}/^{86}\text{Sr}$ (2)	% S.E. (3)	2 sigma s.e.	Sr, ppm	1/Sr, ppm
	Surface water samples							
6/25/2007	T 1341	SW-1	no	0.712735	0.0006	0.000009	0.0749	13.3511
6/25/2007	T 1342	SW-2	no	0.713560	0.0006	0.000009	0.0836	11.9617
6/25/2007	T 1342(2)	SW-2	no	0.713562	0.0005	0.000007	0.0835	11.9760
6/25/2007	T 1343	SW-3	no	0.713018	0.0006	0.000009	0.0742	13.4771
	Bedrock wells							
6/25/2007	T 1357	Well 13B	no	0.713153	0.0005	0.000007	0.1121	8.9206
6/25/2007	T 1353	Well 17	no	0.712357	0.0006	0.000009	0.4138	2.4166
6/25/2007	T 1352	Well 18	no	0.711960	0.0006	0.000009	0.2138	4.6773
6/25/2007	T 1354	Well 19	no	0.712157	0.0007	0.000010	0.1829	5.4675
6/25/2007	T 1345	Well 20	no	0.713174	0.0006	0.000009	0.0341	29.3255
6/25/2007	T 1345(2)	Well 20	no	0.713161	0.0005	0.000007	0.0338	29.5858
6/25/2007	T 1346	Well 21	no	0.713817	0.0006	0.000009	0.2659	3.7608

(1) Indicates presence of hydrocarbons in sample.

(2) Long term reproducibility of NBS-987 at MIT: 0.710240 ± 0.000014 (2-sigma s.d.).

(3) Within-run internal precision of measured ratio.

Date collected	MIT #	Well #	HC? (1)	$^{87}\text{Sr}/^{86}\text{Sr}$ (2)	% S.E. (3)	2 sigma s.e.	Sr, ppm	1/Sr, ppm
	Sand and gravel wells							
6/25/2007	T 1359	Well 5	no	0.711962	0.0006	0.000009	0.1883	5.3107
6/25/2007	T 1347	Well 6	no	0.714261	0.0006	0.000009	0.0898	11.1359
6/25/2007	T 1347(2)	Well 6	no	0.714249	0.0006	0.000009	0.0893	11.1982
6/25/2007	T 1350	Well 7	no	0.712187	0.0006	0.000009	0.0984	10.1626
6/25/2007	T 1344	Well 8A	no	0.715097	0.0006	0.000009	0.0917	10.9051
6/25/2007	T 1349	Well 9	no	0.713263	0.0006	0.000009	0.1364	7.3314
6/25/2007	T 1358	Well 10	no	0.714996	0.0005	0.000007	0.2405	4.1580
6/25/2007	T 1348	Well 11	no	0.715027	0.0005	0.000007	0.1272	7.8616
6/25/2007	T 1356	Well 12	no	0.715132	0.0007	0.000010	0.1964	5.0916
6/25/2007	T 1351	Well 14	no	0.713556	0.0005	0.000007	0.0890	11.2360
6/25/2007	T 1351(2)	Well 14	no	0.713550	0.0006	0.000009	0.0886	11.2867
6/25/2007	T 1355	Well 16	no	0.714397	0.0006	0.000009	0.1406	7.1124
	Surface water samples							
8/30/2007	T 1383	SW-2	no	0.713950	0.0006	0.000009	0.1888	5.2966
8/30/2007	T 1384	SW-3	no	0.709269	0.0007	0.000010	4.7710	0.2096
	Surface water samples							
2/28/2008	T 1486	SW-1	no	0.712466	0.0007	0.000010	0.0824	12.1359
2/28/2008	T 1487	SW-2	no	0.713012	0.0008	0.000011	0.0629	15.8983
2/28/2008	T 1488	SW-3	no	0.712618	0.0007	0.000010	0.0498	20.0803

Date collected	MIT #	Well #	HC? (1)	$^{87}\text{Sr}/^{86}\text{Sr}$ (2)	% S.E. (3)	2 sigma s.e.	Sr, ppm	1/Sr, ppm
	Bedrock wells							
2/29/2008	T 1497	Well 13A	no	0.713682	0.0006	0.000009	0.1954	5.1177
2/29/2008	T 1497-1	Well 13B	no	0.713122	0.0006	0.000009	0.1161	8.6133
2/29/2008	T 1500	Well 17	no	0.712144	0.0006	0.000009	0.3698	2.7042
2/29/2008	T 1501	Well 18	no	0.712012	0.0005	0.000007	0.2539	3.9386
2/29/2008	T 1502	Well 19	no	0.712144	0.0006	0.000009	0.1848	5.4113
2/29/2008	T 1503	Well 20	no	0.713630	0.0007	0.000010	0.0606	16.5017
2/29/2008	T 1504	Well 21	no	0.713648	0.0008	0.000011	0.0220	45.4545
	Sand and gravel wells							
2/29/2008	T 1489	Well 5	no	0.712465	0.0005	0.000007	0.1488	6.7204
2/29/2008	T 1490	Well 6	no	0.714245	0.0006	0.000009	0.1035	9.6618
2/29/2008	T 1491	Well 7	no	0.712192	0.0006	0.000009	0.0956	10.4603
2/29/2008	T 1492	Well 8A	no	0.715104	0.0008	0.000011	0.0934	10.7066
2/29/2008	T 1493	Well 9	no	0.713297	0.0006	0.000009	0.1354	7.3855
2/29/2008	T 1494	Well 10	no	0.714885	0.0006	0.000009	0.2587	3.8655
2/29/2008	T 1495	Well 11	no	0.715025	0.0006	0.000009	0.1393	7.1788
2/29/2008	T 1496	Well 12	no	0.715080	0.0007	0.000010	0.3129	3.1959
2/29/2008	T 1498	Well 14	no	0.713677	0.0006	0.000009	0.0475	21.0526
2/29/2008	T 1499	Well 16	no	0.714363	0.0007	0.000010	0.1603	6.2383
	Surface water samples							
3/5/2008	T 1505	SW-1	no	0.712604	0.0008	0.000011	0.0489	20.4499
3/5/2008	T 1506	SW-1a	no	0.712601	0.0006	0.000009	0.0495	20.2020
3/5/2008	T 1507	SW-2	no	0.712858	0.0007	0.000010	0.0439	22.7790
3/5/2008	T 1508	SW-3	no	0.712543	0.0005	0.000007	0.0395	25.3165

Table 3: Pb IC data, separated by sample type (data table continues on next two pages).

Date of			HC?		Pb Isotopic Ratios (2)								
Collection	MIT #	Well #	(1)	²⁰⁶ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁴ Pb	% SE	²⁰⁸ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁶ Pb	% SE	²⁰⁸ Pb/ ²⁰⁶ Pb	% SE
	Surface water samples												
6/25/2007	T 1341	SW-1	no	18.810	0.032	15.664	0.032	38.500	0.032	0.83274	0.00300	2.0468	0.0026
6/25/2007	T 1342	SW-2	no	18.812	0.027	15.670	0.027	38.526	0.028	0.83295	0.00270	2.0479	0.0021
6/25/2007	T 1342(2)	SW-2	no	18.720	0.750	15.691	0.750	IC did not run		0.83820	0.0770		
6/25/2007	T 1343	SW-3	no	18.765	0.009	15.643	0.010	38.407	0.009	0.83361	0.00130	2.0467	0.0010
	Bedrock wells												
6/25/2007	T 1357	Well 13B	no	18.825	0.043	15.648	0.043	38.382	0.043	0.83125	0.00390	2.0389	0.0027
6/25/2007	T 1353	Well 17	no	18.761	0.068	15.640	0.066	38.347	0.066	0.83364	0.00630	2.0440	0.0045
6/25/2007	T 1352	Well 18	no	18.637	0.103	15.627	0.103	38.210	0.103	0.83850	0.00760	2.0503	0.0058
6/25/2007	T 1354	Well 19	no	18.645	0.039	15.644	0.037	38.264	0.040	0.83904	0.00340	2.0523	0.0031
6/25/2007	T 1345	Well 20	no	18.737	0.031	15.648	0.031	38.351	0.030	0.83517	0.00320	2.0469	0.0035
6/25/2007	T 1345(2)	Well 20	no	18.754	0.062	15.663	0.064	38.380	0.065	0.83516	0.0053	2.0465	0.0046
6/25/2007	T 1346	Well 21	no	18.581	0.014	15.626	0.014	38.193	0.014	0.84099	0.00170	2.0555	0.0015
	Sand and gravel wells												
6/25/2007	T 1359	Well 5	no	18.469	0.017	15.626	0.017	38.192	0.017	0.84606	0.00210	2.0679	0.0019
6/25/2007	T 1347	Well 6	no	18.617	0.030	15.656	0.030	38.345	0.033	0.84094	0.00370	2.0597	0.0030
6/25/2007	T 1347(2)	Well 6	no	18.632	0.036	15.663	0.036	38.364	0.034	0.84067	0.0031	2.0591	0.0033
6/25/2007	T 1350	Well 7	no	18.431	0.066	15.612	0.068	38.073	0.066	0.84705	0.00700	2.0657	0.0045
6/25/2007	T 1350(3)	Well 7	no	18.404	0.125	15.594	0.125	38.017	0.125	0.84731	0.0106	2.0657	0.0081
6/25/2007	T 1344	Well 8A	no	18.682	0.010	15.651	0.010	38.345	0.009	0.83776	0.00110	2.0526	0.0014
6/25/2007	T 1349	Well 9	no	18.438	0.050	15.634	0.051	38.167	0.050	0.84790	0.00530	2.0700	0.0040
6/25/2007	T 1358	Well 10	no	18.287	0.062	15.630	0.064	38.023	0.064	0.85468	0.00620	2.0792	0.0049
6/25/2007	T 1348	Well 11	no	18.108	0.033	15.591	0.031	37.862	0.032	0.86102	0.00380	2.0909	0.0032
6/25/2007	T 1356	Well 12	no	18.454	0.016	15.630	0.016	38.231	0.016	0.84696	0.00170	2.0717	0.0015
6/25/2007	T 1351	Well 14	no	18.495	0.028	15.635	0.030	38.213	0.027	0.84535	0.00320	2.0661	0.0025
6/25/2007	T 1351(2)	Well 14	no	18.484	0.008	15.626	0.009	38.205	0.008	0.84542	0.0013	2.0670	0.0016
6/25/2007	T 1355	Well 16	no	18.538	0.123	15.658	0.120	38.222	0.122	0.84462	0.01030	2.0618	0.0093

Date of			HC?		Pb Isotopic Ratios (2)								
Collection	MIT #	Well #	(1)	²⁰⁶ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁴ Pb	% SE	²⁰⁸ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁶ Pb	% SE	²⁰⁸ Pb/ ²⁰⁶ Pb	% SE
	Surface water samples												
8/30/2007	T 1383	SW-2	no	18.806	0.063	15.655	0.057	38.518	0.062	0.83245	0.0068	2.0482	0.0063
8/30/2007	T 1383(3)**	SW-2	no	18.814	0.017	15.678	0.021	38.492	0.020	0.83175	0.0022	2.0459	0.0018
8/30/2007	T 1384	SW-3	no	18.784	0.029	15.649	0.029	IC did not run		0.83307	0.0030		
8/30/2007	T 1384(3)**	SW-3	no	IC did not run									
	Surface water samples												
2/28/2008	T 1486	SW-1	no	18.936	0.230	15.786	0.232	38.777	0.232	0.83361	0.0144	2.0478	0.0135
2/28/2008	T 1486(2)	SW-1	no	18.841	0.120	15.715	0.13	38.562	0.12	0.83404	0.0120	2.0466	0.0106
2/28/2008	T 1487	SW-2	no	IC did not run									
2/28/2008	T 1487(2)	SW-2	no	18.846	0.21	15.670	0.25	38.533	0.23	0.83148	0.0172	2.0446	0.0174
2/28/2008	T 1488	SW-3	no	18.832	0.051	15.676	0.051	38.469	0.051	0.83241	0.0052	2.0427	0.0040

(1) Indicates presence of hydrocarbons in the sample.

(2) Measured ratios corrected for mass fractionation of 0.12 ± 0.03 ‰/a.m.u. based on replicate analyses of NBS-981; precision of ratios is $\leq 0.1\%$.

** Ammonium hydroxide extraction for sulfate-rich waters.

Shaded values indicate poor analyses. These values were not graphed.

Date of Collection	MIT #	Well #	HC? (1)	²⁰⁶ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁴ Pb	% SE	²⁰⁸ Pb/ ²⁰⁴ Pb	% SE	²⁰⁷ Pb/ ²⁰⁶ Pb	% SE	²⁰⁸ Pb/ ²⁰⁶ Pb	% SE
	Bedrock wells												
2/29/2008	T 1497	Well 13A		18.768	0.115	15.690	0.116	38.544	0.116	0.83601	0.0084	2.0537	0.0070
2/29/2008	T 1497(2)	Well 13A		18.769	0.058	15.669	0.059	38.469	0.060	0.83482	0.0066	2.0496	0.0055
2/29/2008	T 1497-1	Well 13B		18.610	0.072	15.680	0.074	38.306	0.075	0.84254	0.0063	2.0584	0.0048
2/29/2008	T 1500	Well 17		18.843	0.210	15.697	0.210	38.472	0.209	0.83300	0.0147	2.0417	0.0116
2/29/2008	T 1500(2)	Well 17		18.818	0.04	15.679	0.04	38.432	0.04	0.83317	0.0038	2.0423	0.0032
2/29/2008	T 1501	Well 18		IC did not run									
2/29/2008	T 1502	Well 19		18.676	0.045	15.639	0.046	38.267	0.046	0.83738	0.0042	2.0489	0.0036
2/29/2008	T 1503	Well 20		18.376	0.182	15.646	0.183	38.235	0.188	0.85140	0.0125	2.0807	0.0110
2/29/2008	T 1503(2)	Well 20		18.384	0.05	15.650	0.05	38.232	0.05	0.85129	0.0050	2.0797	0.0041
2/29/2008	T 1504	Well 21		18.297	0.075	15.682	0.074	38.156	0.077	0.85708	0.0057	2.0854	0.0056
	Sand and gravel wells												
2/29/2008	T 1489	Well 5		18.521	0.083	15.679	0.083	38.312	0.084	0.84654	0.0080	2.0685	0.0067
2/29/2008	T 1490	Well 6		18.586	0.03	15.637	0.03	38.263	0.03	0.84134	0.0034	2.0587	0.0025
2/29/2008	T 1491	Well 7		18.413	0.027	15.651	0.026	38.138	0.027	0.84998	0.0034	2.0713	0.0030
2/29/2008	T 1492	Well 8A		18.673	0.110	15.603	0.110	38.216	0.110	0.83562	0.0117	2.0466	0.0082
2/29/2008	T 1493	Well 9		18.539	0.046	15.653	0.047	38.205	0.049	0.84431	0.0048	2.0607	0.0040
2/29/2008	T 1494	Well 10		IC did not run									
2/29/2008	T 1495	Well 11		18.528	0.031	15.656	0.032	38.218	0.032	0.84496	0.0029	2.0627	0.0032
2/29/2008	T 1496	Well 12		18.609	0.223	15.695	0.215	38.363	0.211	0.84344	0.0203	2.0615	0.0198
2/29/2008	T 1496(2)	Well 12		18.530	0.004	15.622	0.004	38.146	0.005	0.84308	0.0011	2.0587	0.0016
2/29/2008	T 1498	Well 14		18.574	0.016	15.637	0.016	38.221	0.016	0.84184	0.0018	2.0577	0.0017
2/29/2008	T 1499	Well 16		18.470	0.038	15.646	0.037	38.182	0.038	0.84710	0.0032	2.0673	0.0027
	Surface water samples												
3/5/2008	T 1505	SW-1		18.834	0.031	15.653	0.030	38.504	0.031	0.83113	0.0030	2.0444	0.0024
3/5/2008	T 1506	SW-1a*		18.823	0.036	15.651	0.04	38.493	0.04	0.83147	0.0033	2.0450	0.0029
3/5/2008	T 1507	SW-2		18.860	0.044	15.655	0.05	38.492	0.05	0.83009	0.0049	2.0409	0.0038
3/5/2008	T 1508	SW-3		18.792	0.031	15.647	0.03	38.386	0.03	0.83261	0.0037	2.0426	0.0023

*SW-1a was sampled downstream from SW-1.

Table 4: Pb ID data , separated by sample type (table continues on next 2 pages).

Date of Collection	MIT #	Well #	Pb (ng) (1)	Sample weight (g)	Pb (ppb)
	Surface water samples				
6/25/2007	T 1341	SW-1	0.9465	0.9703	0.9755
6/25/2007	T 1342	SW-2	1.192	0.9727	1.225
6/25/2007	T 1342(2)	SW-2	0.9970	0.9993	0.9977 ⁽²⁾
6/25/2007	T 1343	SW-3	4.359	0.9997	4.361
	Bedrock wells				
6/25/2007	T 1357	Well 13B	1.217	0.9979	1.220
6/25/2007	T 1353	Well 17	0.8130	0.9788	0.8307
6/25/2007	T 1352	Well 18	0.5823	0.9864	0.5903
6/25/2007	T 1354	Well 19	0.7640	0.9968	0.7665
6/25/2007	T 1345	Well 20	0.3240	0.9994	0.3242
6/25/2007	T 1345(2)	Well 20	0.288	1.0012	0.288
6/25/2007	T 1346	Well 21	0.3773	0.9998	0.3774
	Sand and gravel wells				
6/25/2007	T 1359	Well 5	4.589	0.9973	4.601
6/25/2007	T 1347	Well 6	0.7461	0.9805	0.7609
6/25/2007	T 1347(2)	Well 6	0.275	0.9995	0.2746
6/25/2007	T 1350	Well 7	0.2263	0.9951	0.2274
6/25/2007	T 1350(3)	Well 7			
6/25/2007	T 1344	Well 8A	3.476	0.9981	3.483
6/25/2007	T 1349	Well 9	1.722	0.9971	1.727
6/25/2007	T 1358	Well 10	0.2933	0.9974	0.2941
6/25/2007	T 1348	Well 11	0.3799	0.9966	0.3812
6/25/2007	T 1356	Well 12	3.002	0.9967	3.012
6/25/2007	T 1351	Well 14	1.834	0.9870	1.858
6/25/2007	T 1351(2)	Well 14	1.738	0.9837	1.767
6/25/2007	T 1355	Well 16	0.3208	0.9936	0.3229

(1) Nanograms Pb extracted from sample determined by ID using ²⁰⁸Pb spike.

(2) assumes IC from T 1342

Date of Collection	MIT #	Well #	Pb (ng) (1)	Sample weight (g)	Pb (ppb)
	Surface water samples				
8/30/2007	T 1383	SW-2	59.93	5.08	11.8
8/30/2007	T 1383(3)**	SW-2			
8/30/2007	T 1384	SW-3	51.11	5.09	10.04 ⁽³⁾
8/30/2007	T 1384(3)**	SW-3			
	Surface water samples				
2/28/2008	T 1486	SW-1	0.2989	1.0046	0.2975
2/28/2008	T 1486(2)	SW-1			
2/28/2008	T 1487	SW-2	0.2795	1.0039	0.2784 ⁽⁴⁾
2/28/2008	T 1487(2)	SW-2			
2/28/2008	T 1488	SW-3	1.704	0.99876	1.706

(3) assumes IC from T1343

(4) assumed IC from T 1487(2)

** Ammonium hydroxide extraction for sulfate-rich waters.

Date of Collection	MIT #	Well #	Pb (ng) (1)	Sample weight (g)	Pb (ppb)
	Bedrock wells				
2/29/2008	T 1497	Well 13A	0.569	5.0129	0.1134
2/29/2008	T 1497(2)	Well 13A			
2/29/2008	T 1497-1	Well 13B	0.96	5.0185	0.1914
2/29/2008	T 1500	Well 17	5.849	5.0367	1.161
2/29/2008	T 1500(2)	Well 17			
2/29/2008	T 1501	Well 18	3.221	5.0315	0.6401 ⁽⁵⁾
2/29/2008	T 1502	Well 19	2.018	5.0308	0.4010
2/29/2008	T 1503	Well 20	0.989	5.0180	0.1971
2/29/2008	T 1503(2)	Well 20			
2/29/2008	T 1504	Well 21	0.624	5.0190	0.1244
	Sand and gravel wells				
2/29/2008	T 1489	Well 5	2.215	5.0441	0.4390
2/29/2008	T 1490	Well 6	1.614	5.0360	0.3205
2/29/2008	T 1491	Well 7	1.317	5.0221	0.2623
2/29/2008	T 1492	Well 8A	0.593	5.0291	0.1180
2/29/2008	T 1493	Well 9	0.678	5.0293	0.1347
2/29/2008	T 1494	Well 10	0.862	5.0248	0.1715 ⁽⁶⁾
2/29/2008	T 1495	Well 11	1.554	5.0195	0.3096
2/29/2008	T 1496	Well 12	ID did not run	5.0158	
2/29/2008	T 1496(2)	Well 12	16.43	5.0527	3.251
2/29/2008	T 1498	Well 14	3.215	5.0174	0.6408
2/29/2008	T 1499	Well 16	1.330	5.0249	0.2647
	Surface water samples				
3/5/2008	T 1505	SW-1	4.275	5.0030	0.8545
3/5/2008	T 1506	SW-1a	4.937	5.0191	0.9837
3/5/2008	T 1507	SW-2	2.875	5.0150	0.5732
3/5/2008	T 1508	SW-3	7.443	5.0010	1.488

(5) assumes IC from T 1352

(6) assumes IC from T 1358

Table 5: U IC data, separated by sample type.

Date of collection	MIT #	Well #	$^{234}\text{U} / ^{238}\text{U}$ raw	% SE	$^{235}\text{U} / ^{238}\text{U}$ raw	% SE	$^{236}\text{U} / ^{238}\text{U}$ raw	% SE	$^{234}\text{U} / ^{238}\text{U}$ N*	$^{235}\text{U} / ^{238}\text{U}$ N*	$^{236}\text{U} / ^{238}\text{U}$ N*	Alpha** 235/238
6/25/2007	Surface water samples											
	T 1341	SW-1	did not run									
	T 1342	SW-2	did not run									
6/25/2007	T 1343	SW-3	5.840E-05	0.645	0.007290	0.088	no data		5.7954E-05	0.00725268		-0.00170
6/25/2007	Bedrock wells											
	T 1357	Well 13B	2.30E-04	0.548	0.007211	0.128	no data		2.3172E-04	0.00725268		0.00193
	T 1353	Well 17	8.42E-05	0.594	0.007295	0.074	4.0E-07	42	8.3526E-05	0.00725268	3.9647E-07	-0.00192
	T 1352	Well 18	1.93E-04	0.611	0.007257	0.070	no data		1.9331E-04	0.00725268		-0.00018
	T 1354	Well 19	2.08E-04	0.409	0.007288	0.069	8.1E-07	19	2.0665E-04	0.00725268	8.0546E-07	-0.00163
	T 1345	Well 20	9.73E-05	0.411	0.007285	0.041	1.5E-07	57	9.6680E-05	0.00725268	1.5378E-07	-0.00149
	T 1346	Well 21	1.09E-04	0.399	0.007288	0.051	6.9E-07	16	1.0818E-04	0.00725268	6.8548E-07	-0.00163
	Sand and gravel wells											
6/25/2007	T 1359	Well 5	5.50E-05	1.874	0.007225	0.165	6.3E-06	13	5.5328E-05	0.00725268	6.3282E-06	0.00129
6/25/2007	T 1347	Well 6	5.49E-05	0.510	0.007271	0.043	5.1E-07	15	5.4722E-05	0.00725268	5.0717E-07	-0.00082
6/25/2007	T 1350	Well 7	5.68E-05	0.548	0.007394	0.053	1.0E-06	11	5.5369E-05	0.00725268	1.0119E-06	-0.00637
6/25/2007	T 1344	Well 8A	5.61E-05	0.943	0.007308	0.070	2.7E-06	9	5.5540E-05	0.00725268	2.7282E-06	-0.00254
6/25/2007	T 1344(2)	Well 8A	5.32E-05	1.485	0.007287	0.119	no data		5.2851E-05	0.00725268		-0.00157
6/25/2007	T 1349	Well 9	5.30E-05	0.522	0.007281	0.049	5.7E-07	13	5.2762E-05	0.00725268	5.7154E-07	-0.00127
6/25/2007	T 1358	Well 10	6.49E-05	0.846	0.007200	0.076	no data		6.5493E-05	0.00725268		0.00246
6/25/2007	T 1348	Well 11	5.42E-05	0.491	0.007290	0.046	1.1E-07	39	5.3849E-05	0.00725268	1.1362E-07	-0.00169
6/25/2007	T 1356	Well 12	5.90E-05	0.763	0.007275	0.079	1.3E-06	18	5.8774E-05	0.00725268	1.3090E-06	-0.00101
6/25/2007	T 1351	Well 14	5.64E-05	0.437	0.007346	0.044	1.8E-07	39	5.5472E-05	0.00725268	1.7847E-07	-0.00424
6/25/2007	T 1355	Well 16	6.82E-05	0.745	0.007272	0.057	1.0E-06	26	6.7995E-05	0.00725268	1.0175E-06	-0.00089

*Fractionation corrected data

**Fractionation factor per atomic mass unit

Standards and Replicates

Uncertainties associated with mass spectrometry were evaluated by measuring standards of Sr, Pb, and U. The reproducibility of measurements for the samples were evaluated by measuring sample replicates.

Standards

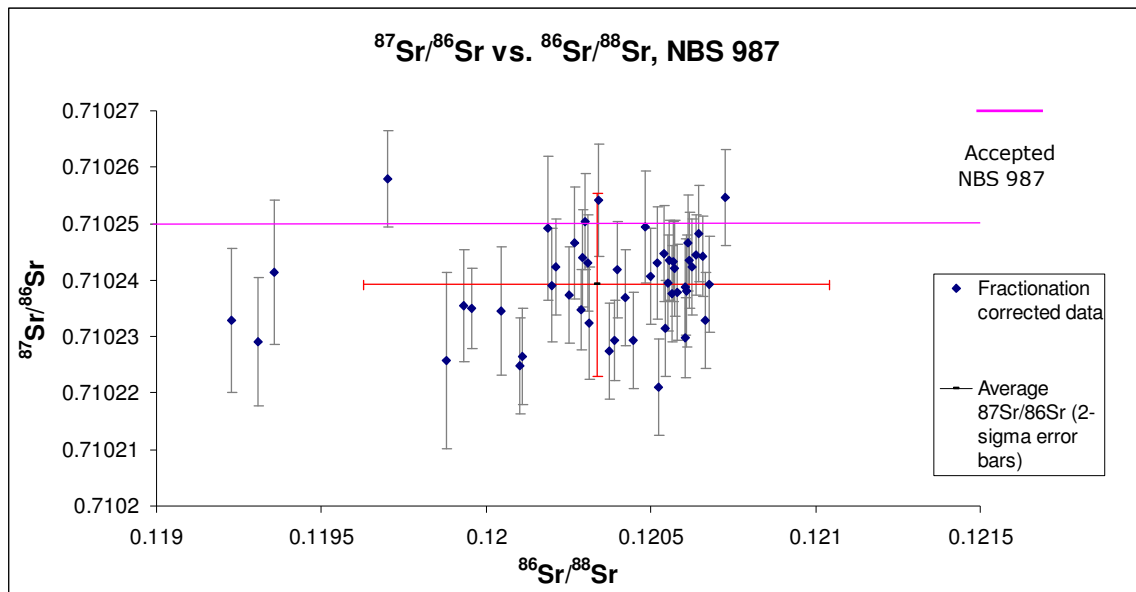


Figure 14: A plot of $^{87}\text{Sr}/^{86}\text{Sr}$ vs. $^{86}\text{Sr}/^{88}\text{Sr}$ for the Sr standard NBS-987. Fractionation-corrected data and the average value from the analyses are shown with 2- σ error bars (all data were fractionation-corrected using $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and the exponential fractionation law). The accepted value of NBS-987 (0.710250) is depicted by the horizontal line (Thirlwall, 1991).

Fifty measurements of the Sr standard NBS-987 yielded a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.710239 ± 0.000016 (2 σ error). Compared to the "accepted" NBS-987 value of 0.710250, the measured value is accurate to 0.001522%. The measured value of NBS-987 varies between different labs, so for the purposes of interlaboratory comparisons, one could normalize all the data to the accepted value .

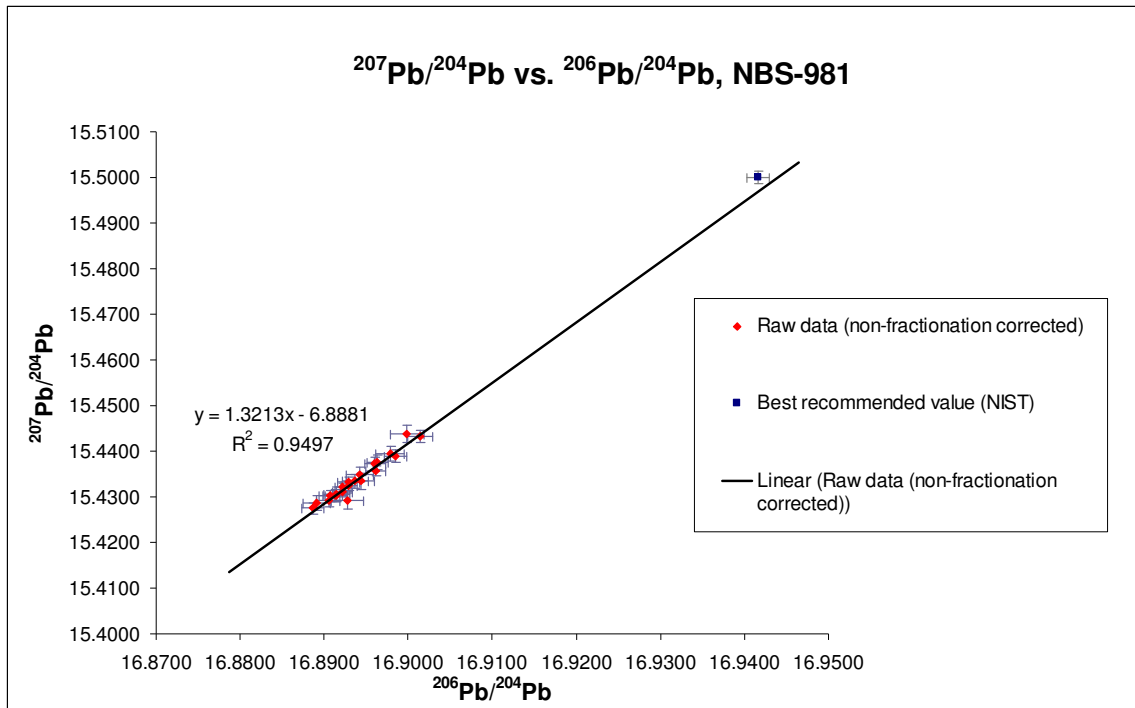


Figure 15: A plot of $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ for the Pb standard NBS-981 (Baker et al., 2004). The measured values (raw data) are displayed with 2- σ error bars.

Results from 22 measurements of the NBS-981 Pb standard yielded $^{207}\text{Pb}/^{204}\text{Pb}$ and $^{206}\text{Pb}/^{204}\text{Pb}$ ratios of 15.4337 ± 0.0092 and 16.8938 ± 0.0068 , respectively (both with 2 σ error). All data were fractionation-corrected using a value of 0.12% per a.m.u.

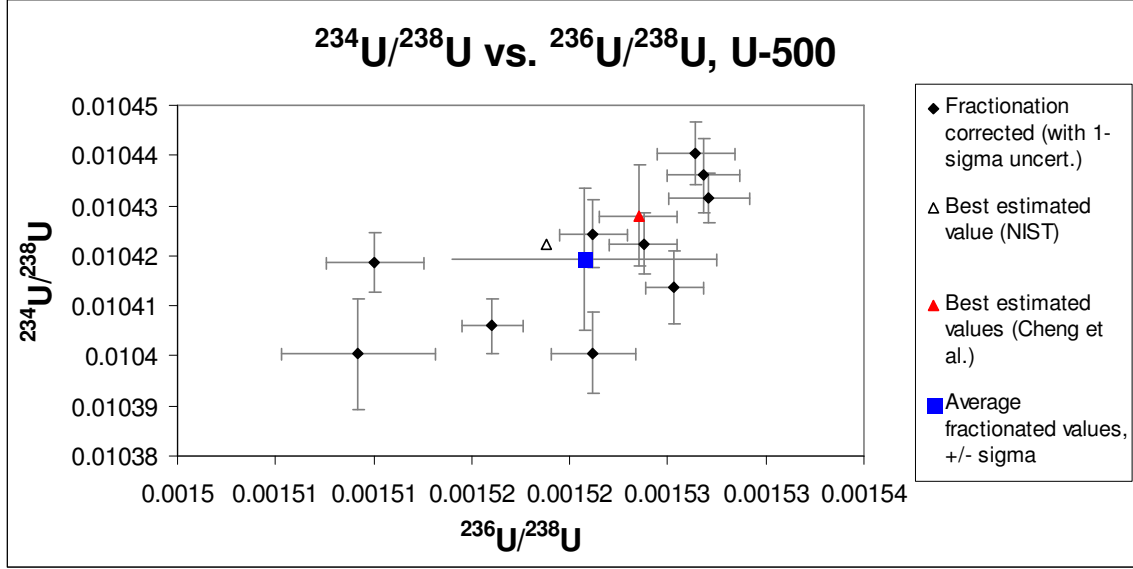


Figure 16: A plot of $^{234}\text{U}/^{238}\text{U}$ vs. $^{236}\text{U}/^{238}\text{U}$ for the U standard U-500. Fractionation-corrected values and the average measured value are shown with 1- σ error bars. The best estimated values for U-500 from two different sources (NIST, 1981 and Cheng et al., 2000) are also shown for comparison.

The $^{234}\text{U}/^{238}\text{U}$ ratio from ten measurements of the U standard U-500 was $0.010400968 \pm .000091505$ (2σ error), which is within 1σ of the NIST and Cheng et al. values.

Replicates

The average difference between replicate pairs of isotopic composition measurements are comparable to or less than the 2- σ precision of the respective standards.

Table 6: a comparison between the precision of sample measurements and those of the standards.

	Average difference between sample replicates	2- σ precision of standards
$^{87}\text{Sr}/^{86}\text{Sr}$ (4 replicates)	0.0000083	0.000016
$^{206}\text{Pb}/^{204}\text{Pb}$ (4 replicates)	0.013	0.0068
$^{207}\text{Pb}/^{204}\text{Pb}$ (4 replicates)	0.013	0.0092
$^{234}\text{U}/^{238}\text{U}$ (1 replicate)	0.0000027	0.0000915

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