A ^{36}Cl Investigation of Ground-water Flow in the Morrison Formation of the San Juan Basin, New Mexico.

by

B. Geoffrey Jones

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ABSTRACT

Determinations have been made of the isotopic ratio $^{36}Cl/Cl$ in groundwater samples taken from wells penetrating the Morrison Formation in the San Juan Basin, New Mexico. Wells were sampled for analysis over the period 1986-1989 as part of the U.S. Geological Survey, San Juan Basin RASA program. Samples from a few wells penetrating the Dakota Sandstone and other wells penetrating the Gallup Sandstone have also been analyzed. Measurements were made on the University of Rochester, Nuclear Structure Research Lab, tandem accelerator mass spectrometer. Measured ratio data has been considered in conjunction with chloride concentration data for samples collected at the wells simultaneously.

A basic ^{36}Cl mass balance development of standard dating equations has been presented, and the approach used to develop the equations has been explored for extended applications. Methods for calculating the secular equilibrium isotopic ratio were examined using a simple digital program, CLSEC, provided herein. Geochemical data from the Morrison Formation were sought and used to calculate possible rock and rock-water secular equilibrium ratio values.

The usual ^{36}Cl decay equations produced variable results, and a diagrammtic approach was developed to assist with chloride data interpretation. The diagrammatic approach proved sucessful, and a complex system of multiple source decays and mixing of waters is described for the Morrison aquifer system. Mixing endmembers are suggestive of ion filtration, ^{36}Cl buildup, and mixing of discharge waters. Comparisons of chloride data to other available data are outlined, but remain incomplete.

TABLE OF CONTENTS

BACKGROUND

METHODS AND RESULTS

DISCUSSION

APPENDIX A: NEW MEXICO TECH ISOTOPE HYDROLOGY LAB PROCEDURES USED

APPENDIX B: CLSEC COMPUTER FILES

LIST OF FIGURES

LIST OF TABLES

INTRODUCTION

The San Juan Basin is a large structural basin located in northwest New Mexico that contains a number of major ground-water aquifer systems. Surface water in the area is fully appropriated, and the increasing water demands of mining, electric power companies, municipalities, and American Indian communities has intensified interest in the ground-water resources of the basin. In response to this interest, the U.S. Geological Survey is conducting studies of the basin as part of its nation-wide Regional Aquifer System Analysis (RASA) program. The San Juan Basin RASA includes the collection, organization, and analysis of geologic, hydrologic, and hydrochemical data from the basin. The effort is intended to provide a conceptual as well as quantitative understanding of water movement and water quality in the major aquifer units. The increased understanding will help provide effective management of the area ground-water resources.

This radioactive isotope investigation is part of the San Juan Basin RASA effort. Radioactive isotopes can frequently assist in the study of regional aquifer systems, and they have been commonly applied to basin settings (e.g. Bentley et al., 1986; Phillips et al., 1984). Not only can sources of water in the system be potentially identified by characteristic isotope compositions, but radioactive decay introduces a time variable that can be compared with rates of chemical transport. If the aquifer system hydrology and isotope chemical behavior are well enough understood, the decay of the isotope provides an independent measure of travel time and thereby regional aquifer hydrologic properties.

The main objective of the current study is to investigate the deeper potable aquifers of the San Juan Basin using the naturally occurring radioactive isotope of chlorine, ^{36}Cl. The deeper groundwaters of the basin are old enough to be dead of ^{14}C (>60,000 years), and hydrologic modeling in progress suggests that water in the central regions of the deeper aquifers exceeds 1 million years old. Chlorine-36, with its long half-life and conservative (rarely chemically reactive) anionic behavior, is suitable for such applications.

The aquifers studied include the Jurassic Morrison Formation and, to a lesser extent, the younger Cretaceous Dakota and Gallup Sandstones. The Morrison is artesian and a primary water supply in parts of the basin, even when located at depths below 4500 feet. Our research efforts were concentrated on those areas in the northwest part of the basin where the Morrison is a principal water supply aquifer and away from the complications of major historic ground-water production.

BACKGROUND

36Cl in Old Groundwaters

The occurrence of ^{36}Cl in the terrestrial environment and its application as an investigative tool in the study of various dynamic hydrologic processes has been well documented in a number of recent articles. The processes involve a wide range of geoscience applications, and have been summarized in a book chapter (Bentley et al., 1986a). The application of ^{36}Cl to old groundwater in sedimentary basins has been summarized in an additional article (Phillips et al., 1986a). These works, as well as the more detailed descriptions of the application of ^{36}Cl to the Great Artesian Basin, Australia (Bentley et al., 1986b), and the Milk River Aquifer, Canada (Phillips et al., 1986b), should be consulted for a complete description of ^{36}Cl as a groundwater investigation tool. Herein, a general description is provided, with emphasis on the aspects important for this study.

The concept of using radiometric tracers to study groundwater systems is not new, and tritium and ^{14}C have seen considerable application in shallow, relatively young groundwater systems (e.g. Pearson and White, 1967; Egboka et al., 1982). This includes the ^{14}C of the San Juan Basin shallow aquifer systems, which have been studied by Phillips and Tansey (1984), and related to the stable isotope data in Phillips et al. (1986c).

In deep sedimentary basin systems, however, the groundwater is often slow moving and the waters are very old. The limiting 5730 year half-life makes ^{14}C inapplicable, regardless of the analytical methods available. Chlorine-36, with its 301,000 year half-life, is better suited, particularly considering its relatively well-behaved anionic chemistry. The chloride ion is very hydrophillic and does not tend to precipitate or sorb in groundwater systems except under low pH conditions (Feth, 1981).

The difficulty in using ^{36}Cl lies in the analytical determination of the quantity present. As the ratio of radioactive to stable chloride is typically on the order of 10^{-15} to 10^{-12} there is usually little ^{36}Cl present, making accurate measurement onerous. With the small quantities typically present, indirect methods of measuring ^{36}Cl based on measuring beta particles from ^{36}Cl decay have proved too inaccurate for the lower levels needed to study deep groundwaters. The breakthrough came in the late 1970's with the development of tandem accelerator mass spectrometry (TAMS) analysis (Elmore et al., 1979). This technique allows direct measurement of the number of ^{36}Cl atoms present, and is companion to a number of developments in the area of accelerator mass spectrometry (AMS) for measurement of long-lived radioisotopes. The current detection limit using TAMS analysis is a $^{36}Cl/Cl$ ratio (radioactive/stable) of about 1×10^{-15}.

Chlorine-36 is produced in the atmosphere and in rocks or water near the land surface by cosmic-ray activation of various atomic species (Davis and Schaeffer, 1955; Yokoyama et al., 1977). In the deeper subsurface, ^{36}Cl is produced by thermal-neutron flux activation of ^{35}Cl. This hypogene production is induced by the generally modest neutron flux in the subsurface which arises from decay of the uranium and thorium series radionuclides (Fabryka-Martin, 1988). In addition to the natural production of ^{36}Cl, human activities have also released large quantities of ^{36}Cl to the environment. The most significant of such activities was the atmospheric testing of thermonuclear weapons near the ocean, which served to activate sea water chloride. This global "bomb pulse" of ^{36}Cl is very similar to that observed and used in tritium studies of shallow groundwater (Bentley et al., 1982).

Fallout from precipitation and dry aerosol deposits containing atmospheric chloride contribute to the meteoric ^{36}Cl/Cl ratio. This ratio, anthropogenic factors aside, can be described by the superposition of the latitude dependence of ^{36}Cl meteoric fallout and the near costal dependence of stable chloride meteoric fallout. Bentley et al. (1986) present the results of this determination for the continental United States (Fig. 1). Production of ^{36}Cl by cosmic-ray activation of atomic species in rocks and water close to the ground surface is referred to as epigene production. Rock buildup is a function of composition, latitude, sunspot cycle, altitude, depth of burial, and time of exposure (Phillips et al., 1986d). Equilibrium in the ^{36}Cl/Cl ratio as a function of time (secular equilibrium) is reached when

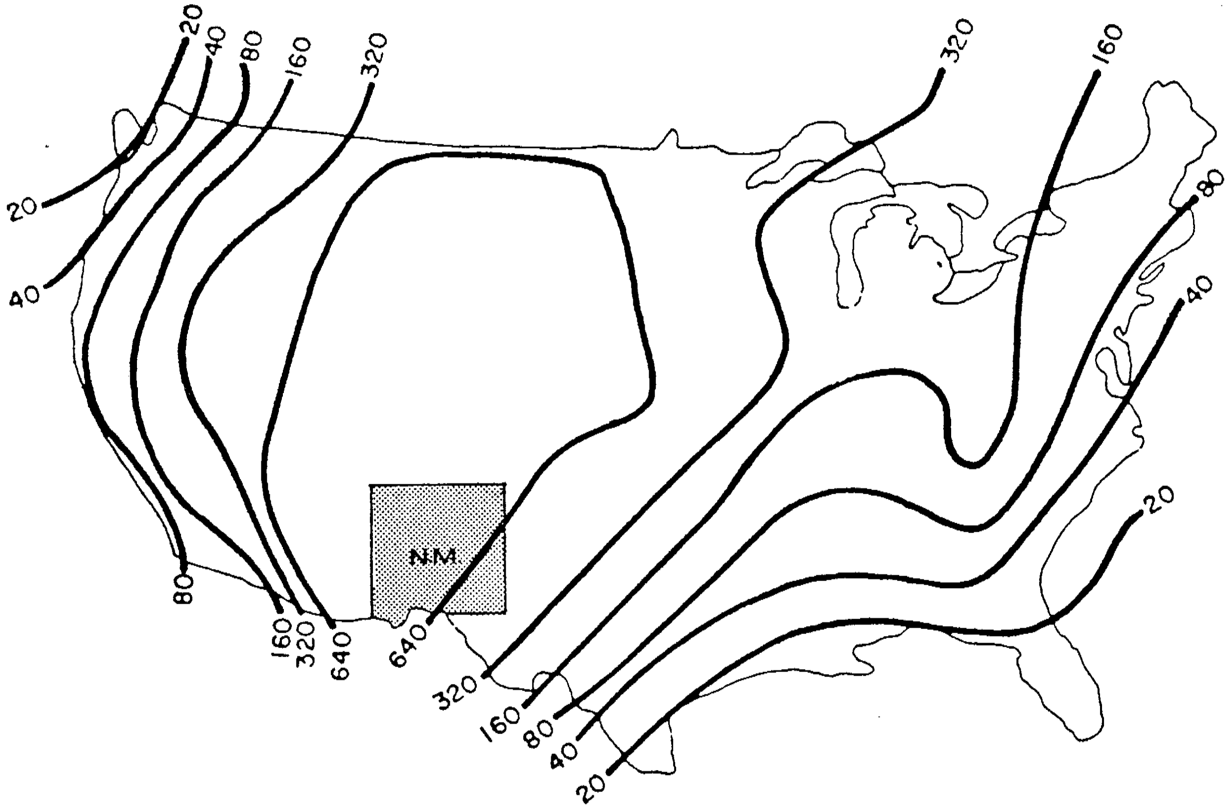

Figure 1. Calculated $^{36}Cl/Cl$ ratios (x 10^{15}) in precipitation and dry fallout over the United States (Bentley et al., 1986).

the rate of production and rate of decay of the ^{36}Cl become equal. However, this typically takes at least five half-lives (about 1.5 x 10^6 years) and most rocks are weathered away before reaching that age.

Hydrogeology of the San Juan Basin

The hydrogeology of the San Juan Basin has been studied in numerous reports, and the U.S. Geological Survey (USGS) is currently compiling the data available. Unfortunately much of the data available was collected some time ago, and the reliability of that data for other applications is not always known. Great effort is being made to distinguish the accuracy and reliability of the data available for cross purposes. Current hydrogeologic understanding is based largely on a study, Stone et al., 1983, published by the New Mexico Bureau of Mines and Mineral Resources in cooperation with the USGS and the New Mexico State Engineer. New hydrogeologic data collection for the San Juan RASA is being restricted to a few wells in relatively lesser known areas of the central basin. Other past efforts will be mentioned in context.

The San Juan Basin is a large structural basin filled with sedimentary deposits that reflect the transgressive-regressive and arid continental sequences responsible for their deposition. The basin is located primarily in the northernmost corner of the state of New Mexico and is generally surrounded by structural uplifts and related high elevations (Fig. 2). These uplift

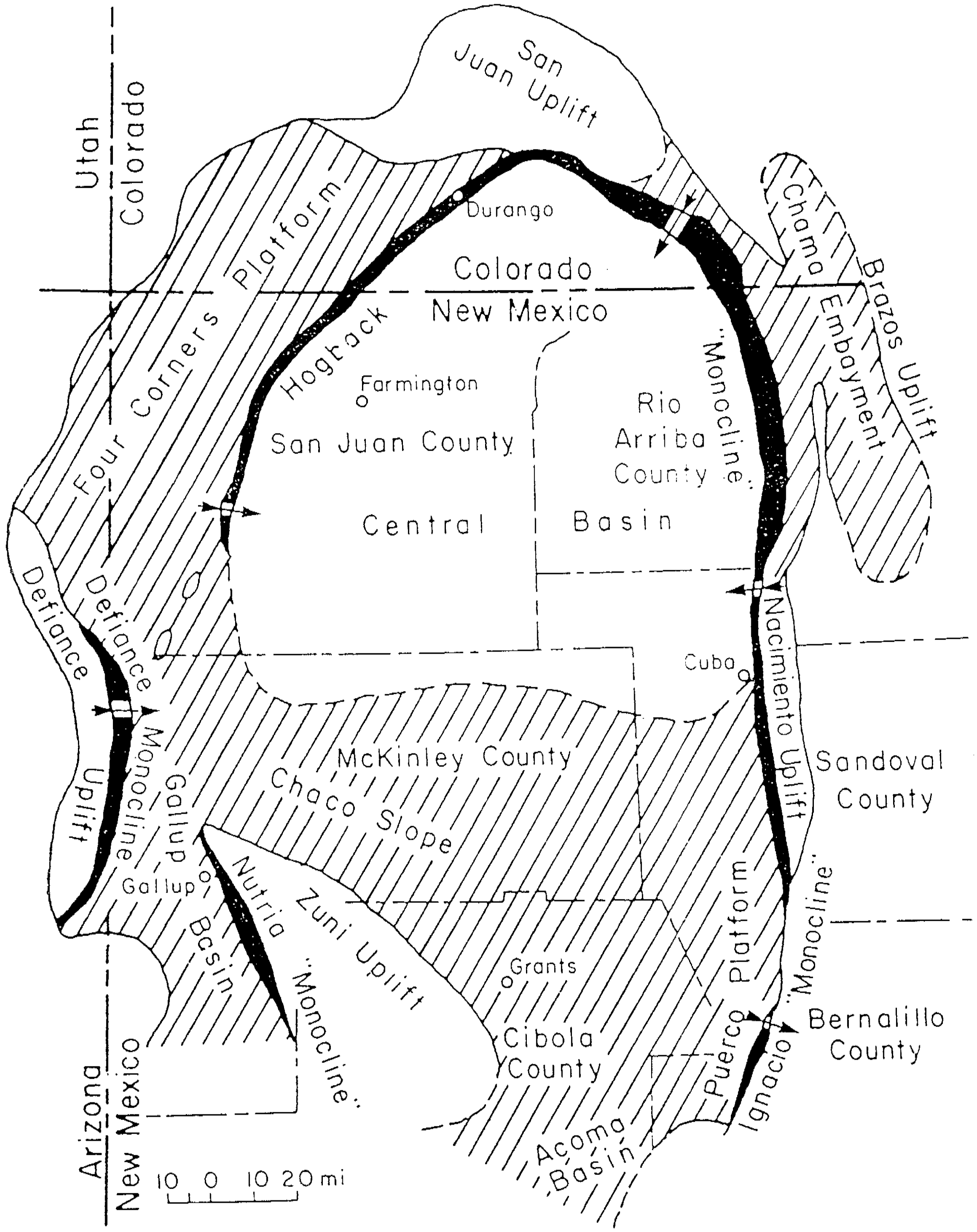

Figure 2. Structural elements recognized in the San Juan Basin (Stone et al., 1983; modified from Kelly, 1951).

structures include the Defiance Uplift and associated Chuska Mountains on the west side, the Zuni Uplift and associated Mount Taylor, Cebolleta, and Zuni Mountains on the south, the Nacimiento Uplift and the associated Sierra Nacimiento Mountains on the east, and the San Juan Uplift and associated San Juan Mountains on the north. The deep central part of the basin is delimited to the north by the Hogback monocline, a large arcuate shaped structure with significant surface expression.

The stratigraphic section for the Triassic to Tertiary periods is shown on Fig. 3 in a generalized north-south cross section. As might be expected, the units regarded as aquifer units are correlative with the occurrences of significant sandstone bodies; however, a given sandstone body or unit might vary tremendously in water yield and quality from one part of the basin to another. Not demonstrated by the time-stratigraphic type relationship shown in Fig. 3, the basin is asymmetric in cross-section and shows a maximum depth of sediments at the basin axis, close to the San Juan River, towards the northern margin of the study area.

Surface water flow through the basin is controlled by the topography. The continental divide passes through the basin as a subdued topographic expression, and approximately 4/5 of the basin drains north to the San Juan River, whereas the other 1/5 drains south through the Rio Puerco to the Rio Grande. A relatively small area in the southwest extreme of the basin is

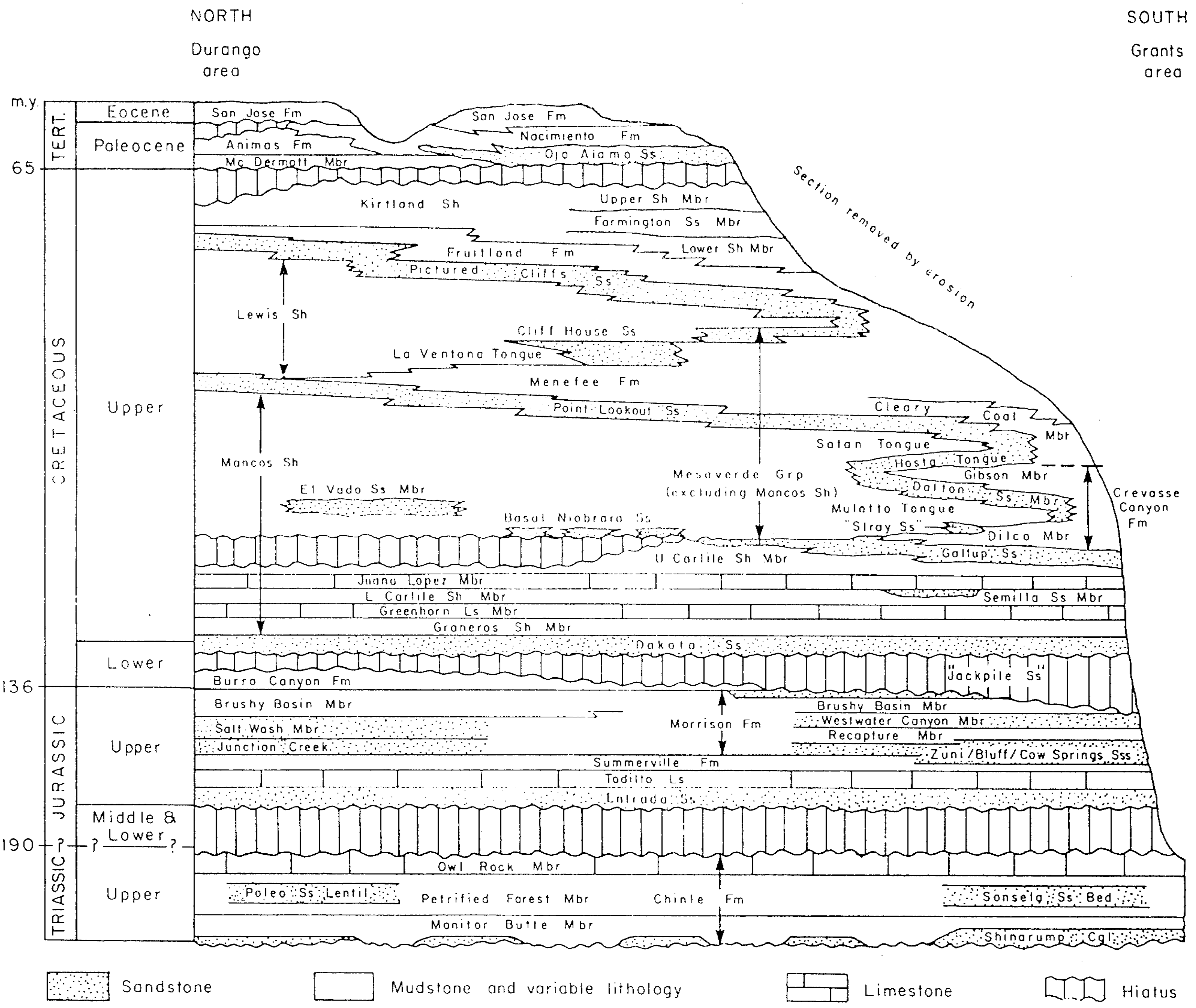

Figure 3. Time-stratigraphic framework and nomenclature (Triassic-Tertiary) of the San Juan Basin (Stone et al., 1983; modified from Molenaar, 1977).

drained to a third system, flowing west through the Puerco River (not the same as the Rio Puerco) to the Little Colorado River. This third drainage has been made more significant in the recent past by the large quantities of effluent it carries from uranium-mine dewatering operations.

The topographic low of the basin is at the San Juan River as it leaves the state in the Four Corners area. Precipitation is greatest within the higher elevation areas of the basin, close to its boundary, and groundwater recharge is probably low in the central more arid regions. The source of summer moisture generally is convective storms originating from the Gulf of Mexico; the source of winter precipitation is frontal storms from the Pacific Ocean.

The principal source of water in the San Juan Basin is groundwater obtained from wells completed in surficial valley-fill deposits of Quaternary age and sandstones of Tertiary, Cretaceous, Jurassic and Triassic age (Figure 3). The Tertiary and younger units of the central basin have been studied using the radioactive isotope ^{14}C (Phillips et al., 1986c) and will not be further considered in this study. Flow in the lower units is generally believed to be from the recharging basin edges, where all the major aquifer units outcrop, to discharge points along the San Juan River and Rio Puerco. The aquifer units are confined throughout the non-outcrop part of the study area by thick sections of shale, and wells away from the outcrop regions are commonly flowing artesian wells.

Withdrawal of groundwater varies across the basin in terms of both the quantities of water pumped and the aquifers that are used. In the southern part of the basin water is withdrawn from a number of different units, mostly of pre-Tertiary age. The withdrawals are large and are associated with uranium-mine operations and municipal water supply. Water level declines from this pumping have been greater than 200 feet (Stone et al., 1983). In the central part of the basin, water is withdrawn primarily from the Tertiary units for low-volume stock and domestic use, although some larger withdrawals are made for coal reclamation purposes. A few central basin wells, such as that at the Burnham Trading Post, go to greater depths to tap the better quality waters of the Jurassic Morrison Formation, however the Morrison is used more on the western side of the basin, on the Navajo Reservation, where exploratory uranium drilling has left a number of flowing wells now used for stock and domestic purposes.

The layer-cake geology of the pre-Tertiary sedimentary units has led to an initial hydrologic interpretation of largely lateral flow from the outcrop recharge areas to the river discharge areas (see Fig. 4). This interpretation is supported by the occurrence of low permeability units between major aquifers, and by an investigative modeling study indicating low vertical flow rates (Frenzel and Lyford, 1982). The lithologies and hence the hydrologic properties of many of the individual units tend to be

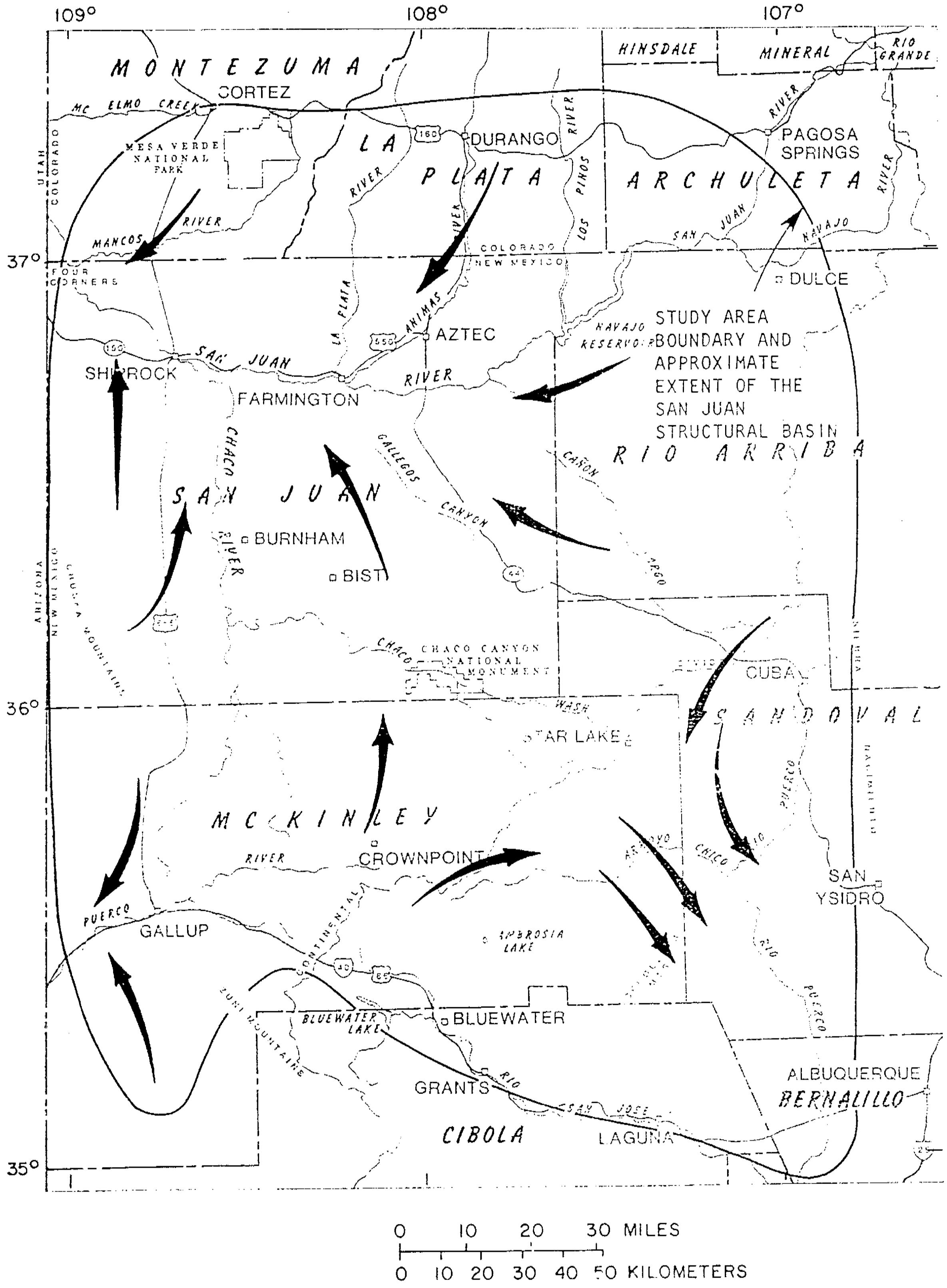

Figure 4. Generalized pattern of groundwater low (arrows) in rocks of Jurassic and Cretaceous age (Frenzel and Lyford, 1982).

somewhat lenticular or discontinuous, but relative to the differences between aquifer and aquitard units Frenzel and Lyford (1982) viewed this variation as minor. An additional study regarding the modeled hydraulic head distribution in Morrison is in progress, and preliminary results resemble the directions of flow indicated on Figure 4.

The occurrence of some amount of vertical flow in the system is inevitable. There are, in places, large hydraulic head differences between aquifer units that could be driving vertical flow, but the continued existence of such head differences also serves to indicate a certain pressure independence between the units. This independence is also suggested by water quality differences. Fracturing near geologic structures and past volcanic intrusives may serve as conduits for vertical flow (Stone et al.,1983). This is particularly probable near the Hogback Monocline, the Rio Puerco fault zone, and possibly near the Cebolleta Mountains. Although not specifically mentioned in the works reviewed, abandoned wells can also serve as such conduits, and there have been a great number and variety of exploratory wells drilled in the basin.

APPROACH

It is helpful to consider the application of ^{36}Cl to groundwater studies from an atomic mass balance point of view. If the hydrodynamic system and geochemical conditions are such that the movement of chloride in the groundwater can be well represented by a piston flow approximation of flow from one point, call it the recharge point, to another, call it the sample point, then the mass balance of ^{36}Cl atoms can be quantitatively described. Assuming no significant additional chloride sources or sinks, the mass of ^{36}Cl at the sample point is the summation of the ^{36}Cl still left after exponential decay of the recharge ^{36}Cl mass and the ^{36}Cl added from hypogene production. Note that decay is an exponential function of the half-life of ^{36}Cl and the residence time, where the residence time is the time required for the water to move in piston fashion from the recharge to sample points. This ^{36}Cl atomic mass balance can be generalized by the equation:

$$ {}^{36}M_s = {}^{36}M_o \cdot e^{-\lambda t} + {}^{36}M_{se} \tag{1a} $$

where, $^{36}M_s$ is the mass of ^{36}Cl within an elemental water volume at the sample point, $^{36}M_o$ is the mass of ^{36}Cl within an elemental water volume at the recharge point, $^{36}M_{se}$ is the mass of ^{36}Cl contributed by hypogene production, and $e^{-\lambda t}$ is that fraction of the recharge mass that has not yet radioactively decayed.

The concentration of ^{36}Cl is not measured directly, as in the mass terms presented in equation 1. Rather, the isotopic ratio of ^{36}Cl to stable chloride is the determination made. The ^{36}Cl mass present in a sample volume is a product of the ^{36}Cl concentration and the elemental volume at that point. The ^{36}Cl concentration is a product function of the measured $^{36}Cl/Cl$ ratio and the chloride concentration. If the chloride concentration is measured in mg/L the atomic concentration of ^{36}Cl can be expressed:

$$1b) \qquad {}^{36}Cl(atoms/L) = R \bullet Cl^{-}(mg/L) \bullet k$$

where R is the measured isotopic ratio, Cl^- is the measured chloride concentration, and k is a conversion factor for the given measurement units equal to 1.6988×10^4.

Equation 1a can be rewritten in terms of R and Cl^-, and the constant, k, of equation 1b can be divided through. In addition, consider that the hypogene contribution of ^{36}Cl is that mass of decayed recharge chloride which has achieved secular equilibrium with respect to the saturated porous media. With these relationships in mind, atomic mass balance is still conserved with the equation:

$$2) \qquad R \bullet C \bullet V = V_o \bullet R_o \bullet C_o \bullet e^{-\lambda t} + V_o \bullet R_{se} \bullet C_o (1 - e^{-\lambda t})$$

where: R, C, and V are the $^{36}Cl/Cl$ ratio, total chloride concentration, and elemental volume at the sample point; R_o, C_o, and V_o

are the $^{36}Cl/Cl$ ratio, total chloride, and elemental volume at the recharge point; R_{se} is the secular equilibrium $^{36}Cl/Cl$ ratio of the saturated porous media (the calculation of R_{se} is further addressed in the results section and Appendix B); $e^{-\lambda t}$ is as before; and $(1-e^{-\lambda t})$ is the fraction of the secular equilibrium ^{36}Cl mass that has accumulated.

Equation 1a, upon which equation 2 is based, does not account for the presence of additional chloride sources or sinks. Therefore, in equation 2, any changes in the concentration are a direct result of relative changes in the elementary pore volume. In this case, the product VC is equal to the product V_oC_o and these terms cancel. The resultant equation is a simple atomic mass balance based on the $^{36}Cl/Cl$ ratios (Bentley, et al., 1986a), as follows:

$$(3) \qquad R = R_o \cdot e^{-\lambda t} + R_{se}(1 - e^{-\lambda t})$$

Equation 3 may be applied to any case where the $^{36}Cl/Cl$ ratio follows simple decay, regardless of processes that might serve to change the water volume. Without the presence of additional chloride sources or sinks, mass balance of ^{36}Cl is still conserved even though the concentration history may be unknown. With a volume change, the concentration of both the radioactive and stable chloride species would both be changed in inverse proportion to the volume change, and the ratio would be unaffected. Equation 3, therefore, is applicable for simple decay even where additional processes such as evaporation, transpiration, and ion-filtration are working to change water volumes and chloride concentrations. Hence, this widely applicable equation shows a complete insensitivity to the often slightly variable total chloride concentration of the recharge water.

The residence or decay time in equation 3 associated with the piston travel from the recharge point to the sample point may be solved for. Assuming that the time to develop the ^{36}Cl ratio at the recharge point is insignificant, this residence time can be considered the water "age" (Bentley et al., 1986a; 1986b):

$$(4) \qquad t = \frac{-1}{\lambda} \ln \frac{R - R_{se}}{R_o - R_{se}}$$

Recalling our assumption of piston flow from the recharge to sample points, the residence time can be used to directly calculate groundwater velocities. If hydraulic gradients are known, then effective rock hydraulic properties can be evaluated using Darcy's Law.

The ^{36}Cl atomic mass balance approach can be readily extended to include an additional chloride source. Additional terms associated with the atomic mass of ^{36}Cl contributed by a chloride source can be added to equation 2. Terms to be added are similar to the terms describing the recharge ^{36}Cl input, and almost any well defined chloride input can be accommodated with a mixing equation; however, if some assumptions can be made about the chloride source the equations are greatly simplified. For example, if it can be assumed that decay or re-equilibration of the ^{36}Cl added by from the source is negligible, only one additional term is required, and no additional time variables are introduced. The general equation for this condition is:

$$(5) \quad R \cdot C \cdot V = V_o \cdot R_o \cdot C_o \cdot e^{-\lambda t} + V_o \cdot R_{se} \cdot C_o (1 - e^{-\lambda t}) + V_i \cdot R_i \cdot C_i$$

where R_i is the ^{36}Cl/Cl ratio of the source chloride, C_i is the total chloride concentration of the source input, V_i is the water volume of the source input, and all other variables are as previously defined. In this case, as Cl^- is being added from an additional source of different origin, the total chloride cannot be expected to remain constant and chloride concentrations must be explicitly considered. As there must also be a total chloride mass balance:

$$(6) \quad \text{Mass of } Cl^- = V \cdot C = V_o \cdot C_o + V_i \cdot C_i$$

Considering this total chloride mass balance, and further assuming that water volume is approximately constant ($V=V_o$), equation 5 can be rewritten to describe the general case where C_o is a known constant and the source chloride contribution is variable, as follows (Phillips 1986b):

$$(7) \quad R \cdot C = R_o \cdot C_o \cdot e^{-\lambda t} + R_{se} \cdot C_o (1 - e^{-\lambda t}) + R_i (C - C_o)$$

Equation 7 may solved for the residence or decay time associated with the recharge ^{36}Cl. Since the water volume was assumed to be approximately unaltered, the decay time should be equivalent to the water "age":

$$(8) \qquad t = \frac{-1}{\lambda} \ln \frac{(C/C_o)(R-R_i) + (R_i - R_{se})}{R_o - R_{se}}$$

Two special cases of equations 7 and 8 are particularly interesting; when the source $^{36}Cl/Cl$ ratio is approximately zero, and when the source $^{36}Cl/Cl$ ratio equals the secular equilibrium ratio. A $^{36}Cl/Cl$ ratio equal to the secular equilibrium ratio is applicable if the source of the water is from mineral dissolution or the flushing of high-concentration connate water. As can be seen from equations 7 and 8, the resulting residence time equation in this case is:

$$(9) \qquad t = \frac{-1}{\lambda} \ln \frac{C(R - R_{se})}{C_o(R_o - R_{se})} = \frac{-1}{\lambda} \ln \frac{(C/C_o)(R - R_{se})}{R_o - R_{se}}$$

Similarly, a chloride source with a $^{36}Cl/Cl$ ratio of zero is applicable if the source of the additional chloride is the dissolution of thick, bedded marine evaporites or the flushing of high chloride concentration "dead" connate waters. The chloride source does not change the concentration of ^{36}Cl, instead the $^{36}Cl/Cl$ ratio is diluted by the addition of only stable chloride.

Again as can be seen from equations 7 and 8, the resulting residence time equation is (Bentley et al., 1986b):

$$(10) \qquad t = \frac{-1}{\lambda} \ln \frac{C \cdot R - R_{se} \cdot C_o}{C_o(R_o - R_{se})} = \frac{-1}{\lambda} \ln \frac{(C/C_o)R - R_{se}}{R_o - R_{se}}$$

Note that the development of equation 8 to address cases with a chloride source, has included a few limiting assumptions beyond those applicable to equation 4, the no source case. First, the recharge chloride concentration is now assumed to be a known constant; second, the source or other processes are assumed to have negligibly changed the water volume; and third, the chloride that has been introduced by the additional source is assumed to maintain a constant $^{36}Cl/Cl$ ratio, even after its addition. These assumptions were made to simplify equation 8 and do not preclude the development of more complex equations where the assumptions are relaxed; however, the extreme, allowing decay or buildup of additional source ^{36}Cl, will introduce a second time variable that may in turn be complicated by the distributed nature of the source. If the source is not a point source and changes in the $^{36}Cl/Cl$ ratio of the source need to be included, a multiple-cell type model would be required.

As demonstrated, equation 8 is potentially applicable for a number of possible chloride sources. Because of the no-decay assumption, the equation requires that sources input distant from the sample point have $^{36}Cl/Cl$ ratios somewhat similar to the media secular equilibrium ratio. Small volume inter-aquifer leakage of chloride which has reached secular equilibrium under somewhat different geochemical conditions can be reasonably well described by equation 8, with R_i equal to the source R_{se}. Conceivably, small-volume chloride contamination at the sample point could also be accounted for by equation 8 if the associated $^{36}Cl/Cl$ ratio was known.

Relaxing the constant water volume assumption could be motivated by two possibilities, either the water volume changes because of some concentrating process as described for equation 4 and/or there is a significant water volume associated with the additional chloride source. In general equation 5 still applies, and the best applicable equation can be developed by simplifying equation 5 using the volumetric and total chloride mass balances to eliminate as many unknown variables as possible. For example, if a volume variable, f is introduced and defined as the proportion of the sample volume attributable to the recharge volume, the equivalent equation 8 becomes:

$$(11) \qquad t = \frac{-1}{\lambda} \ln \frac{(C/f \cdot C_o)(R - R_i) + (R_i - R_{se})}{R_o - R_{se}}$$

where, as stated, $f = V_o/V$. The proportion f may or may not be estimated otherwise, but if the chloride concentration of the additional source, C_i, is known, f can be calculated from the total chloride mass balance as:

$$(12) \quad f = \frac{C_i - C}{C_i - C_o}$$

An example of a hydrologic phenomenon that might warrant this extended application would be mixing in the borehole of water from two different aquifers, one of which is considered as an input of a known fixed ^{36}Cl/Cl ratio.

These mixing equations are the first introduced that include the mixing of different water volumes and so really describe mixing in the hydrologic sense. The original assumption of piston flow could be in question. The concept of the residence time or water "age" must be limited to that fraction of the sample associated with the recharge water. Proportions would have to be constant. Note that in the inverse case, when the recharge residence time can be otherwise approximated, the volumetric proportion variable, f, can be independently solved for using either the ^{36}Cl or Cl mass balances.

Equation 5 can be easily further extended to include any number of non-decay/buildup sources. All sources are assumed to be of a constant ^{36}Cl/Cl ratio. Each new source will require that an additional concentration variable be known, and each new source that is of significant water volume will require that an additional volume variable be known. Again, the application of any mixing equation requires consideration of the reality of the calculated recharge water residence time.

Given the possibility of applying any one of the number of different mixing equations that can be developed, the available data needs to be examined to determine what type of down-gradient chloride history is relevant. Comparative study of measured $^{36}Cl/Cl$ ratios, total Cl concentrations, and ^{36}Cl concentrations serves to reveal important hydrologic processes. Such processes will tend to alter the relation of these parameters in different ways. The nature of the superposition of other processes on the effects of decay determines the applicable mixing equation. In addition, parameters need to be examined spatially. Significant spatial trends must be considered in light of other hydrologic understandings, particularly, whether that part of the system containing the trend is well represented by piston flow along a down gradient flow path. Even if the concept of a residence time appears invalid, the ^{36}Cl data can still provide insight as to the origin and history of different waters in the system. Such insight can help guide the interpretations of other, associated system data.

METHODS AND RESULTS

Field collection and lab preparation of samples

Samples for ^{36}Cl analysis were collected as part of the USGS San Juan Basin RASA study. Samples were taken from wells completed in the Morrison, Dakota, Gallup, and Point Lookout aquifer units, with emphasis on the older units of the Morrison Formation. The locations of the wells sampled are shown on Figure 5, where Table 1 relates the numbers used on the figure (and elsewhere in the remainder of this report) to the well names. Samples were collected during the period 4/86-1/89, and for a few wells (28, 29, 33, 34, and 36) samples were collected on more than one occasion. Care was taken to select wells with a reasonably well known completion history. Sampling efforts were concentrated in the northwest part of the basin, away from most large withdrawals and uranium deposits, and in an area where the hydrogeology was seemingly less complicated. Based on the flow directions indicated for the Morrison in Figure 4, it was hoped that the sample distribution would include some part of a flow path associated with water travel from the recharging flanks of the Chuskas to discharges along the San Juan River.

The known completion history of Morrison wells for the units of study is shown on Table 2. As indicated on the table, most of these wells are flowing wells, with four near-outcrop wells pumped with windmills, and two other wells pumped with down-hole

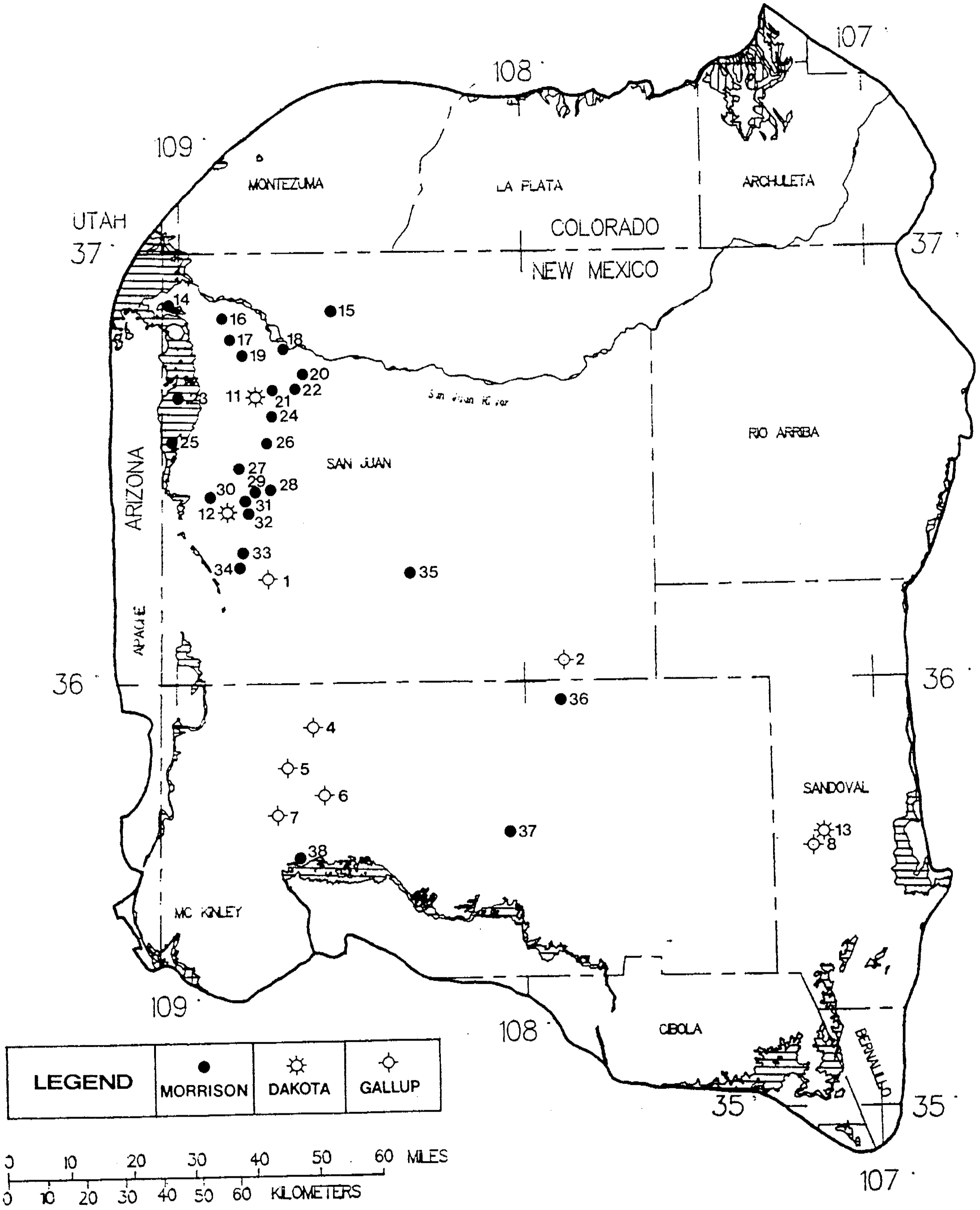

Figure 5. Locations of wells sampled for ^{36}Cl analysis (Morrison outcrop shaded).

Table 1. Well numbering used for this study

Well #	Well name
Morrison Wells:	
14	12K-335
15	Arco-WS2
16	12T-637
17	12T-636
18	12T-520
19	12T-548
20	12T-630
21	12T-628
22	12T-629
23	12T-329
24	12T-638
25	12T-501
26	12K-320
27	12T-644
28	12T-620
29	12T-647
30	12T-662
31	12T-633
32	12T-651
33	12T-640
34	12T-632
35	Burnham Well
36	Chaco-Jm

Well #	Well name
Dakota Wells:	
11	12K-357
12	12T-543A
13	Cerro Well
Gallup Wells:	
1	12T-576
2	Chaco-Kg
4	EPNG#5
5	14T-583
6	14T-321
7	14T-501
8	Ojo Well
Point Lookout Well:	
39	Chaco-Kp

Table 2. Morrison well completions

Name	#	Type	Depth	Cased To	Opens To	Kg Depth	Km Depth	Kd Depth	Jmb Depth	Jmw Depth	Jmr Depth	Jmsw Depth	Jwa Depth	Depth past Jm
12K-320	26	F	1992	1745	1992	460	540	1300	1470	1530	1660	1800	1980	12
12K-335	14	W	604	160	604			48	160	260	420			
12T-329	23	W	555	340	535									
12T-510	25	W	702	553	702			0	400?					
12T-520	18	F	1777	1482	1777	248	335	1015	1185	1342	1485	1610	1760	
12T-548	11	F	2013	1733	2013	0		821	1024					
12T-620	28	F	2034	1200	2034	0	105	789	1008	1178	1640		1879	
12T-628	21	F	2597	1827	2597	655	880	1460	1610	2220	2480	2540		
12T-629	22	F	2520	1764	2511	460	720	1290	1508	1928		2290?	2423?	97
12T-630	20	F	2300	1512	2300	250	470	1080	1300	1478	1745	2124	2222	78
12T-632	34	F	2518	1750	2518	418	878	1363	1546	1750?	2000		2443	75
12T-633	31	F	2125	1512	2125	0	59	874	1080	1270	--	--	1960	
12T-636	17	F	2035	1344	2035	0	280	880	1070	1330	1594	1800	1929	
12T-637	16	F	2000	1218	2000	0	190	804	983	1174	1495	1792	1864	
12T-638	24	F	2682	1806	2682	602	1030	1490	1643	2010		2556	2615	67
12T-640	33	F	2349	1491	2349	195	400	1170	1342	1569	1821	2215	2235	114
12T-644	27	F	1912	1386	1912	0	240	1009	1215	1344	1618	--		
12T-647	29	F	1912	1407	1912		0	1118	1323					
12T-651	32	F	1691	1281	1691	0	90	800	1060					
12T-662	30	F	1751	950	1730		0	600	750				1750	1
15T-567	37	P	2605	1520	2140				1450?					
16T-534	38	W	410	300	410			10	235?					
ARCO-WS2	15	P	2736	2238	2668	--	1315	1804	2050					
BURNHAM	35	F	5250	5000	5250	3565	3747	4427	4750	4980				
CHACO-JM	36	F	3988	3957	3988					3960?	4360			

Kg - Cretaceous Undiferentiated Mancos Shale
Kml - Cretaceous Gallup Sandstone
Kd - Cretaceous Dakota Sandstone
Jmb - Jurassic Morrison Formation: Brushy Basin Member
Jmw - Jurassic Morrison Formation: Westwater Canyon Member
Jmr - Jurassic Morrison Formation: Recapture Member
Jmsw - Jurassic Morrison Formation: Salt Wash Member
Jwm - Jurassic Wanahka Group (inc. Bluff Sandstone, Todilto Limestone, Entrada Sandstone)

F - Flowing Well
W - Windmill
P - Downhole Pump

Notes:

Data from William Dam USGS RASA

Measurements are in feet

pumps. The majority of the non-outcrop wells sampled on the Navajo Indian Reservation (see Figure 5) were drilled for uranium exploration purposes by Exxon and were left as groundwater supply wells. The open intervals in most of the Morrison wells exceed 200 feet, and in many wells exceed 700 feet. All wells are recorded as cased down to the top of the Morrison Formation, and most are cased through the Brushy Basin Shale, a confining layer. The Westwater Canyon Sandstone Member, although it does contain some mudstone layers, is the major water bearing member of the Morrison Formation. This member is below the Brushy Basin Member, and is turn underlain by the Recapture Shale Member. The Recapture Member mainly consists of interbedded, red shale and white sandstone (Stone et al., 1983), and is underlain by the Salt Wash Sandstone Member. The Salt Wash Member intertounges with the underlying Bluff Sandstone, a part of the Jurassic Wanahka Group. As can be noted on Table 2, the deeper wells are all open for depths beyond the Morrison proper and into the Jurassic formations below. The well openings tend to span at least three members of the stratified Morrison Formation.

Samples were collected by either precipitating chloride directly out of water samples as silver chloride or by running well water through an anion exchange column. Exchange columns were used to facilitate easy collection of chloride when the chloride concentrations were expected to be low. Samples containing less than about 10 mg chloride were found difficult to further process in the laboratory without going to the extent of adding carrier, so exchange resins were used when the chloride concentration at the well was suspected to be low enough to require more than 2 liters of water sample. A more specific procedure for sampling by anion exchange columns is provided in some detail as Appendix A.1. After collection, columns were chemically flushed of chloride, and the chloride was precipitated as silver chloride. In addition, a sample was taken at the time of the ^{36}Cl sample collection for separate chloride analysis by other investigators of the USGS San Juan Basin RASA project.

Silver chloride is the target material used in the AMS measurements and is readily precipitated out of a slightly acid chloride solution by addition of silver nitrate. In a few cases, samples for this study were precipitated in the field, the only additional consideration being that AgCl is photo-reactive and should be kept covered or at least in dark bottles. Following collection and precipitation, the silver chloride recovered from the samples was then further treated in the laboratory. The process was designed to remove any other material mixed in with the precipitate that might interfere with the analysis. In particular, the measurement of ^{36}Cl by AMS is made less precise by even small amounts of ^{36}S, an interfering atomic isobar in the measurement. Sulfur is a ubiquitous element, so precautions must be taken to limit the quantities present. Appendix A.2. outlines the development and presents the details of the laboratory procedure utilized for the samples in this study. In summary, the silver chloride was dissolved and re-precipitated several times and, at a time when the silver chloride was in solution, barium carbonate was added to precipitate any remaining sulfate present as barite.

Once treated, samples are loaded into 1-2mm (diameter and depth) cavities drilled in small tantalum steel cylinders. These sample holders were chosen by the TAMS analysis group at the University of Rochester for their low sulfur content and machining characteristics (Conard et al., 1986). Sample weights in the holders usually ranged from 5-10 mg, although smaller sample quantities can be accommodated by smaller cavities and, if necessary, the addition of clean gold powder to increase sample volume. The minimum possible sample size is dictated by the AMS analysis requirements and the quantity of ^{36}Cl in the sample.

AMS analysis

The samples for this study were analyzed for the ratio of ^{36}Cl to stable Cl on the tandem accelerator mass spectrometer (AMS) at the Nuclear Structure Research Lab, University of Rochester, New York. Measurements were made during four different ^{36}Cl runs in the period 12/87-1/89 by David Elmore, Peter Kubik, Ray Tang, and colleagues, using techniques described in Elmore et al., 1979; Elmore et al., 1982; and Elmore and Phillips, 1987. At the time of these measurements, the analysis was instrumented such that as many as seven sample holders could be secured in a shuttle wheel for subsequent targeting by the ion beam source. On any given wheel, two standards (samples with a known isotope ratio) and a blank (sample containing no detectable radioisotope) were emplaced together with four unknown samples. Standards were run periodically between runs on the unknowns to allow for normalization, and blanks were used to measure background.

The raw data from the accelerator measurements include beam, interference, and fractionation parameters needed to correct the isotopic ratio measured and determine the absolute error. The University of Rochester performed these calculations as part of their computer controlled analysis, as described in Elmore et al., 1984. The final results of these calculations, grouped by the wheel on which the sample holder was placed, are presented in Table 3 for the samples processed for this study. The ratios determined for the blanks (designated as BLK) on each wheel are also given to describe background levels. To check for lab contamination, a number of blanks were also measured that had undergone the chemical purification process as part of this study (designation, CHEM BLK). Table 3 also indicates what aquifer was sampled and whether the sample was precipitated from a water sample in the field, in the lab, or in the lab after collection with an exchange column. The number etched on the side of the sample holder used for a particular sample is given for purposes of sample tracking.

The results presented in Table 3 indicate that the Rochester

Table 3. Measured Cl-36/Cl[*] ratios.

WELL NAME	UNIT	DATE COLLECTED	METHOD USED	HOLDER NUMBER	CL-36 ANALYZED	CL-36 RATIO	DEV. +/-	ERROR %
SAMPLE WHEEL: GH09								
CHACO-KP	KP	11/10/87	LAB	1270	12/19/87	18.60	3.2	17.2
NSRL BLK	N/A			556	12/19/87	3.28	2.1	64.0
SAMPLE WHEEL: GH11								
12T-629	JM	06/10/87	FIELD	1253	12/20/87	39.10	5.9	15.1
12T-630	JM	06/10/87	FIELD	1252	12/20/87	37.90	4.0	10.6
ARCO-WS2	JM	07/21/87	LAB	1259	12/20/87	15.40	2.9	18.8
CONARD BLK	N/A			1138	12/20/87	0.03	3.3	***.*
SAMPLE WHEEL: GH12								
12T-520	JM	06/10/87	FIELD	1255	12/20/87	58.90	3.4	5.8
BURNHAM	JM	06/11/87	FIELD	1219	12/20/87	290.00	16.0	5.5
CHACO-JM	JM	10/22/87	LAB	1260	12/20/87	537.00	21.0	3.9
OJO-KG	KG	12/03/87	LAB	1267	12/20/87	512.00	27.0	5.3
NSRL BLK	N/A			1556	12/20/87	4.22	1.3	30.8
SAMPLE WHEEL: GH14								
12T-548	JM	07/14/87	LAB	965	12/20/87	3.98	1.8	45.2
12T-620	JM	07/17/87	LAB	1258	12/20/87	14.90	2.7	18.1
CHACO-KG	KG	10/21/87	LAB	1256	12/19/87	198.00	12.0	6.1
CHACO-KP	KP	11/10/87	LAB	1270	12/20/87	18.20	3.7	20.3
NSRL BLK	N/A			558	12/20/87	0.53	1.0	188.7
SAMPLE WHEEL: GH22								
12T-629	JM	06/10/87	FIELD	1253	12/23/87	44.00	2.9	6.6
OJO-KG	KG	12/03/87	LAB	1267	12/23/87	562.00	30.0	5.3
NSRL BLK	N/A			1558	12/23/87	0.78	1.0	128.2
SAMPLE WHEEL: HB09								
12T-632	JM	07/16/87	LAB	1246	02/28/88	481.00	21.0	4.4
12T-636	JM	07/02/86	LAB	1261	02/28/88	42.00	5.0	11.9
12T-640	JM	07/15/87	LAB	1244	02/28/88	400.00	23.0	5.8
15T-567	JM	10/02/87	LAB	1245	02/28/88	894.00	34.0	3.8
CHEM BLK	N/A	02/15/88	LAB	1247	02/28/88	6.85	4.1	59.9
SAMPLE WHEEL: HB12								
12T-638	JM	07/01/86	LAB	1266	02/29/88	213.00	11.0	5.2
12K-357	KD	07/23/87	LAB	1268	02/29/88	169.00	13.0	7.7
CERRO-KD	KD	12/03/87	LAB	1262	02/29/88	149.00	13.0	8.7
CHEM BLK	N/A	02/15/88	LAB	1187	02/29/88	0.76	1.9	250.0

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[*] isotopic ratio is measured as radioactive/stable

WELL NAME	UNIT	DATE COLLECTED	METHOD USED	HOLDER NUMBER	CL-36 ANALYZED	CL-36 RATIO	DEV. +/-	ERROR %
SAMPLE WHEEL: HF13								
12K-335	JM	06/06/88	RESIN	973	07/27/88	244.00	35.0	14.3
12T-637	JM	06/06/88	RESIN	967	07/27/88	44.50	6.1	13.7
12T-651	JM	06/11/88	RESIN	969	07/27/88	41.90	5.5	13.1
CHEM BLK	N/A	07/20/88	LAB	1263	07/27/88	0.00	2.4	***.*
NSRL BLK	N/A			1783	07/27/88	0.00	2.3	***.*
SAMPLE WHEEL: HF15								
12T-329	JM	06/06/88	RESIN	1271	07/28/88	333.00	23.0	6.9
12T-644	JM	06/06/88	RESIN	966	07/28/88	36.00	3.0	8.3
12T-576	KG	06/28/88	LAB	1257	07/28/88	676.00	31.0	4.6
14T-583	KG	06/28/88	LAB	1265	07/28/88	153.00	16.0	10.5
NSRL BLK	N/A			1776	07/28/88	0.00	1.0	***.*
SAMPLE WHEEL: HF22								
12K-320	JM	07/01/88	RESIN	1905	07/30/88	251.00	25.0	10.0
12T-501	JM	06/06/88	RESIN	1907	07/30/88	295.00	15.0	5.1
12T-640	JM	06/29/88	RESIN	1904	07/30/88	113.00	7.4	6.5
12T-647	JM	06/29/88	RESIN	1906	07/30/88	407.00	19.0	4.7
NSRL BLK	N/A			1784	07/30/88	0.74	1.7	229.7
SAMPLE WHEEL: HF28								
16T-534	JM	07/14/88	RESIN	971	07/28/88	630.00	39.0	6.2
NSRL BLK	N/A			1782	07/31/88	9.14	3.8	41.6
SAMPLE WHEEL: HF29								
12T-543A	KD	06/29/88	RESIN	964	07/28/88	730.00	45.0	6.2
14T-321	KG	06/20/88	RESIN	1264	07/28/88	22.00	5.2	23.6
14T-501	KG	06/28/88	RESIN	975	07/28/88	569.00	39.0	6.9
EPNG#5	KG	07/15/88	RESIN	1903	07/28/88	122.00	10.0	8.2
NSRL BLK	N/A			1793	08/01/88	0.39	1.4	359.0
SAMPLE WHEEL: HF33								
12T-640	JM	06/29/88	RESIN	1904	07/28/88	129.00	16.0	12.4
NSRL BLK	N/A			1798	08/01/88	0.00	2.2	***.*
SAMPLE WHEEL: IA34								
12T-620	JM	06/30/86	LAB	2153	01/12/89	163.00	23.0	14.1
12T-640	JM	11/23/88	RESIN	2155	01/12/89	274.00	16.0	5.8
12T-662	JM	01/05/89	RESIN	2154	01/12/89	573.00	27.0	4.7
CHEM BLK	N/A	01/07/89	RESIN	2152	01/12/89	25.60	5.3	20.7
NSRL BLK	N/A			1780	01/12/89	7.65	3.0	39.2
SAMPLE WHEEL: IA35								
12T-632	JM	11/22/88	LAB	2157	01/13/89	1857.00	62.0	3.3
12T-647	JM	11/22/88	RESIN	2156	01/13/89	417.00	18.0	4.3

measurements can be viewed with confidence. The Rochester blanks show a consistently low or extremely low background level of ^{36}Cl compared to the levels observed in the samples, and the absolute error in the measurements is typically under 10 percent except for the very low level samples. The four times that samples in one holder were run on more than one wheel (samples 8, 22, 33B, and 39) the results are within the calculated deviations of each other.

The chemistry process seems to have been somewhat less consistent. Blanks for the lab at New Mexico Tech are made from a sample of Weeks Island Halite preserved for this purpose. Blank material processed at Tech has been used at Rochester in the recent past as the NSRL blank; however, since that time there has been a change of lab facilities and an increase in other lab use. The samples for this study were not the first samples processed in the new facility and the chemistry of the samples from this study associated with the first three sample runs at Rochester are not in question. It is in the last run, in January of 1989, that the chemistry blanks (measured ratios: 33.5 and 25.6 x 10^{-15}) showed the possibility of laboratory contamination.

A review of laboratory conditions indicated that a malfunctioning water deionizer was probably responsible for the elevated blank values in the last run. Water being used to make solutions for the laboratory process were not as purified as they were supposed to be. The ratio that might be associated with the distilled water added to the deionizer is unknown, but nothing extraordinary would be expected. As long as the contamination can be assumed near constant, the chemistry blank measurements provide an estimate of the ratio of the contamination. Any interpretation of the measurements in the last run must be made with the possibility of minor lab contamination in mind. Samples measured in the last run include two samples with extremely high ratios (sample 31: 2,201; sample 34B: 1,857 x 10^{-15}) which could be explained as contamination by bomb-pulse type waters either in the lab or in the field; however, there is no other reason to suspect such contamination. Other, potentially valid geochemical explanations exist. It is fortunate, most of the ratios measured in the last run were high, so as to be less effected by low level contamination.

Of the two blanks measured in the last run, one was the typical lab precipitate from a solution of the halite, and the other was obtained by pouring 16 liters of a dilute halite solution through an anion exchange column. The similarity in their resulting values indicates that the chloride contamination for the 1989 run may be a fairly constant isotopic ratio of 25-30 x 10^{-15}, and that contamination by the resin does not appear likely. Indeed, there appears to be no correlation between the use of the resin and consistently low or consistently high measured ratios.

Determination of secular equilibrium isotopic ratio

As described earlier, the secular equilibrium ratio is the ^{36}Cl/stable chloride ratio at which production of ^{36}Cl equals the decay of ^{36}Cl. In the case of groundwater, we are interested in the secular equilibrium ratio associated with hypogene production of ^{36}Cl in the porous media. This ratio is a function of the rock composition, specific gravity of the rock, and the effective porosity of the media. The rock composition determines both the neutron flux active in the media and the elemental partitioning of the neutron absorption.

Bentley et al. (1986) present an equation (their equation 6) for calculating the secular equilibrium ratio assuming that the dominant production mechanism for ^{36}Cl is neutron activation of groundwater ^{35}Cl:

$$(13) \qquad R_{se} = \frac{0.7577\Phi_n\sigma_{35}}{\lambda(\sum N_j\sigma_j + N_{35}\sigma_{35} + N_{H_2O}\sigma_{H_2O})}$$

where the bracketed term reflects the neutron absorption by elements in the rock as well as by the chloride and water molecules in the pore water; j refers to those elements in the rock; σ_i is neutron absorption cross section of the isotope, element, or molecule; 0.7577 is the isotopic abundance of ^{35}Cl; Φ_n is the subsurface neutron flux; and λ is the decay constant of ^{36}Cl.

The number of ^{35}Cl atoms in the pore water per kilogram of saturated porous media, N_{35}, and the number of water molecules in the pore water per kilogram porous media, N_{H_2O}, can be calculated, respectively:

(14a) $$N_{35} = 0.7577 V_w C_{Cl} A$$ (14b) $$N_{H_2O} = V_w C_{H_2O} A$$

where C_{Cl} is the chloride concentration in the pore water in moles per liter; A is Avagadro's number; C_{H_2O} is the concentration of water molecules in the pore water in moles per liter (approx. 1000/18); and V_w is the fractional water volume. Note that in Bentley et al. (1986) V_w is calculated as the porosity divided by the dry bulk density (in their equation 3); however, this approximation does not include consideration of the fluid density fraction of the total density. As the values being calculated are clearly relative to the saturated mass of the media, inclusion of the fluid density, which could be 20 percent or more of the mass, is more exact. In addition, the calculation can easily be extended to include water saturation for epigene application:

(15) $$V_w = \frac{1}{\frac{(1 - s)}{s} \rho_a + \rho_w + \frac{\rho_B}{ns}}$$

where s is degree of saturation; n is porosity; and ρ_w, ρ_B, and ρ_a are the specific weights of the water, rock, and air of the media. The density of air, potentially applicable in the partially saturated case, is ignored as insignificant for calculations for this study.

The assumption that neutron activation of groundwater ^{35}Cl is the only significant production mechanism for groundwater ^{36}Cl is an extremely convenient one. The subsurface neutron flux can then be simply partitioned among the rock and water isotopes based on reaction cross-section. The amount of 36Cl buildup in the pore water would depend only the concentration of 35Cl in the water relative to the total rock thermal neutron cross section. Mass transfer of ^{36}Cl between the rock and the pore water would presumably be non-existent or associated with Cl in the rock of a constant ^{36}Cl/Cl ratio.

As indicated by Fabryka-Martin (1988), production of ^{36}Cl by neutron activation of ^{39}K is another mechanism important in some rock types. The reaction cross-section for ^{35}Cl is more than an order of magnitude greater than that of ^{39}K, and the concentrations in water of Cl are frequently greater as well. Nonetheless, in a geochemical system where K-bearing minerals are being aggressively leached, significant quantities of high ratio chloride could be released. Depending on particle energies, neutron activation of ^{39}K can also competitively produce ^{39}Ar. The success of the ^{40}K-^{40}Ar method of dating common rock minerals is evidence against the release of such potassium reaction products except in unusual geochemical conditions, and the significant presence in groundwater of ^{39}Ar from neutron activation of ^{39}K may indicate some degree of simultaneous release of ^{36}Cl (Fabryka-Martin, 1988). For completeness, it should be noted that in some geological environments, minor ^{36}Cl production can also occur from an α-particle reaction with ^{33}S, but this is rarely significant. In any case, hypogene ^{36}Cl production other than neutron activation of groundwater ^{35}Cl is only likely to be significant in low chloride groundwaters in geochemically active conditions.

The subsurface neutron flux term, Φ_n, in equation 13 is the major difficulty in the secular equilibrium determination (Bentley et al., 1986). At depths below the influence of cosmogenic particles, about 30 meters, the neutron flux is reduced to a summation of the neutrons produced by spontaneous fission of ^{238}U and by the U and Th α-decay series α-particle bombardment of light nuclei in the media (Fabryka-Martin, 1988; Andrews, et al., 1986). Based on experiments consisting of hitting light element targets with ionized helium beams from a tandem accelerator, Feige et al. (1968) have calculated neutron production rates associated with U and Th concentrations typical of soils. Fabryka-Martin (1988) considered further data available and included a number of additional extrapolations for additional elements to further develop the calculations.

As compiled by Fabryka-Martin (1988), the contribution of spontaneous fission of ^{238}U to production of neutrons occurs at a rate described by:

$$(16)\quad (P_n)_{sf} = N_{238}\lambda_{sf}\nu = 0.429\ [U]\ ng^{-1}yr^{-1}$$

where N_{238} is the atomic concentration of ^{238}U in the rock; λ_{sf} is the decay constant for spontaneous fission of ^{238}U (8.49 + 0.14 x 10-17 yr-1); ν is the average number of neutrons emitted per spontaneous fission of ^{238}U (2.00 ± 0.03); and [U] is the fractional concentration of U (ppm). The fractional concentration is generally regarded as the product of the total U concentration and the isotopic abundance of ^{238}U, 99.27 percent. This would modify the coefficient in equation 16 to 0.426 when the total U concentration (ppm) is used for [U].

The neutron production associated with the U and Th α-decay series depends on the target elements that compose the media as well as the concentration of the parent materials. Feige el al. (1968) present equations of use for estimating the yield of neutrons from light elements targeted by α-emitters. They indicate neutron yields of mixtures or compounds consisting of a number of elements can be calculated from:

$$(17)\quad (P_n)_{\alpha m} = \sum_i S_iW_iY_i \Big/ \sum_i S_iW_i$$

where S_i is the mass stopping power of element i for α-particles of a given energy (MeV/g·cm^2), W_i is the fractional abundance of element i, and Y_i is the neutron yield of element i per ppm parent material, m. Table 4, from Fabryka-Martin (1988), provides the mass stopping powers and neutron yields for a number of important light elements. Although the mass stopping power of an element decreases with increasing energy of the α-particle, the mass stopping power ratios for any pair of elements does not change more than 4% for α-particle energies between 5.3 and 8.8 MeV (Feige et al., 1968). Calculating yields for both uranium and thorium concentrations using equation 17 and Table 4 and summing together with the spontaneous fission production establishes the total neutron production rate in the media.

The success of estimating the neutron flux using the presented equation has been suggested in a comparison to measured values by Andrews et al. (1986). There are, however, some difficulties in applying the given equations to the determination of the subsurface neutron flux. The development of equation 13 as described in Bentley et al. (1986, a and b) and the inclusion of the media water and its contents from the neutron absorption and neutron activation standpoints imply that the neutron flux needed to calculate the secular equilibrium ratio is that relative to a kilogram of saturated porous media. Therefore, the method chosen to calculate the neutron flux should also consider how to incorporate the presence of the water. Andrews et al. (1986) simply present the constants used in equations similar to 16 and 17 above, for data from the Stripa granite.

Table 4. Neutron yields per ppm U and ppm Th, due to (α,n) reactions (modified from Fabryka-Martin, 1988)

Target element	Mass SP	n produced per ppm U			n produced per ppm Th
		238 U	235 U	Total U	
Li	548	19.8	1.3	21.1	9.6
Be	529	251	14	265	91.2
B	527	59.3	3.1	62.3	19.2
C	561	0.43	0.025	0.45	0.18
O	527	0.22	0.012	0.23	0.079
F	472	29.0	1.8	30.8	11.8
Na	456	13.7	0.78	14.5	6.8
Mg	461	5.5	0.35	5.8	2.6
Al	444	4.7	0.31	5.1	2.6
Si	454	0.65	0.042	0.69	0.335
P	433	0	0	0	0
S	439	0	0	0	0
K	414	0.42	0.032	0.045	0.305
Ca	428	0	0	0	0
Ti	375	0	0	0	0
Fe	351	0.18	0.006	0.19	0.205
UO_2	74.7	0.073	0.004	0.077	0.027
H	1563	0	0	0	0

Neutron yields reported in units of n/gram i/yr per ppm U or per ppm Th.

Mass SP = Mass stopping power of element for α-particle of energy 8.0 MeV, in units of MeV/(g/cm). Values from Ziegler (1977, pp.45-49), converted from eV/(10^{15} atoms/cm^2) to MeV/(g/cm^2) by applying a factor of 10^{-6} N_A/A_i where N_A = Avogadro's number (6.02 X 10^{23} atoms/mol) and A_i = atomic weight (g/mol).

Mass stopping power for UO_2 from Perry and Wilson, 1981(p. 12)

Mass stopping power for H calculated from Feige et. al, 1968

Neutron yields derived from sum totals for each α-decay series in Appendix Table C-2 times rate of α-production per ppm U or Th per yr (Appendix Table C-1)-- where the appendices listed, are in Frabryka-Martin, 1988.

In the neutron flux calculations of equations 16 and 17, the water mass can be disregarded, regarded simply for its mass contribution, or regarded as if in a mixture with the rock. If the water is to be disregarded, the neutron flux calculated would be the same as that of the rock alone. If the mass of the water is incorporated in the calculations, the concentrations in the media of both the parent α-decay materials and the light target elements will be diluted, reducing the calculated flux. If the water is regarded as in a mixture with the rock, concentrations of rock elements will be diluted by the mass contribution, and the water can elementally contribute to α-particle induced neutron production. The water contains oxygen which has a small (α,n) neutron yield, but the dominant contribution in most cases is the significant mass stopping power of both oxygen and hydrogen. The travel distance for α-particles is extremely short, and depending on the mineralogic location of the parent and target materials, the assumption the water exists as a mixture with the rock may or may not reflect the geochemical conditions. However, in general the incorporation of the water mass in the calculations has a much greater effect on the calculated flux than including the water in α-particle neutron production.

The method of calculating the secular equilibrium ratio based on the equations given in this section has been written up in a computer routine which is attached as Appendix B.1., and is referred to as CLSEC. The FORTRAN listing is heavily commented and includes a beginning portion describing the expected contents and structure of an input file. CLSEC will calculate the secular equilibrium ratio for a number of input sets, each set consisting of porosity, saturation, and rock density and composition data.

Rock composition data required for CLSEC consist of major and minor element chemistries, where the major elements can be provided as either oxide or elemental weight percentages. The relative importance of the many chemistry constituents depends on their abundance and reaction cross-section, and the rock quantities of Cl, F, Be, N, and S would only be significant in uncommon conditions. The input data can also include the chloride content of the pore water, which is used simply to correct further the neutron absorption of the water in high total dissolved solids situations. The output given by CLSEC includes calculations of neutron absorption, neutron flux and secular equilibrium ratios. Flux calculation and associated secular equilibrium ratio results are given for the three different approaches to considering the water content of the media as previously described.

In order to verify the program calculations, a test input file was generated using data for three different general rock types as available in Fabryka-Martin (1988). Data available for comparison includes various neutron flux calculations as well as secular equilibrium determinations. CLSEC calculated values are compared in Table 5 with those of Fabryka-Martin (1988). The total neutron flux and secular equilibrium values shown for CLSEC are those calculated assuming the water was in a mixture with the rock, but ratios calculated simply accounting for the mass of the

Table 5. Comparison of results from Fabryka-Martin (1988) with the CLSEC program.

	High Ca-Granite		Clay & Shales		Sandstone	
	Fabryka-Martin	This Report	Fabryka-Martin	This Report	Fabryka-Martin	This Report
Water volume (L/Kg)	4.783E5	4.815E5	5.357E5	4.719E5	5.241E5	5.479E5
Oxygen (ppm) in media from rock	3.700E3	3.310E3	4.17E4	3.64E4	8.700E4	7.790E4
Oxygen (ppm) in media from water	3.700E-3	3.727E-3	4.170E-2	4.098E-2	4.371E5	8.772E-2
Total oxygen (ppm) in media	4.746E5	4.782E5	4.940E5	4.355E5		4.700E5
Rock flux for U (a,n) per Kg media	3.55E3	3.52E3		3.00E3		0.25E3
Rock flux for Th (a,n)	4.76E3	4.75E3		5.05E3		0.43E3
Total rock flux (a,n)	8.3E3	8.27E3	7.6E3	8.05E3	0.72E3	0.68E3
Fisson rock flux	1.3E3	1.27E3	1.4E3	1.35E3	0.19E3	0.19E3
Total neutron flux	9.6E3	9.54E3	9.0E3	9.33E3	0.92E3	0.87E3
Secular equilibrium ratio	21.E-15	21.47E-15	12.E-15	12.82E-15	1.9E-15	1.60E-15

water and not water (α,n) contributions differ by less than 1 percent. The test input also included internal consistency check cases, where the porosity and saturation were set to zero, and is reproduced in detail together with the complete output file as Appendix B.2.a.

In general, the comparison of values in Table 5 is very favorable. The Fabryka-Martin results are calculated using a different method to determine the oxygen content of the rock and pore water, and her neutron flux calculations include (α,n) reactions associated with potassium and sulfur. Despite these differences, the values calculated for the total neutron flux and the secular equilibrium ratios are very close. The slight differences in the methods are more exaggerated in the case where the neutron flux is small (general sandstone case), but this case is associated with a uranium concentration of just 0.45 ppm and the calculated ratio is approaching the limits of analytical detection.

As an additional case of comparative interest, the data of the Great Artesian Basin in Australia as presented by Bentley et al. (1986) has also been incorporated into an input file and analyzed using CLSEC (input/output in Appendix B.2.b.). In this case the major element data was read as oxide weight percents (instead of elemental percents), the same units expected for San Juan Basin data. The secular equilibrium ratio calculated by the CLSEC program for the given input is 7.7×10^{-15}, whereas the ratio calculated in Bentley et al. (1986b) is 9×10^{-15}. Although these calculated ratios are not significantly different, it is interesting to note that if the presence of the water is ignored in the neutron flux used, CLSEC also calculates a ratio of 9×10^{-15} (see output). Also, the ratio actually used in the Bentley et al. (1986b) paper for dating equations was not the ratio they calculated, but a ratio of 5.7 x 10-15 that was actually observed in very old waters in the basin, suggesting the lower calculated value may be more truly representative.

The CLSEC program has been used to examine possible secular equilibrium conditions in the Morrison Formation of the San Juan Basin. Aquifer geochemistry, porosity, and rock specific weight are the inputs required for the program and these were gathered from available literature or estimated as described following. A number of data sets were developed to determine the sensitivity of the results to specific estimated parameter values. Due to the sensitivity of the results to the uranium and thorium content of the rock, this relationship was studied with some detail. The input and output files for the CLSEC program associated with the different data sets are given as appendix B.2.c., and the results will be summarized following a more detailed description of input parameter data available.

The general geochemistry of the Morrison rocks was obtained from two sources. The first, a paper by Spirakis and Pierson (1986), contains data for an average chemical background of over a hundred samples from the southern San Juan Basin. The data was also presented in Spirakis et al. (1981) and is referred to in these reports and in the appendix input files as the SSJB data set. They obtained this background data by computer searches of the USGS Rock Analysis Storage System and used it for comparison with geochemistries associated with uranium mineralization. One of these mineralized geochemistries, the Mariano Lake Deposit in the Smith Lake Uranium District of the Grants Uranium Belt, was also used as a data set to provide insight regarding the effects of mineralization on calculated secular equilibrium. The other source of general geochemical data used for this study consists of a computer search of the USGS Branch of Geochemistry Archive database otherwise known as PLUTO. The search was restricted to an area within the state of New Mexico north of 36 degrees latitude and west of 108 degrees longitude. The idea was to avoid well-known uranium mineralization areas and to concentrate on the region where the majority of the groundwater ^{36}Cl samples for this study were collected (see Figure 5).

The SSJB and Smith Lake data sets were lacking values for silica, lithium, chloride, boron, phosphorus, and a few of the more exotic trace elements. The data sets were filled out using data from the PLUTO search where possible, and rubidium, samarium, and gadolinium were estimated for all sets from the trace elements studies listed under the uranium and thorium data search mentioned following. Values for chloride and fluoride were not found, and fluoride was ignored, whereas the more important chloride was estimated from general estimates of chloride in sedimentary rocks.

Within the PLUTO search were seven samples along the southern edge of the defined area with uranium values over 250 ppm that were thrown out as uranium mineralized. In addition, out of the 23 remaining Morrison samples in the PLUTO set, those 8 that were sampled from drill core rather than mines or outcrop were considered less biased, and these were the only samples considered in determination of an average value unless data availability necessitated otherwise. The SSJB and Smith Lake data sets differ from the PLUTO data set in that the PLUTO average data values were determined as simple arithmetic averages, whereas the SSJB and Smith Lake average values were determined as geometric averages.

Information on uranium and thorium concentrations in the Morrison Formation was obtained in both of the sources used for the general geochemistry, as well as in numerous reports from the Department of Energy and by Brookins and others at the University of New Mexico (Brookins, 1975; Brookins, 1979a; Brookins, 1979b; Brookins and Della Valle, 1977; Haddad et al., 1981). Data are highly variable, and even samples adjacent to each other within an individual member unit can vary by orders of magnitude. The SSJB data set (Spirakas et al., 1981; Spirakas and Pierson, 1986) indicates an average uranium value of 18 ppm and does not provide any thorium data. The PLUTO data set has an average uranium value of 7.7 ppm and an average thorium value of 3.8 ppm. More detailed uranium and thorium data are available in the other reports listed; samples are located as to the specific Morrison member, and comparisons are made between the whole rock and the less than 2 micron fraction. Whole-rock uranium and thorium data for the Morrison found in Brookins (1979a and b) and uranium data in Brookins and Della Valle (1977) are provided as Table 6, which also furnishes uranium values for some surrounding formations.

Further examination of the Morrison uranium data in Brookins (1979a) reveals an overall Morrison sample average value of 7.25 ppm U. If instead, the overall average is estimated by in turn averaging the average values for the different units, a value of 8.46 ppm U is calculated; however, this calculation poorly reflects the fact that many of the high uranium units, such as the Jackpile and Poison Canyon sandstones, are relatively thin and not laterally extensive. An average determined by weighting the unit averages by the thickness of the individual units would be significantly lower, perhaps 6 ppm or so, depending on the thicknesses used for the determination. Similarly, unit thorium concentrations can be averaged or weighted averaged resulting in values of 9.53 ppm or perhaps 6.5 ppm, respectively.

The uranium concentrations discussed so far have all been for rocks considered not uranium mineralized. Uranium concentrations for mineralized samples (greater than 100ppm U), as provided in Spirakis and Pierson (1986) for many of the uranium mining dis-

Table 6. Average uranium and thorium concentrations of whole rock (non-ore) samples from selected units.

	Brookins & Della Vella, 1977			Brookins, 1979a							
Formation, Map Unit, or Member	Range	N	Av. U (ppm)	Range	N	Av. U (ppm)	Range	N	Av. Th (ppm)	Th/U (1)	Th/U (2)
Madera (P-Pm)	1.68-16.9	19	5.65								
Todilto (Jt)	0.57-3.45	12	1.39								
Morrison (Jm)											
Recapture	0.63-21.1	7	6.64	0.72-21.1	9	6.30	0.47-17.7	5	6.77	1.07	1.93
Westwater Canyon											
non-ore;outcrop	0.91-10.3	13	3.63	1.11-10.7	12	3.77	0.81-12.6	9	4.97	1.32	1.47
core;oxidized	1.65-56.2	20	14.5								
core;reduced	2.04-70.6	11	20.6								
Chaco Canyon(3)	0.68-564.	80	2.0								
Poison Canyon				1.97-31.1	17	8.73	6.79-7.82	2	7.31	0.84	0.82
Brushy Basin				2.65-21.2	8	8.91	8.46-16.6	5	11.8	1.33	1.82
Jackpile Sand.				5.05-31.0	5	15.7	ND-41.76	5	16.8	1.07	2.89
Dakota (Kd)				0.37-14.6	13	4.54					
bentonite-rich	5.32-14.6	5	8.86								
sandstone	0.36-6.15	8	1.82								
Mancos Shale (Km)	2.85-4.99	10	3.98								

Notes:
(1) Ratio of average values
(2) Average ratio of sample ratios
(3) Chaco Canyon data from Brookins, 1979b

tricts in the Grants Uranium Region, indicate average uranium values for Morrison ore rocks of 1,068 to 1,817 ppm (approximately 0.125 to 0.214 weight percent U308). Ore has also been reported within the basin for units not in the Morrison Formation, including the Shinarump Conglomerate, Chinle Formation, Entrada Sandstone, Todilto Limestone, Dakota Sandstone, and Mesaverde Group. Distribution of mineralization, and uranium within mineralized zones, is often unclear. For example, Brookins (1979b) reported the interception of nine different zones of low-grade uranium mineralization by five boreholes in the Westwater Canyon Member in the East Chaco Canyon area. The whole-rock average uranium concentration for the Westwater Canyon in the area was estimated as 2 ppm, but whole-rock values as high as 594 ppm were measured. In the same report, Brookins also described an eight foot section of 0.125 weight percent U_3O_8 in the Brushy Basin Member that had been observed in one of the drill holes.

Figure 6 shows active and proposed uranium mines in the San Juan Basin as of 1978. A number of mine sites and probable mine sites are designated in areas well away from the Grants Uranium Region. Interest in these sites has diminished with the changed economics of uranium mining and information on these sites is not readily available. The many such sites and their scattered distribution indicates the need for caution in interpreting decay calculations.

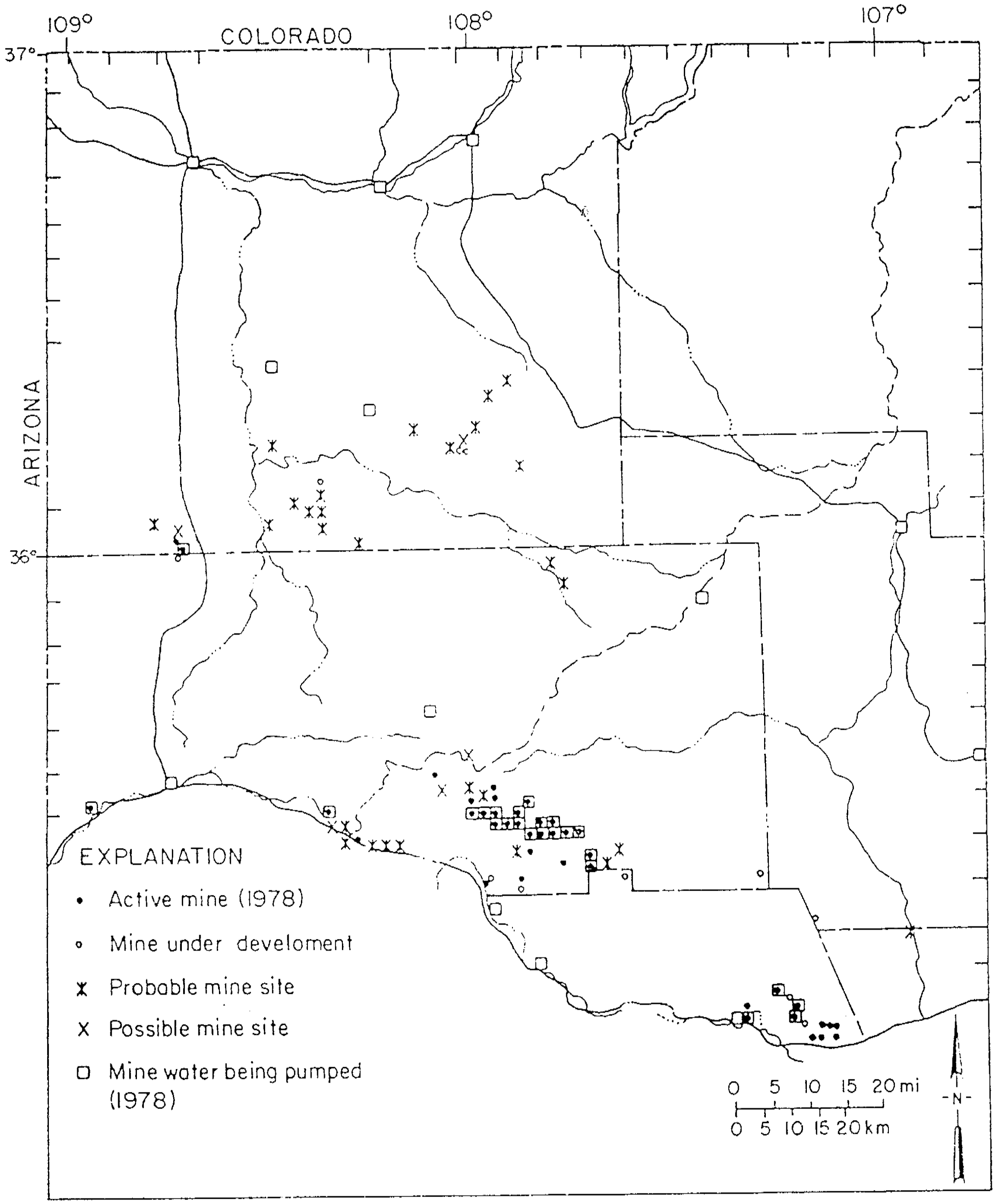


Figure 6. Location of active and proposed uranium mines in Grants mineral belt (Stone et al., 1983; originally from Perkins, 1979).

Ratios of uranium to thorium are also of interest in developing reasonable estimates of data values. Table 6 gives the ratio of thorium to uranium as determined for individual units from the data of Brookins (1979a). Values for a given unit are given for two methods of calculation; the ratio of the average uranium and thorium values, and the average of individually measured sample ratios. The ratios of the average uranium and thorium values are consistently lower than the ratios determined by averaging ratios calculated for individual samples. The individual sample ratios range from 0.42 to 5.42, and based on the data presented in the table, an overall average ratio might be between 1.0 and 1.8. In addition, examination of the sample ratio data led to an observation that high-uranium mineralized samples need not have comparable levels of thorium; or, in other words, that a lower than average ratio was applicable for high uranium samples.

Besides the chemistry, the porosity and specific weight are important inputs to the CLSEC model. The high percentage of silica observed in the geochemical analyses indicates the specific weight can be well estimated by using that of quartz, 2.65. The porosity can be expected to be variable; the Morrison Formation is composed of both shale and sandstone units. Based on ranges and average values given for different rock materials in the hydrology texts McWhorter and Sunada (1977) and Freeze and Cherry (1979) the porosity is estimated as between 20 and 40 percent. Particularly as the porosity applicable to the CLSEC calculations is the entire water filled porosity, an expected average of about 30 percent is suggested.

The neutron flux and $^{36}Cl/Cl$ results for a number of secular equilibrium calculations are shown on Table 7. The input and output information given on the table summarizes the runs given in Appendix B.2. Sensitivity of the calculations to some of the estimated parameters is apparent. Various concentrations of uranium and thorium are tried, although many of the sensitivity runs are focused on a base data set with uranium 8 ppm, thorium 12 ppm, the SSJB geochemical data, a porosity of 30 percent, a rock chloride concentration of 100ppm, and samarium and gadolinium concentrations of 4ppm (run # 5). As shown on the table, the Cl concentration in the rock and the choice of data set appear to make little difference to the calculations. The mineralized Smith Lake data set does appear to be conducive to a somewhat higher neutron flux, but data in Haddad et al. (1981) suggest that the neutron adsorbers Sm and Gd may also be more concentrated in the more mineralized rocks, which would lead to relatively unchanged equilibrium ratios.

As demonstrated in runs 7 and 8, the secular equilibrium calculation is somewhat sensitive to porosity estimates. The calculated secular equilibrium is inversely proportional to the estimated porosity. The base-case porosity of 30 percent may be somewhat higher than the true average, but then again the true average uranium concentration might well be less than 8 ppm. Therefore, unless locally measured input parameters are used, an

Table 7. Results of secular equilibrium calculations.

	Input						Output	
Run#	U (ppm)	Th (ppm)	Data Set	Porosity	ppm Cl in Rock	Sm and Gd (ppm)	Neutron Flux per Kg Media (n/yr)	Secular Equilibrium
1	3	4.5	SSJB	0.30	100	4	3.6E3	7.24E-15
2	5	7	SSJB	0.30	100	4	5.9E3	11.8E-15
3	8	4	SSJB	0.30	100	4	7.8E3	15.7E-15
4	8	4	PLUTO	0.30	100	4	7.9E3	15.8E-15
5	8	12	SSJB	0.30	100	4	9.6E3	19.3E-15
6	8	12	PLUTO	0.30	100	4	9.8E3	19.5E-15
7	8	12	SSJB	0.20	100	4	10.6E3	24.4E-15
8	8	12	SSJB	0.40	100	4	8.5E3	14.9E-15
9	8	12	SSJB	0.30	200	4	9.6E3	19.2E-15
10	8	12	SSJB	0.30	50	4	9.6E3	19.4E-15
11	8	12	SSJB	0.30	100	8	9.6E3	17.6E-15
12	8	12	SSJB	0.35	200	8	9.1E3	15.6E-15
13	8	8	SSJB	0.30	100	4	8.7E3	17.5E-15
15	18	9.8	SSJB	0.30	100	4	17.7E3	35.6E-15
16	18	9.8	PLUTO	0.30	100	4	18.1E3	35.9E-15
17	100	30	SSJB	0.30	100	4	93.E3	185.E-15
18	100	30	S. LAKE	0.30	100	4	100.E3	189.E-15
19	1000	100	SSJB	0.30	100	4	886.E3	1,770E-15
20	1000	100	S. Lake	0.30	100	4	947.E3	1,790E-15

estimate of the average secular equilibrium ratio in non-mineralized Morrison rocks of 15-20 x 10^{-15} appears reasonable based on Table 7 results.

The equilibrium ratio applicable to the more mineralized rocks would obviously be much higher. Even using the average uranium and thorium values from the SSJB and PLUTO data sets respectively, results in a ratio over 35 (run # 14), and ore concentrations can easily lead to calculated equilibrium ratios over 1,800 x 10^{-15} (see run 19). Work by Andrews et al. (1986) has demonstrated the complications of subsurface production of ^{36}Cl.

Dating equation calculations

The ability of the ^{36}Cl results to help with an understanding of aquifer system properties is largely dependent on the calculated system water "ages" and their spatial distribution. As described in detail in the approach section, different methods of calculating the decay time, or "age", are appropriate for different flow path processes. Often the question of the most appropriate equation can be addressed by comparative examination of the calculation results. This section presents the results of the most commonly applied dating equations and examines the sensitivity of theses results to different given inputs.

The three equations attempted are; the simple decay equation

(equation 4), the addition of secular equilibrium chloride (equation 9 version of equation 8), and the addition of dead chloride (equation 10 version of equation 8). These three equations are equivalent to those applied in Bentley et al. (1986a) and Phillips et al. (1986b). All of these equations require estimates for the recharge ratio and the secular equilibrium ratio. Equation 4 is otherwise based only on the sample ratio and assumes conservation of ^{36}Cl mass; however, the equation is not restrictive with regards to processes that alter the water volume. Equations 9 and 10 require an estimate of the recharge chloride concentration, and sample chloride concentration values greater than the recharge value are attributed to the respective sources. In these last two equations, the water volume is assumed unchanged, and, as the quantity of chloride input from the additional source is calculated from the concentration change, they are not applicable to volumetric dilution or concentration processes.

Estimates for the secular equilibrium ratio have already been discussed; however, estimates are also needed for the recharge ratio and the recharge chloride concentration. The graph on Figure 1, of the ratios associated with dry precipitation across the United States, suggests that the precipitation ratio for pre-bomb conditions is greater than 640×10^{-15} . In fact, projecting the trend on the diagram into the area of the San Juan Basin suggests a precipitation value well over 640×10^{-15}. Ratios of measured samples from the younger units in the vicinity of outcrop also suggest higher recharge ratios; well 27 in the Dakota is reasonably close to potential recharge from the flanks of the Chuska Mountains and has a measured ratio of 730 x 10^{-15} , and well 29 in the Gallup Sandstone is adjacent to Gallup outcrop in the same area as the Dakota well and has a measured ratio of 676. As the 730 x 10^{-15} value observed in the Dakota may be influenced by some high ratio waters seen in the Morrison in that area, a compromise value of 700 x 10^{-15} seems a reasonable estimate of the recharge ratio. This value is similar to that of 717 x 10^{-15} measured for waters below 2 meters in depth at site near Socorro, New Mexico (Phillips et al., 1984)

The appropriate value for the recharge chloride concentration can be estimated from the measured system values. The two lowest chloride concentrations measured were 0.8 and 1.1 mg/L, but these values are associated with ^{36}Cl ratios of only 36 and 41.9, respectively, and so do not represent recharge waters. A number of samples show chloride concentrations of approximately 3 mg/L and have near-recharge type ^{36}Cl ratios; however, there are a number of samples that suggest a recharge ratio of closer to 8 mg/L. The most generally applicable value to use is left undecided for now, but the higher value will call for less correction due to chloride addition, which may be advantageous. In any event, the affects of differing the recharge chloride concentration will be further examined in our sensitivity runs.

The equations are not always solvable for each sample data;

the log term in each of the equations can be examined to determine the possible problems. For the results presented in this section (Table 8), an error code has been devised to indicate the cause of the problem. First, if the sample ratio is greater than the recharge ratio, then there has been buildup rather than decay and this cannot be accounted for by any of the equations. Such an occurrence should be uncommon, but is physically realistic if local sample point conditions have led to either contamination by bomb-pulse waters or very high secular equilibrium ratio chloride. A result of *1 is presented for this case.

Second, the corrections to the log term in equations 9 and 10 associated with the addition of aquifer matrix chloride can result in apparent negative ages. A calculated negative age occurs when the log term exceeds a value of 1, which it is apt to do when the sample to initial chloride ratio is large and the measured sample ratio is high. This condition, which implies that the chloride concentration has been changed by processes other than those described by equations 9 and 10, has been reported as *2 in the presented results.

Third, if the sample ratio is less than the secular equilibrium ratio estimate, then the only equation that makes sense is that regarding the addition of dead chloride; the result for the other equations can not be calculated because the log term becomes negative. For cases other than overwhelming addition of dead chloride, such a sample is apparently near decayed in terms

Table 8. **Sample ages (x 10 $^{-5}$) calculated using decay equations (as indicated) from text.**

Sample #	^{36}Cl/Cl Ratio	Cl (Mg/L)	^{36}Cl (atm/L)	Age (4A)	Age (4B)	Age (9)	Age (10)
Morrison Samples:							
14	244	11	4.6	4.63	4.83	3.44	3.34
15	15.4	210	5.5	17.9	*3	*3	2.48
16	44.5	11	0.8	12.4	14.5	13.1	12.2
17	42.0	61	4.4	12.6	14.9	6.08	3.55
18	58.9	57	5.7	11.0	12.4	3.90	2.31
19	4.0	750	5.1	*3	*3	*3	2.85
20	37.9	120	7.7	13.1	15.8	4.04	0.93
22	41.5	83	5.9	12.7	15.0	4.85	2.19
23	333	23	13.0	3.26	3.37	*2	*2
24	213	38	13.8	5.23	5.48	*2	*2
25	295	10	5.0	3.79	3.94	2.97	2.90
26	251	3.9	1.7	4.50	4.69	--	--
27	36.0	0.8	0.1	13.4	16.3	--	--
28A	163	28	7.8	6.42	6.78	1.33	0.92
28B	14.9	64	1.6	18.1	*3	*3	8.37
29A	407	2.5	1.7	2.38	2.45	--	--
29B	417	3.0	2.1	2.27	2.34	--	--
30	573	2.7	2.6	0.88	0.90	--	--
31	2201	1.6	6.0	*1	*1	--	--
32	41.9	1.1	0.1	12.7	14.9	--	--
33A	400	43	29.2	2.45	2.53	*2	*2
33B	121	12	2.5	7.75	8.29	6.53	6.25
33C	274	8.4	3.9	4.12	4.28	4.07	4.05
34A	481	7.8	6.4	1.64	1.69	--	--
34B	1857	7.1	22.4	*1	*1	--	--
35	290	19	9.4	3.87	4.02	0.26	0.07
36A	648	14	15.4	0.34	0.35	*2	*2
36B	537	17	15.5	1.16	1.19	*2	*2
37	894	6.5	9.9	*1	*1	--	--
38	630	7.2	7.7	0.46	0.47	--	--
Dakota Samples:							
11	169	57	16.4	6.26	6.60	*2	*2
12	730	2.6	3.2	*1	*1	--	--
13	149	78	19.7	6.82	7.23	*2	*2
Gallup Samples:							
1	676	7.8	9.0	0.15	0.15	--	--
2	198	49	16.5	5.55	5.83	*2	*2
4	122	4.1	0.9	7.72	8.25	--	--
5	153	45	11.7	6.70	7.09	*2	*2
6	22.0	5.0	0.2	15.9	25.4	--	--
7	569	9.5	9.2	0.91	0.93	0.18	0.16
8	537	23	21.0	1.16	1.19	*2	*2
Point Lookout Sample:							
39	18.4	840	26.3	16.9	*3	*3	*2

Notes:

*1	R > Ro	Ages in years	
*2	Log term negative	--	C < Co
*3	R < Rse		

of recharge chloride, and the result (on Table 8) is set to *3.

Finally, if the sample chloride concentration is less than the estimated recharge chloride concentration, equations 9 and 10 give erroneous results. The equations are not designed for application to dilution processes; they would be actually correcting for a chloride sink having lowered the chloride concentration instead of a source having raised it. Assuming that the result best suited in this case is that associated with no chloride addition, the reduction in the chloride value is ignored and the result of the simple decay equation, equation 4, is best regarded.

The results of a few sample "age" equations are given in Table 8. The first two columns of ages are both the result of equation 4; they differ in the secular equilibrium ratio that was used. A ratio of 4 was used in the first column, a value equal to the lowest of the sample measurements. A value of 20 was used for the second column, a value considered more generally applicable to the samples taken. As can be observed, the results of the two columns are very similar, with the higher secular equilibrium ratio leading to slightly older calculated ages. This is as expected since the greater equilibrium ratio implies that more ^{36}Cl will have to decay to achieve the same sample ratio.

The other two columns are the results of equations 9 and 10, respectively. The recharge ratio used was 700 x 10 $^{-15}$ and the recharge chloride concentration used was 8 mg/L. There are not many samples in the 3 to 8 mg/L range to be affected by the potentially high value chosen for the recharge concentration, and a number of the equation 9 and 10 results would probably have listed as a *2 error if a lower value was used. Even so, Table 8 shows there are quite a few samples for which equations 9 and 10 could not be successfully applied. In general, the results show the lowering of ages associated with the addition of a low ratio chloride source, and in some cases the difference in the age calculated is quite dramatic. The results of the last two columns are very similar, with the last column reflecting slightly younger ages due to the lower ratio of the added matrix chloride.

The ages from the first column for the Morrison samples have been plotted as Figure 7 to allow examination of the areal distribution of the calculated ages. The ages show some expected trends, as the ages near the San Juan river are older and ages near the Morrison outcrop are generally younger, but there is a great deal of unexpected variability; Ages in the outcrop areas are older than expected, whereas ages in the central basin are much younger than expected. Figure 8 shows the same type of plot for the results in the last column of ages. Correcting the ages for addition of dead chloride greatly affects a number of calculated values, with many of the wells by the river now showing much younger ages due to their high chlorides and resulting high degree of correction. Clearly, trends along the flow-lines sus-

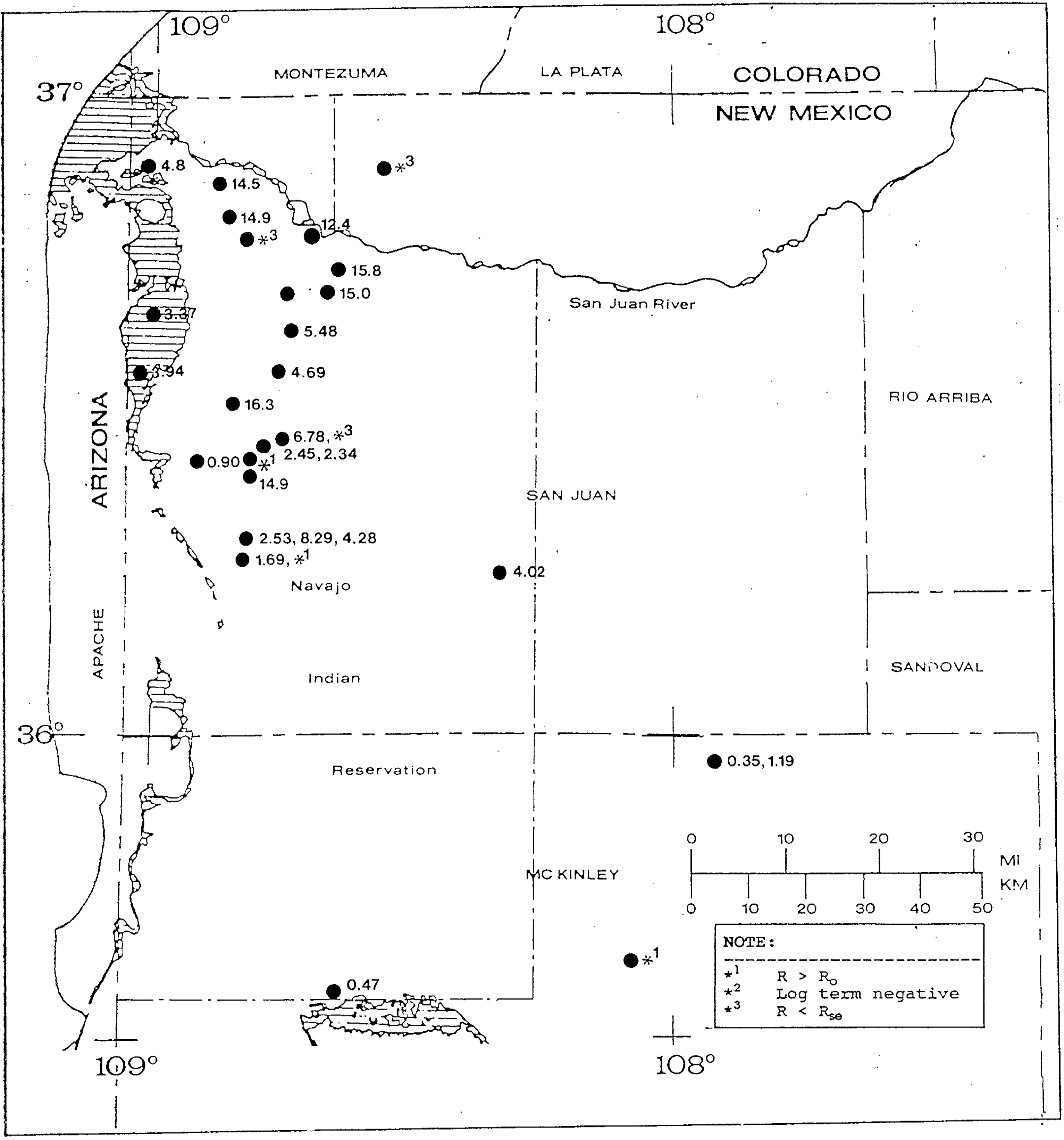


Figure 7. Ages (years x 10^{-5}) calculated using equation 4 (simple decay) with $R_{se}=20 \times 10^{-15}$ and $R_o=700 \times 10^{-15}$. Multiple samples listed in chronological order.

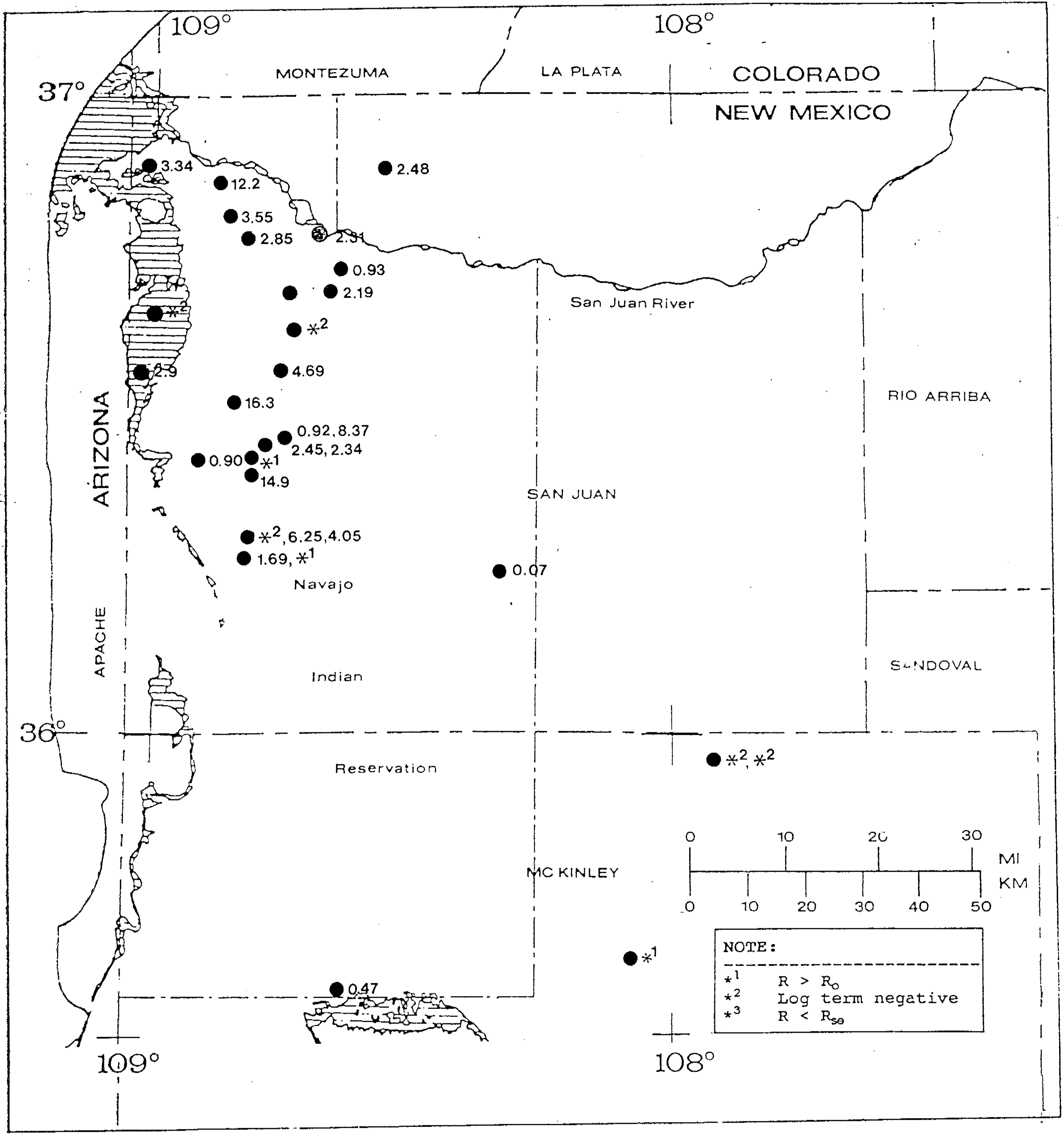


Figure 8. Ages (years x 10^{-5}) calculated using equation 10 (simple decay with dead chloride addition) with $R_{se}=20 \times 10^{-15}$, $R_o=700 \times 10^{-15}$, and C_o=8 mg/L. Multiple samples listed in chronological order.

pected from Figure 4 do not immediately suggest the correctness of either of the age equations, and further analysis will be necessary to determine the best equation to apply to different sample measurements.

The sensitivity of the age calculations is explored in Table 9. This table presents the results of each of the equations, 4, 9 and 10 for each of three samples with a variety of specified input parameters. The samples for which the ages were calculated are from wells of suspected chloride addition, 16, 22, and 25, and represent a number of ^{36}Cl ratios and chloride concentrations. All of the input data sets are easily within a range of possible values. The simple decay calculations are relatively consistent. Ages of 350,000 to 1.5 million years seem somewhat old, but are possible. Younger ages are calculated by assuming addition of chloride with a low isotopic ratio, but the calculation is sensitive to the input parameters. Even corrected ages imply the water is old; well 25 is in an outcrop area, and the youngest calculated age was 36,000 years. Clearly, the solute assemblage could have been changed through mixing with chloride from other sources.

Table 9. Sensitivity of Cl-36 calculated ages for three wells (x 10-5) to changes in input data.

Input Data			Well 22 (Cl-83.,R=41.5)			Well 25 (Cl=10., R=295.)			Well 16 (Cl=11., R=44.5)		
Ro	Rse	Co	Age 4	Age 9	Age 10	Age 4	Age 9	Age 10	Age 4	Age 9	Age 10
700.	20.	8.	15.02	4.85	2.19	3.94	2.97	2.90	14.45	13.07	12.13
700.	20.	4.	15.02	1.83	-.92	3.94	-.05	-.23	14.45	10.05	8.23
700.	20.	12.	15.02	6.61	4.06	3.94	3.94	3.94	14.45	14.45	14.45
700.	4.0	8.	12.70	2.53	2.13	3.79	2.82	2.81	12.37	10.98	10.87
700.	4.0	4.	12.70	-.48	-.91	3.79	-.19	-.23	12.37	7.97	7.70
700.	4.0	12.	12.70	4.29	3.91	3.79	3.79	3.79	12.37	12.37	12.37
650.	20.	8.	14.69	4.51	1.86	3.60	2.63	2.57	14.12	12.37	11.86
650.	20.	4.	14.69	1.50	-1.26	3.60	-.38	-.57	14.12	9.72	7.90
650.	20.	12.	14.69	6.28	3.73	3.60	3.60	3.60	14.12	14.12	14.12
650.	4.0	8.	12.38	2.20	1.80	3.47	2.50	2.49	12.04	10.66	10.54
650.	4.0	4.	12.38	-.81	-1.23	3.47	-.52	-.55	12.04	7.64	7.38
650.	4.0	12.	12.38	3.97	3.59	3.47	3.47	3.47	12.04	12.04	12.04
800.	20.	8.	15.61	5.44	2.79	4.53	3.56	3.50	15.05	13.66	12.79
800.	20.	4.	15.61	2.43	-.33	4.53	.55	.36	15.05	10.65	8.83
800.	20.	12.	15.61	7.21	4.66	4.53	4.53	4.53	15.05	15.05	15.05
800.	4.0	8.	13.28	3.11	2.71	4.38	3.40	3.39	12.95	11.56	11.45
800.	4.0	4.	13.28	.10	-.32	4.38	.39	.36	12.95	8.55	8.29
800	40.	8.	27.08	16.91	2.89	4.75	3.78	3.64	22.30	20.92	15.56

Note: Negative calculated ages given for comparison purposes.

Ages are in years

DISCUSSION

Concerns

The hypothesis of simple decay along suspected piston-flow water paths does not fit much of the measured ^{36}Cl data. This is true even if the addition of dead or secular equilibrium chloride is included. The extremely varied distribution of the age calculation results presented in Figures 7 and 8 clearly illustrate the need to consider additional important processes. The ^{36}Cl data must be more completely regarded as that of a hydrochemical tracer--a tracer capable of volumetric mixing, underground production, and simultaneous decay of chloride from multiple sources with different salinities. Large differences in the isotopic ratio and concentration values, even for wells located in close proximity to each other, commonly are observed in the Morrison Formation data. In addition, the multiple samples at three wells, 28, 33, and 34, yield inconstant data. Changes are with regard to the chloride concentration as well as the ^{36}Cl isotopic ratio.

The analytical results are not in question, but the assumptions applied with regard to the hydrology and related hydrochemistry are. Mixing of waters in wells with such long open intervals is undeniable. The mixing described by equation 11 is applicable to distinct sources mixing in a simple manner, at an outlet. In complex mixtures, the concept of a composite groundwater age is of questionable use. As stated by Andrews et al. (1986);

> "In cases where the groundwater chloride content increases due to interaction with the rock matrix, the specific activity of ^{36}Cl in solution must attain the equilibrium specific activity of ^{36}Cl in the rock matrix, as solution proceeds. No information concerning groundwater residence time can then be deduced, but the admixture of chloride from different rock matrices or with input chloride can be estimated."

The ^{36}Cl ages calculated may reflect a relative change in the entire chloride composition, not just a residence time decay along a flow-path. To develop and understand the possible explanations, a more in-depth look at the relation of the ionic chloride measurements to the $^{36}Cl/Cl$ measurements is required.

To assist with a further examination of the data, the data from the Morrison Formation have been first grouped geographically to allow for easier identification of nearby data points. Figure 9 shows the five groups used. These include: group 1--a group of wells near the river, most of which show elevated chloride concentrations; group 2--a group of wells in the northwestern outcrop area with moderate ^{36}Cl ratios; group 3--a group of wells in the western portion of the basin leading away from the Defiance Uplift recharge area; group 4--a group of wells just off of the Defiance Uplift area; and group 5--a group of wells in the central and southern portions of the basin, many with ^{36}Cl ratios higher than expected. Data from the Gallup and Dakota sandstones is much less extensive and is presented only in reference to an

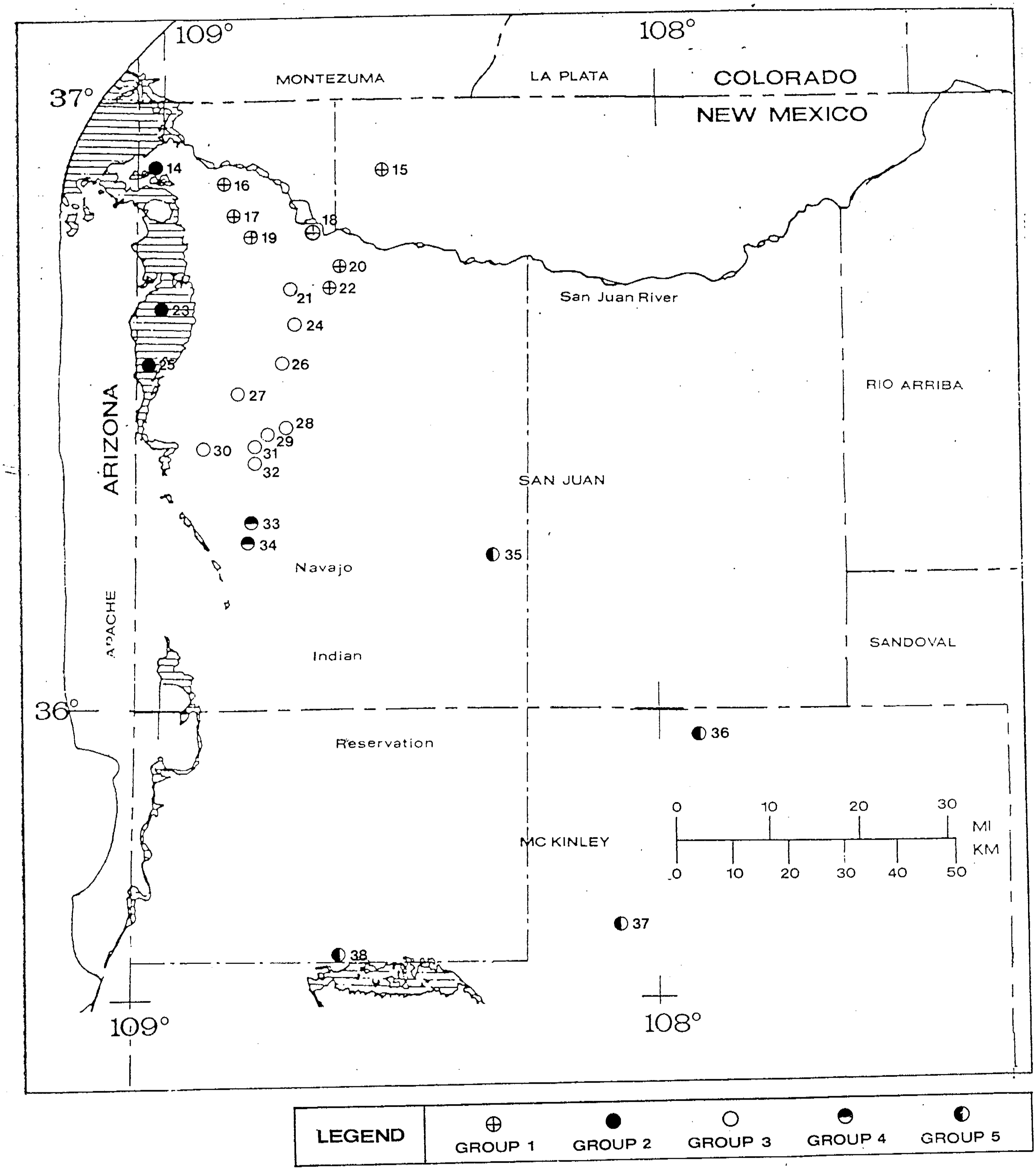


Figure 9. Map showing Morrison Formation wells sampled and geographic groups used in analysis.

analysis of the Morrison data.

An interpretation of the chloride data (both Cl^- and $^{36}Cl/Cl$ data) involves the search for a process or combination of processes that can explain the data measured. The processes described in developing the dating equations in the approach section of this report suggest that there are three types of processes to consider; those associated with the decay or buildup of ^{36}Cl in the aquifer pore water, those associated with the dissolution or release of chloride from the rock matrix, and those processes that involve mixing of waters. In complex systems, more than one explanation for a given data trend is often conceivable.

Decay of recharge ^{36}Cl, and the relation of that decay to residence time, have been the focus of the age dating equations applied in Table 8. The entire Morrison sampling area was regarded as one system requiring one explanation. In fact, processes important in one area may be distinct from processes important in other areas. Variability in the aquifer system is to be expected, and a universally applied conceptual model may be very inappropriate.

The variability in the ratio measurements and secular equilibrium calculations suggest the potential importance of ^{36}Cl buildup due to uranium/thorium induced production. Much improved sources of uranium distribution data (New Mexico Energy and Minerals Department, 1981; McLemore, 1983; McLemore and Chenoweth, 1989) were reviewed, with the area sampled in mind. The distribution of uranium in the northwest portion of the San Juan Basin as reported by these sources is well represented by Figure 10 (from McLemore, 1983). The New Mexico Energy and Minerals Department report further supports the occurrences shown on the figure to the northeast of the Enos Johnson mine toward Sheep Springs, an area not discussed in McLemore and Chenoweth (1989) and an area poorly described by Stone et al. (1983, this report Figure 6).

The isotopic ratio versus the Cl-36 concentration

Distinct decay and buildup processes are best examined using the relationship of the measured ^{36}Cl/Cl ratio to the calculated ^{36}Cl concentration. Recall, the ^{36}Cl concentration is a function of the product of the ^{36}Cl/Cl ratio and the chloride concentration as described in equation 1b. In the case of a simple decay or buildup process, the total chloride concentration remains unchanged, and the relation between the ^{36}Cl/Cl ratio and the ^{36}Cl concentration is linear. The slope of the line is dependent on the constant chloride concentration (and the constant unit conversion factor k). On a plot of ^{36}Cl/Cl ratio versus ^{36}Cl concentration, "... samples which are derived from a common origin by only radioactive decay will fall on a straight line between the initial composition and the origin" (p. 146, Phillips et

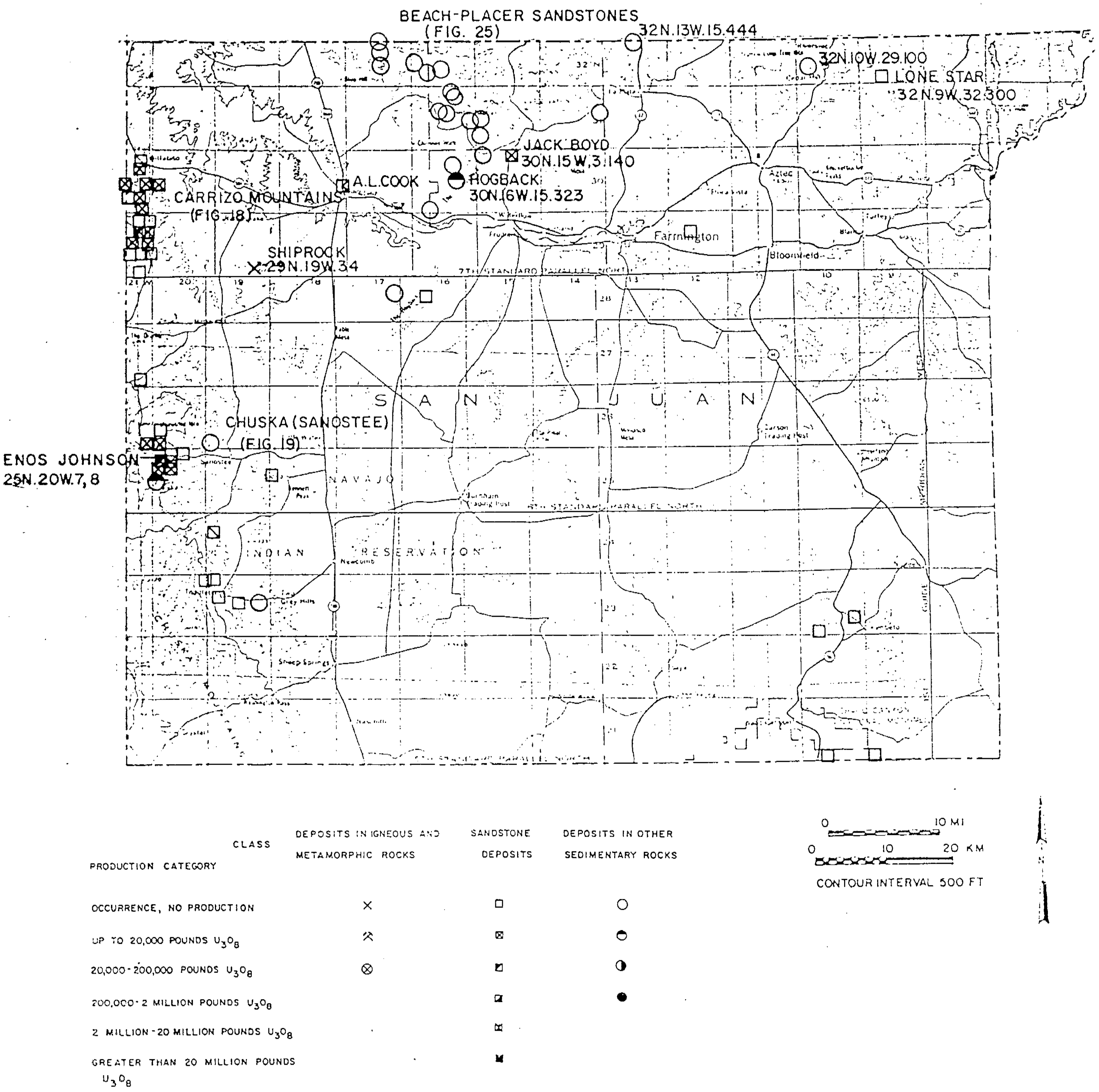


Figure 10. Radioactive occurences in San Juan County, New Mexico (McLemore, 1983).

al., 1986). This is true of buildup from an initial composition as well as decay.

Figures 11 and 12 present the $^{36}Cl/Cl$ ratio versus ^{36}Cl concentration relationships for the measured Morrison samples. Two plots are given to allow for detailing of densely plotted data. The sample data are labeled as to the well sampled, and, in cases of multiple sampling, are also lettered chronologically. A few neighboring Dakota and Gallup wells, which can be located on Figure 5, are included. Figure 11 illustrates the displacements caused by various processes other than volumetric mixing.

On both figures, three proposed decay/buildup lines are drawn, reflecting the decay or buildup of ^{36}Cl in water of differing initial salinities. The steepest decay line represents the most dilute water, and is drawn through sample 30 and to the origin. Sample 30 has measured $^{36}Cl/Cl$ ratio and chloride concentration values (573 x 10-15 and 2.7 mg/L, respectively) approaching those expected of a recharge water. In addition, well 30 is geographically located at a short distance from a major recharge area--the flanks of the Chuska Mountains near the Defiance Uplift (see Figures 2 and 9).

The middle decay/buildup line is drawn through sample 34B to the origin. The line goes nearly through the data point for sample 34A, which indicates 34B may represent a potential buildup of ^{36}Cl in water of a similar salinity. Interestingly, the

Figure 11. $^{36}Cl/Cl$ vs. ^{36}Cl for Morrison Formation waters. The displacement caused by various processes are illustrated. Figure 12 provides additional detail.

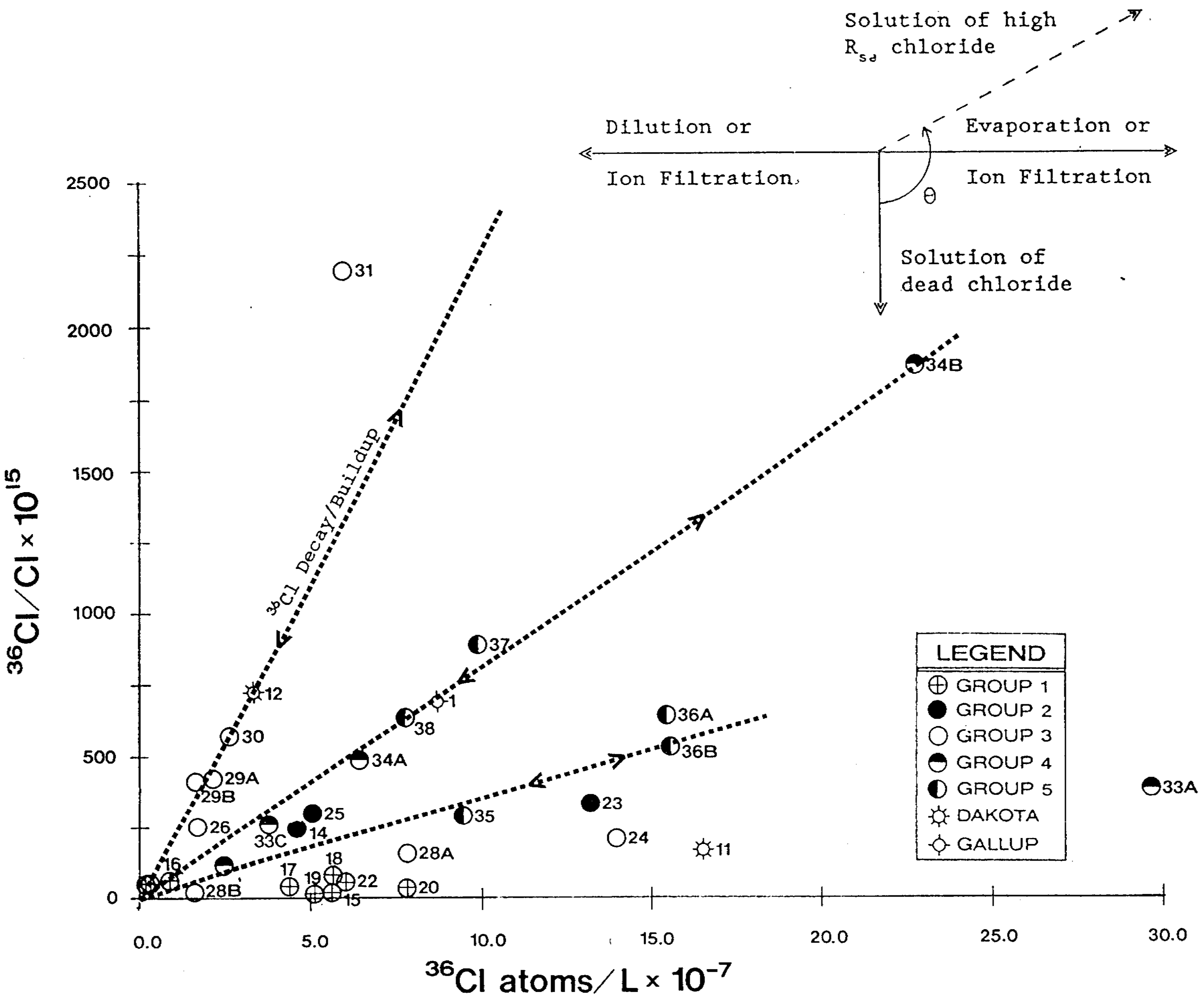
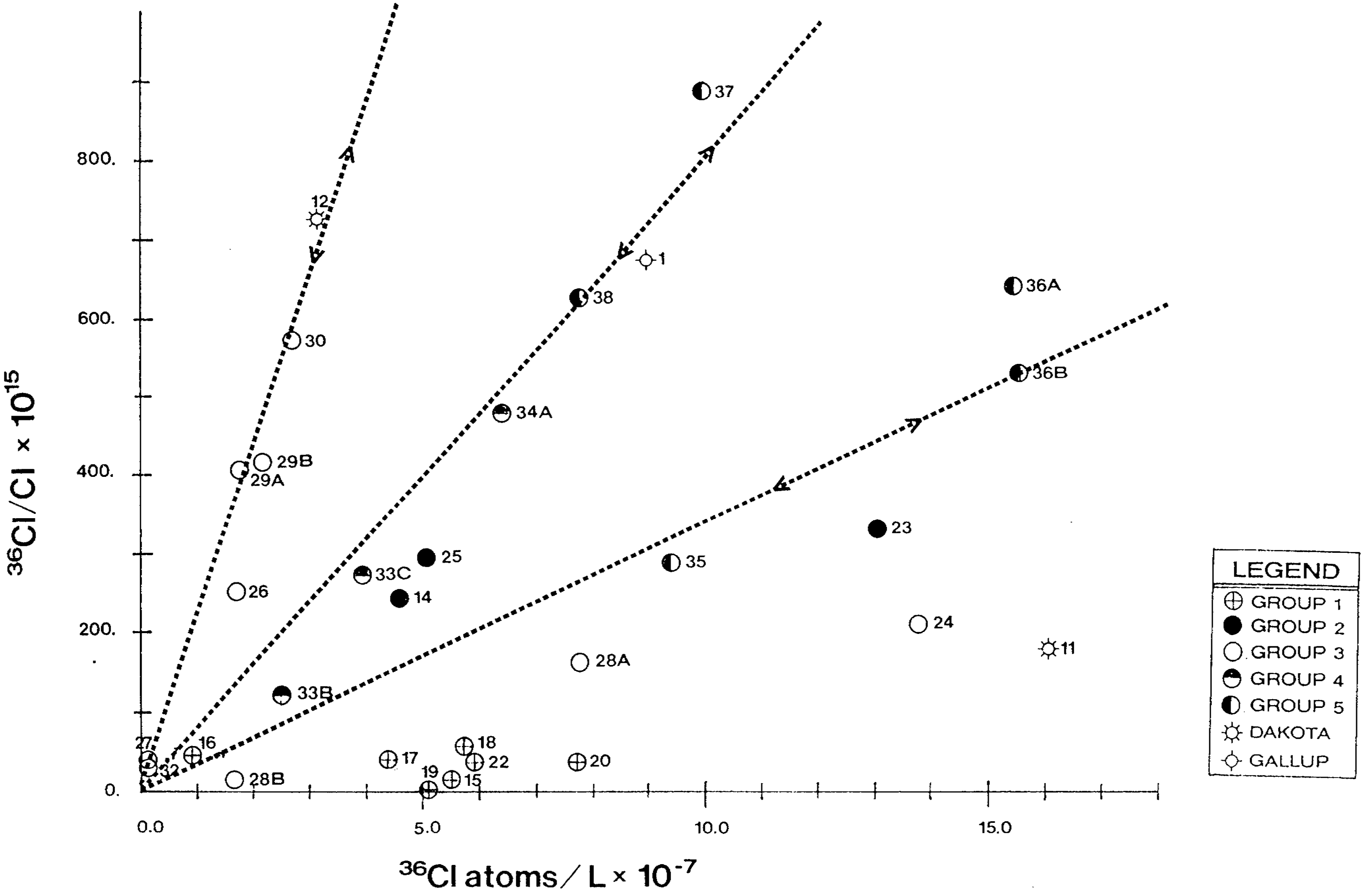


Figure 12. $^{36}Cl/Cl$ vs. ^{36}Cl for Morrison Formation waters. Expanded scale shows portion of figure 11 in more detail.



samples representing wells in or immediately adjacent to outcrop areas all plot in the vicinity of this decay line.

The decay line drawn with the lowest slope represents decay or buildup in waters with higher salinities. This line is drawn through sample 36B and to the origin. The line is drawn to suggest the contribution of water in well 35 containing chloride decayed while in travel from well 36. Treating the water at well 36 as an initial water reflects the significant secular equilibrium contribution to the ^{36}Cl concentration observed at Chaco Canyon. The very high chloride waters from wells near the river plot as a group, below the slope of any suspected decay line.

Figure 11 also shows the displacements caused by various processes other than decay. Again, non-decay processes shown are those that alter the chloride composition other than by volumetric mixing of different waters. Evaporation, dilution, and ion filtration can each serve to alter the total chloride concentration, but neither of them change the 36Cl/Cl ratio (Phillips et al., 1986b). In these cases, the 36Cl concentration value is the only variable affected, and a horizontal displacement takes place, as can be seen on Figure 11.

Evaporative processes have been invoked in the Bentley et al. (1986) study of the Great Artesian Basin to explain variability of chloride concentrations in recharge waters. Ion filtration has been used in the Phillips et al. (1986) study of the Milk

River Aquifer to explain chloride increases in a distally pinching aquifer. As noted on Figure 11, ion filtration can have either a diluting or concentrating effect, depending on which side of the geologic "membrane" is being sampled and how the concentration changes are proceeding.

The concentration changes in an ion filtration system are dependent on location in the system, direction of water flow, and the open or closed nature of the system. The direction of water flow depends on the balance of concentration and pressure gradients in the system (driving mechanisms) and system conditions and boundaries. In the system proposed by Phillips et al. (1986) for the Milk River Aquifer, the aquifer water is on the residual side of a membrane, leading to an increase in concentration. Presumably this would continue until some type of equilibrium is achieved where the flux of chloride in and out of the aquifer system become equal. Assuming an initial chemical balance with the residual side of a the system, the solute flux on the filtrate side of such an aquifer system would be initially dilute and only later become more concentrated.

Figure 11 shows the displacements on this type of plot caused by the solution of aquifer chloride. Since chloride is being added, not water, the concentration of ^{36}Cl cannot be decreasing. The added chloride causes a positive (or zero) ^{36}Cl concentration displacement. The direction of this displacement (angle θ on Figure 11) is dependent on the quantity and ^{36}Cl/Cl ratio of the chloride added. Solution of dead chloride does not add ^{36}Cl, and the displacement is vertically downward ($\theta=0$). Solution of chloride with the same ^{36}Cl/Cl ratio as the aquifer water does not alter the ratio and the displacement is horizontal ($\theta=90$ degrees). In the case where secular equilibrium chloride being dissolved has an isotopic ratio greater than that of the aquifer water, the displacement is not only in the increasing ^{36}Cl concentration direction, but has a positive slope ($\theta>90$ degrees).

Figures 11 and 12 show a wide scattering of data points reflective of the complex nature of the Morrison Formation ^{36}Cl data. This complexity has been suggested previously by the attempts at applying simple dating equations (Figures 7 and 8). A significant aspect of this complexity is the frequent lack of consistency between samples taken from the same well at different times. Wells 33, 34, and 28 all have multiple samples that are extremely different, whereas wells 29 and 36 have multiple samples that are more consistent. Surprising, is the occurrence of sample variability in wells that have been, for the most part, freely flowing for more than a decade. It is true, however, that all three wells showing variability have open intervals of greater than 750 feet (as indicated on Table 2). Well 33 is also one of the few wells not cased through the Brushy Basin Member of the Morrison, and wells 33 and 28 both extend more than 100 feet beyond the Morrison into the Wanahka Group.

With even a slight transient change in the pressure regime, the mixture in the well of waters from stratified layers within and around the Morrison will change composition. Hydraulic conductivity variations from layer to layer will dictate which layer produces relatively more water then before. The persistence of such dramatic changes in the solute composition after such a long period of well flow suggests that the flux distributions in the wellbore have not yet reached even a quasi-steady state. A system consisting of an open well penetrating horizontally continuous layers should eventually approach a steady-state, and the potential importance of a three-dimensional flow regime within the Morrison is suggested.

There are two data points that stand out on Figure 11 as having extreme $^{36}Cl/Cl$ ratios, samples 31 and 34B. Sample 31 has the highest measured $^{36}Cl/Cl$ ratio at 2,201 x 10-15, and sample 34B has a similarly high ratio of 1,857 x 10-15. The unusually high ratios of these samples suggest ^{36}Cl buildup or possible contamination of the samples with much younger, bomb-pulse influenced, water. These samples were measured in the ^{36}Cl run at Rochester when the chemistry blanks showed evidence of minor lab contamination (see results section), and an earlier sample from well 34 measured in another run, sample 34A, has a more expected ratio of 481. x 10-15. Despite this, there is no clear evidence to support the idea that the high ratios are related to the much lower ratio lab contamination event.

The preferred explanation for the high ratios is uranium-thorium related ^{36}Cl buildup. As Figure 10 has suggested, there are uranium-thorium occurrences other than those located on Figure 6. In fact, McLemore and Chenoweth (1989) reported that almost 500,000 pounds of U_3O_8 have been produced from areas along the western border of San Juan County. The first recorded discoveries of uranium in New Mexico stemmed from the mining of vanadium-bearing carnotite ore in the Carrizo Mountains west of Shiprock (Perkins, 1979). Following World War II uranium deposits were discovered in the Sanostee area south of the Carrizo Mountains and in the Cuba-San Yisdro area on the eastern side of the basin. The major deposits of the Grants Mineral Belt were discovered in the 1950's, and drilling down-dip from these first discoveries is continually encountering increased uranium in more areas of mineralization (Perkins, 1979).

The secular equilibrium runs made with the CLSEC program indicated the sensitivity to both the uranium/thorium content and to a lesser degree to the general mineral assemblage. The runs primarily explored the best choice for a generally applicable value. If values were calculated for more specific locations, their variability would be great. The occurrence of heavy mineral deposits, particularly those containing minerals such as radioactive zircon (Bingler, 1963; Chenoweth, 1957), may also be important to local ^{36}Cl buildup.

Wells 31 and 30 are in the same geographic group, group 3, and there are other samples from wells in group 3 that fall near the decay line drawn through sample 30 on Figure 11; however, sample 31 plots significantly above this decay line. This displacement could possibly be explained by the difference in the chloride content. Sample 30 has a chloride concentration of 2.7 mg/L whereas sample 31 has a chloride concentration of 1.6 mg/L. If sample 31 is associated with the buildup of ^{36}Cl in a recharge water, then the decay/buildup line would be expected to be at a steeper slope than the line through sample 30 because of the lower chloride concentration.

Similarly, sample 34A plots just below the decay line drawn from the origin through sample 34B. Again, the displacement could be explained by the difference in the chloride concentrations. Sample 34A has a chloride concentration of 7.8 mg/L, whereas the chloride concentration of 34B is 7.1 mg/L. In addition, the relative displacements of samples 36A and 35 from the line drawn through 36B can be explained by chloride concentration differences. The chloride concentrations of 36B is 17, whereas 36A, which plots above the line, is less at 14, and 35, which plots below the line, is greater at 19.

The isotopic ratio versus the chloride concentration

Decay and buildup processes are clearly important and can not be ignored, but a further look at mixing is needed. The discussion can be built on the mass balance considerations noted in the approach section. If the volume at the sample point is a mixture of two volumes from two distinct sources, or:

18) $$V = V' + V''$$

then the ^{36}Cl atomic mass balance can be written:

19) $$R \cdot C \cdot V = R' \cdot C' \cdot V' + R'' \cdot C'' \cdot V''$$

where, as previously introduced, R is the $^{36}Cl/Cl$ ratio, C is the chloride concentration, and V is the elemental volume of the source contributions. Like as done for equation 2, a constant that relates the ^{36}Cl concentration to the isotopic ratio, R, and the chloride concentration, C, has been divided out of the mass balance of equation 19. If C is measured in mg/L, the atomic concentration of ^{36}Cl in atoms/L is described by equation 1b.

Re-introducing the proportionality constant f for V'/V, mixing equation 19 becomes:

20) $$R \cdot C = R' \cdot C' \cdot f + R'' \cdot C''(1-f).$$

The proportionality constant, f, can be solved for using the chloride mass balance, as previously introduced in equation 12. The ratio and chloride values possible for the endmembers is non-unique, but if mixing of two endmembers is to be represented by a linear relationship, the relation of R to C needs to be linearized. A linear relationship can be described by substituting equation 12 into 20. The result, after some algebra, is:

$$21)\qquad R = \frac{R'\bullet C' - R''\bullet C''}{C' - C''} + \frac{1}{C}\left[\frac{R''\bullet C''\bullet C' - R'\bullet C'\bullet C''}{C' - C''}\right]$$

This equation can be rewritten to emphasize the concentration behavior of the ^{36}Cl tracer during mixing. The concentration of ^{36}Cl in atoms/L is a function of the product of the isotopic ratio and the chloride concentration as presented in equation 1b. Considering this, the conservation of mass balance presented in equation 21 can be rewritten for an atom/L mass balance:

$$22)\qquad k\bullet R = \frac{^{36}Cl' - {}^{36}Cl''}{C' - C''} + \frac{1}{C}\left[\frac{C'\bullet{}^{36}Cl'' - C''\bullet{}^{36}Cl'}{C' - C''}\right]$$

where k and the units are as described for equation 1b. This equation is essentially the same as that used by Gifford et al. (1985) in their report on ^{36}Cl mixing in Hanford, Washington groundwaters.

The addition of chloride from dissolution or introduction of highly concentrated pore fluid is not regarded as a volumetric mixing, but can be still regarded as the mixing in of added solute. Assuming that the change in concentration is due to the added chloride, and ignoring any volume change, the atomic mass balance can be written:

22) $$R \cdot C = R' \cdot C' + R_i(C - C')$$

which can be linearized with respect to R and C as:

23) $$R = R_i + \frac{1}{C}(R' \cdot C' - R_i \cdot C')$$

where R_i is the $^{36}Cl/Cl$ ratio of the added chloride (perhaps R_{se}). Again, using equation 1b to substitute in a atoms/L ^{36}Cl concentration, equation 23 becomes:

24) $$R = R_i + \frac{1}{k \cdot C}(^{36}Cl' - k \cdot R_i \cdot C')$$

Gifford et al. (1985) present a similar equation, with R_i equal to R_{se}.

Equations 21 and 23 suggest that on a plot of $^{36}Cl/Cl$ versus $1/Cl^-$, the mixing scenarios described by the mass balance equations 22 and 24, would both plot linearly. In addition, the equations suggest mixing of added solute can be conceptually regarded as mixing with water of a very large concentration. In general, the isotope ratio of a mixture will be more heavily influenced by the more concentrated water.

Figure 13 provides an analysis of different processes on a $^{36}Cl/Cl$ versus 1/Cl plot of the Morrison data. The three decay lines from Figures 11 and 12 are again plotted, this time with a smooth interpretation of the effects of the addition of dead chloride. Figure 13 indicates the displacements on this type of plot caused by all the processes considered. The mixing displacements indicated apply to any displacement into the quadrant that they plot in. The lines were drawn in directions suggested by the current data. The dense portion of the figure is enlarged in Figures 14 and 15.

Two mixing displacements, labeled A and B are indicated on Figure 13. These displacements address the tremendous variations in the group 3 samples. The presence of a Morrison Formation ion filtration phenomenon is proposed at least in the vicinity of wells 27, 28, 29, and 32, and perhaps is effecting data from wells 11, 24, and 31. The two lines (A and B) reflect the compositional influence of waters from both sides of the geologic membrane in different wells in the area. The wells listed include virtually all of the samples in group 3; however, the decay line developed in the same vicinity is indicative of the multiple influences on the solute composition.

Mixing displacement line A indicates mixing of waters on the sample 30 decay line with very dilute, radiometrically older water to explain the extremely dilute water samples, 27 and 32. The dilute nature of these samples could be explained by waters

Figure 13. $^{36}Cl/Cl$ vs. 1/Cl for Morrison Formation waters. Figures 14 and 15 provide additional detail.

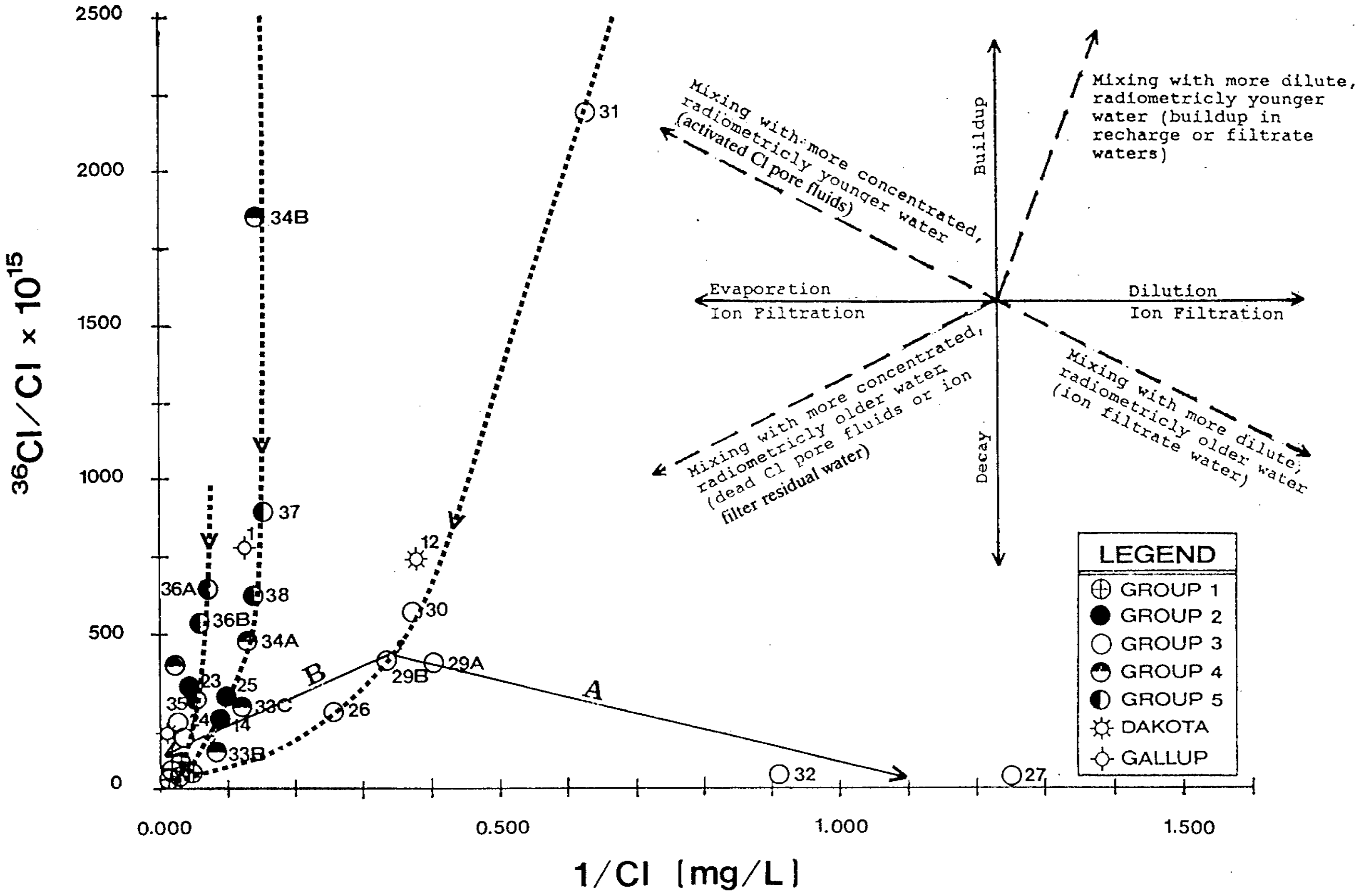


Figure 14. ^{36}Cl/Cl vs. 1/Cl for Morrison Formation waters. Expanded scale shows portion of figure 13 in more detail. Figure 15 provides further detail.

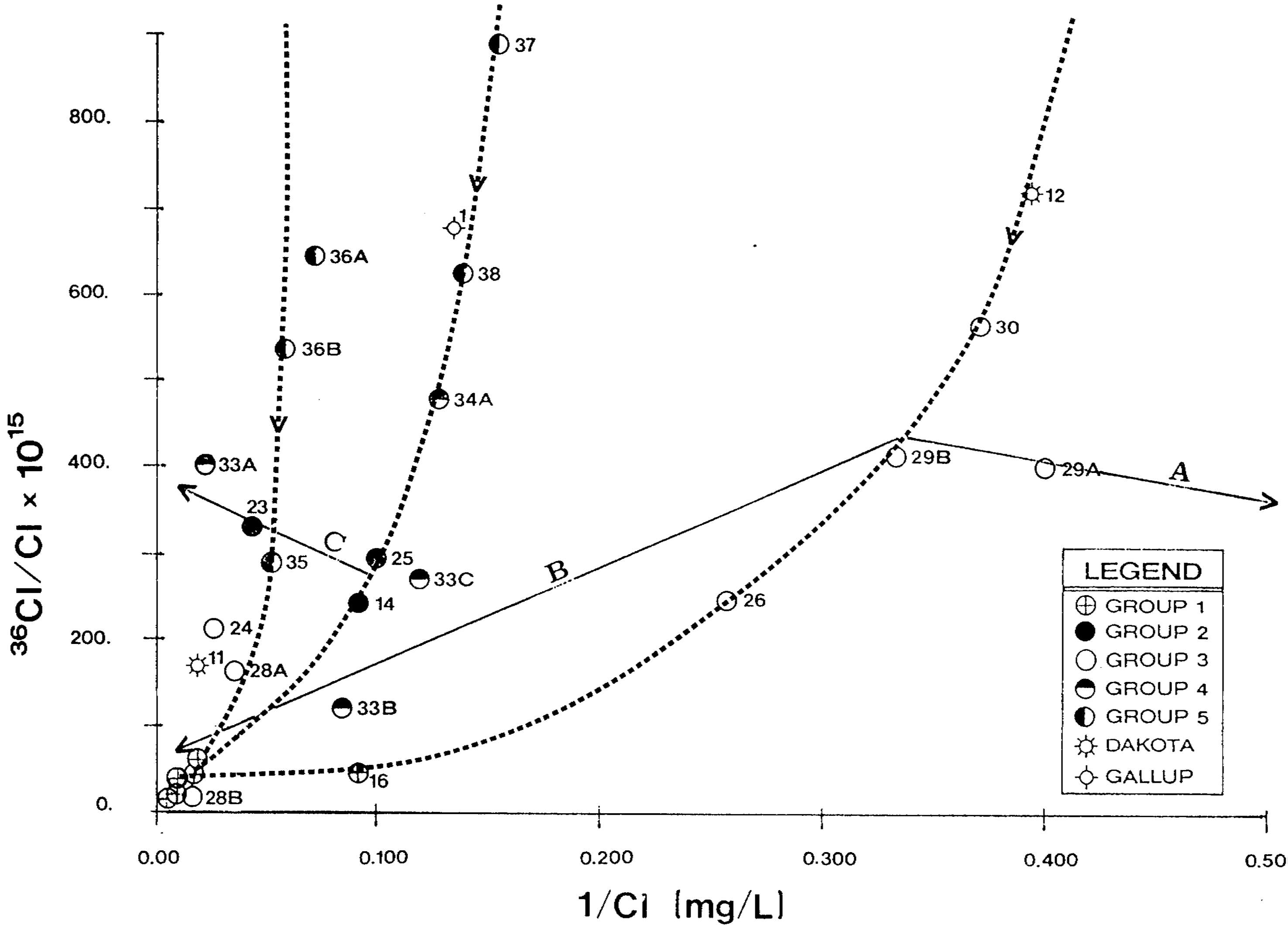
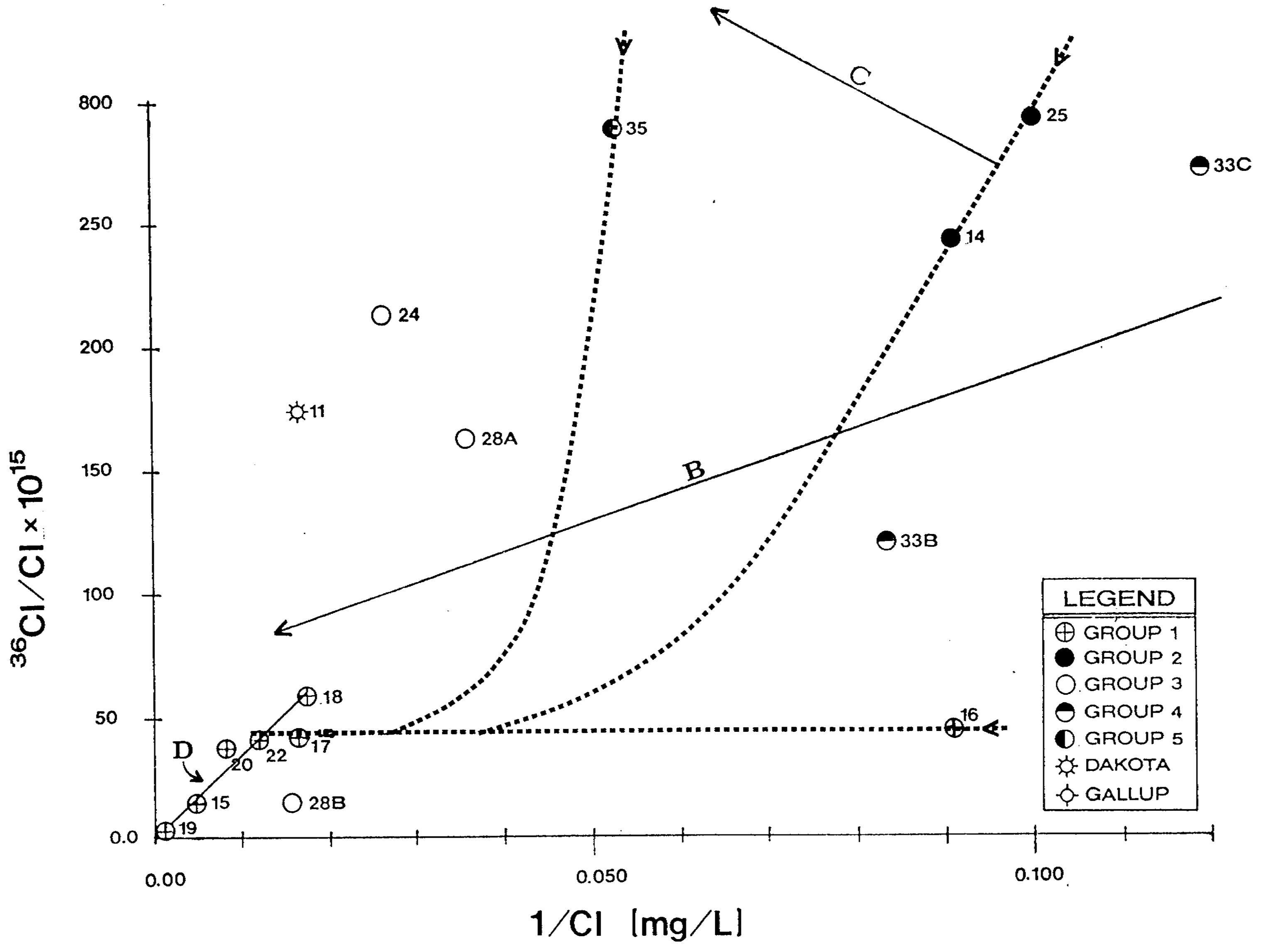


Figure 15. $^{36}Cl/Cl$ vs. 1/Cl for Morrison Formation waters. Expanded scale shows portion of figure 14 in more detail.



initially introduced, perhaps during a more recharge conducive period in the past, as very dilute recharge water. However, it seems doubtful that a dilute recharge water could remain dilute throughout the time needed to decay the ^{36}Cl to the extremely low ratios measured for these samples. Therefore, since there is other evidence (line B) for ion filtration effects among these samples, mixing with an ion filtrate water is proposed. Since the mixture would be dominated by the more concentrated endmember, the very dilute waters of samples 27 and 32 must indicate a nearly pure endmember, or mixing with an endmember with an even lower concentration. The placement of the line is also directly supported by the changes with time observed in the nearby, well 29 sample data. Sample 29A is shifted in the direction of line A away from sample 29B suggesting a linear mixing with older, dilute waters occurs in the well.

The linear mixing displacement B is best observed on Figure 14. The occurrence of mixing with ion filter residual waters is most strongly suggested by the data from well 28, and the line drawn to represent line B is an average of the plotted data for samples 28A and 28B. The plotted location of sample 28A, along with the locations of samples 24 and 11, could also be readily explained by mixing with waters associated with decay of ^{36}Cl in waters from a higher salinity source. The plotted location of 28B suggests that some of the mixed in water is very old, perhaps associated with flow from the direction of well 35 in the more central region of the basin (group 5). As Figure 15 further suggests, the endmember for this mixing is unique, and in fact extreme (assuming moderate secular equilibrium), in its low isotopic ratio. Well 28 may be on the eastern edge of the samples in group 3 (towards the central basin), but the suspected flow directions (Figure 4) do not strongly suggest a large contribution from such a direction.

Considered together with line A, line B suggests that a number of wells in group 3 penetrate both sides of an old, ion filtration system, with the residual side of the membrane system associated with the overlying Dakota unit. Since wells 28 and 24 are cased more than 100 feet beyond the Brushy Basin Member (see Table 2), their correlation with Dakota sample 11 suggests the Recapture Member as the geologic membrane unit.

A linear mixing line C is also introduced on Figure 14. Water flow was suspected northward along the western Morrison outcrop area (Figure 4), and whereas samples 25 and 14 appear to fall on the decay line associated with most of the outcrop samples taken, the sample from well 23, which is physically located between the other two wells, plots in a shifted manner as suggested by line C. A mixing is proposed between the waters on the decay line of samples 25 and 14 and a more concentrated, radiometrically younger water. The endmember water of this mixing is probably more saline pore waters being released from areas of high subsurface ^{36}Cl production. As suggested by Figure 10, there are certainly areas of high uranium and thorium concentration, and the waters of samples 23 and possibly 33A are well explained by mixing with nearby waters. Note that this shift places the plotted points for samples 23 and 33A near the decay line drawn for the decay of the central basin waters (group 5), a group whose initial point is considered to be the result of release of high secular equilibrium chloride.

The enlarged scale of Figure 15 allows a closer examination of the group 1 wells, located near the river. At this scale, the decay line from sample 30 toward the origin becomes almost flat. The chloride being added becomes a more important process than the degree of decay. The chloride being added is mixing with the chloride decayed as described by the group 3 decay line. Samples 16 and 17 represent this mixing with a water similar to sample 22. Sample 22 and the majority of the group 1 wells plot on linear mixing line, D, that can be drawn from the points plotted for samples 18 and 19. The relative position of the samples along line D are related simply to a mixture of discharging waters from different units, not to the areal location of the sampling points.

CONCLUSION

The Morrison Formation, as described by the well data available for this study, is an aquifer system with complex mixing of different waters. Well waters have chloride compositions characteristic of numerous processes. Members, and layers within members, must contribute a very different array of solute compositions to explain the variability in the data. The large open well intervals serve as mixed collection points. The relative contribution of individual units to the water composition measured appears to be inconsistent from well to well. Such variability implies a lack of horizontal connection, or the overwhelming influence of the draining of local reservoirs.

The ^{36}Cl data is not well described by the simple dating equations commonly used. Flow-paths are not well known, and multiple processes are certain. The use of calculated ages in such a system is uncertain. Data interpretation is greatly assisted by a diagrammatic understanding of the relation of important chloride data.

Three different decay histories are suggested by the measured data. The decay line drawn on figure 11 through sample 30 represents the decay line most representative of expected flow path behavior. Sample 30 has isotopic ratio and chloride concentration values expected for a slightly decayed recharge water. The flow-path suggested by the wells that fall along this decay line fits the suspected hydrologic conditions, but the deviations present in wells in their midst (group 3) suggest that oversimplifications may be premature.

A different decay line is suggested by the samples from wells in, or immediately adjacent to, outcrop areas (group 2 and samples from groups 4 and 5). The line is drawn on figure 11 through samples 38 and 34B. The decreased slope of the line reflects the greater chloride concentrations of these samples than those of the group 3 decay line. A different source (e.g., recharge area inter-aquifer flow) or a change in the recharge composition is suggested.

The influence of subsurface production of ^{36}Cl is clearly reflected in modestly saline samples from the central basin. The continued subsurface production influence on waters in the southerly portion of the basin is suggested by the high isotopic ratio observed in well 37 and the Chaco Canyon well (well 36), although at Chaco local increased production is also supported. As suggested by the third decay line drawn on figure 11, the chloride composition at well 35 is well explained by the presence of decayed chloride originating from waters like those of well 36.

Samples taken from Morrison wells near the San Juan River, clearly show the compositional influence of a dead chloride solution. Discharge of old, concentrated waters near a basin discharge area is expected and clearly indicated on figure 11.

The decay histories developed on figures 11 and 12 can be examined on the mixing orientated diagrams of figures 13, 14, and 15. Decay processes along potential flow-paths can seemly be well described by smooth functions that incorporate chloride concentration increases. In the Morrison system, concentration increases due to dissolution or leaky additions lead smoothly into mixing with a higher chloride discharge water.

A number of the mixing processes described on figure 13 are suggested by the chloride data measured. The data strongly suggest the sampling of a large ion filtration system in the area of wells 11, 24, 27, 28, 29, and 32 (see figure 9 for locations). Both sides of the geologic membrane appear to have been sampled, as represented by the mixing lines A and B on figures 13 and 14. Data suggests the Recapture (not Brushy Basin) Member of the

Morrison is serving as the membrane unit. The stratigraphically higher Dakota, and Westwater Canyon waters appear to be on the chloride concentrated side of the membrane, and the lower units, perhaps the Cow Springs or Bluff Sandstone units of the Wanahka Group, appear to be on the dilute side of the system. The very old, very dilute waters from wells 27 and 32 are best explained as ion filtrate. Current flow directions are uncertain, but flow across the membrane at one time or another produced the dilute filtrate type water observed.

The importance of subsurface ^{36}Cl buildup in the northwestern part of the basin is suggested in the extreme isotopic ratio measurements of samples from wells 31 and 34. In addition, buildup of ^{36}Cl in more concentrated, perhaps pore-fluid type, waters is suggested by samples 23 and 33A. These wells are close to known radioactive deposits as described on figure 10. The addition of neutron activated, "pore-fluid" chloride is the processes suggested with mixing line C on figure 14. The line reflects the mixing of waters of the type typically observed in the outcrop, with neutron activated, more concentrated pore fluids.

Samples from wells near the San Juan River are compositionally dominated by the addition of a high chloride discharge water. This discharge would presumably smoothly mix with the chloride in the Morrison water as it proceeds along the flow-path decay line, as shown interpretatively on figure 15. The mixing of water from the high chloride source and the sample 30 decay line is implied for samples 16 and 17. The high chloride samples appear to be a group of mixtures themselves, as described by line D on figure 15. The group represents a variety of old waters contributing to the discharge area solute compositions, with the end-points of the linear mix represented by samples 18, and 19. Perhaps the mixing line represents the compositional variation in a contributing aquifer system, such as the Todilto Limestone or Entrada Sandstone in the lower Wanahka Group.

Comparisons of the chloride data to other data being compiled by the USGS RASA for the same wells are still preliminary. Data (Dam, 1988) show that the higher chloride waters from wells near the river are actually of a concentrated sodium sulfate type, suggestive of the influence of the gypsiferous Todilto. The few wells that have significant ^{14}C show decay-based ages in poor agreement with the ^{36}Cl data. Groundwater ages suggested by the two methods differ by two orders of magnitude, and the youngest ages calculated for the ^{14}C method (Dam, 1989, personal communication) are shown by the chloride data to be very old dilute waters (wells 27 and 32). Mixing scenarios can address the differences, as chloride and carbonate can exist in entirely different proportions in the waters being admixed. Small amounts of a younger carbonate water will have a profound effect on the composite measurement when mixed with waters dead of ^{14}C, an isotope with a much shorter half-life than ^{36}Cl (about two orders of magnitude).

At initial inspection, the stable isotope data (Dam, 1988) demonstrate some interesting coincidences. One of the isotopically lightest waters is observed in well 27, the water proposed as an ion filtrate. Other samples from the same geographic group also group together near this sample on the standard meteoric water line plot. No large deviations from the meteoric water line are observed in the data, and further analysis is required before a definitive conclusion can be drawn as to any comparison with the ^{36}Cl data.

The CLSEC program as introduced is capable of nearly replicating the neutron flux and secular equilibrium calculations presented by Fabryka-Martin (1988). The program is sensitive to poorly known rock composition elements (such as U and Th) and the calculations vary somewhat depending on how the water rock mixture is handled. Early comparisons of measured and calculated neutron flux are potentially encouraging (Andrews et al., 1986), but the importance of locations within the rock matrix of specific minor elements could be critical to the buildup of ^{36}Cl. Comparisons to other radioactive measurements made for samples taken at the well are incomplete.

Complications of mixing have suggested the dangers of trying to follow decay of recharge chloride along flow-paths. Yet, the analysis of the data does suggest decay histories that would presumably be associated with flow path type evolution. The decay line suggested on figure 13 for wells 30, 29, and 26 is associated with a chloride concentration change of 2.7 to 3.9 mg/L. These are small chloride changes, and the ages calculated for simple decay should be nearly unchanged by their consideration. By limiting the view on figure 7 to those wells that might fit on such a decay line, the calculated ages may have some meaning. The simple decay calculated ages for the three well waters just listed are 90,000; 240,000; and 470,000 years, A fairly smooth progression whose increase implies residence times that are in a range that might be expected for this system. Relating ages to rates associated with processes has further possibilities, but a more complete development of associated hydrochemistry and hydrology data is first recommended.

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APPENDIX A

NEW MEXICO TECH ISOTOPE HYDROLOGY LAB PROCEDURES USED

Appendix A.1. Procedure for Chlorine-36 sampling using anion-exchange columns.

Modified from 6/84 laboratory notes of June Fabryka-Martin.

Materials:

20-50 mesh Dowex™ 1-X8, Cl^- form, anion exchange resin (as sold by BioRad).
Exchange columns (eg. PVC columns, 3 cm in diameter, 25 cm long, with screw-on endcaps and metal nipples that can be capped or valve closed)
Reagent grade chemicals (check for minimal Cl^- content)--
$NaNO_3$ or HNO_3, NaAc, $AgNO_3$

Resin preparation:

1) Resin initially in Cl^- form. Elute with 2M $NaNO_3$ (AR grade) or 2M HNO_3 until no Cl^- is detected in the eluant when tested with a solution of $AgNO_3$. The flowrate should be about 0.4 ml/min/cm^2 bed per the recommendation of the BioRad Catalog (p. 12). Takes about 5 or more bed volumes of NO_3^- solution.

2) Rinse with distilled deionized water until the pH returns to normal (pH 5-6).

3) Elute resin with 2M NaAc. Check for Cl^- in eluant by $AgNO_3$ test. Column should be eluted to reduce residual NO_3^- and make the later Cl^- capture more efficient (eluant can by checked for NO_3^- using, for example, a Hach kit-Cd reduction method).

4) Rinse with distilled H_2O. Pack columns or store resin in bottles for later packing.

Column Preparation:

1) With one end capped and plugged, fill PVC (or other) column partially with distilled water. Add saturated glass wool plug, tamp into place. Minimize presence of entrapped air as much as possible (changes effective flowrate in field).

2) Slurry into column at least 30 cm^3 of exchange resin. At 1.4 meq/cm^3, this amount should be adequate to collect easily the desired quantity of Cl^-.

3) Add saturated glass wool plug to top of resin, fill column with distilled H_2O and screw on top endcap.

Field operation:

1) Collect sufficient sample to contain at least 200 mg Cl^-. Sampling can be performed from either a water sample previously collected and stored, or from a water obtained by connection to the well. The size of stored water sample required is dependent on the chloride concentration of the sample. Two liters of water typically can be used for direct precipitation of AgCl for waters with chloride concentrations down to just less than 20 mg/L. The use of anion exchange collection is especially helpful for waters with less than 5 mg/L chloride. Larger precipitate quantities are less prone to contamination.

2) Connect one column end to saturated tubing leading from the sample supply (well water).

3) Set up column so that it flows up through column to maximize exchange efficiency. Flow rate should be on order of 3 ml/min/cm² bed cross-section. for a column of 3 cm diameter, this rate is about 1.2 L/hr. (Note that extreme efficiency and exact rates are not much of a concern if the column can just be connected to a near-continous supply for a reasonable time). The well pressure should not be directly transferred to the column, some form of regulation that does not greatly affect well sampling conditions is required. In the case where sample supply is limited, the efficiency of Cl^- capture can be judged using $AgNO_3$ to check eluant.

4) The flow may slow considerably if the sample degases in the tubing or in the column. If this is of concern, the air bubbles can be let out and the flow restarted.

Sample recovery:

1) Slurry resin from column into buret containing small glass wool plug on bottom (or setup column directly, with a inlet reservoir).

2) Elute resin with 2M $NaNO_3$ until Cl^- content of eluant is negligible (or until its clear plenty of sample is available, fractionation is not expected). Takes about four bed volumes. Resin volume will decrease by about 20%.

3) Add sufficient $AgNO_3$ solution to eluant to precipitate at least 200 mg AgCl. Let sit overnight.

4) Filter out precipitate and purify by normal procedure.

Appendix A.2. Lab procedure for purification of silver chloride precipitate.

Modified from 6/85 laboratory notes of Nancy O. Jannik

Equipment Needed:

- fume hood
- low temperature oven
- hot plate - optional
- vacuum pump or line
- 1000ml Erlenmeyer filtering flask
- glass test tubes, 25x200mm
- beakers, 200ml and 400ml
- watch glasses
- stirring rods
- 300ml millipore filter funnels
- filter paper, 0.45 micron (to fit filter funnel, e.g. 47 mm)
- laboratory squeeze bottles containing: a) distilled deionized water, b) dilute HNO_3, c) dilute NH_4OH, and d) reagent grade NH_4OH
- small, preferably amber sample vials to contain final precipitate
- parafilm
- disposable polyethylene gloves
- plastic forceps
- distilled water
- distilled-deionized water (type I: 18MΩ water)
- reagent grade chemicals:
    - barium nitrate
    - ammonium hydroxide
    - nitric acid
    - silver nitrate
- aluminum foil
- NaCl sample to use as background blank or carrier

Procedure:

Purification of chlorine-36 samples prior to analysis in the tandem-accelerator mass spectrometer is necessary to reduce the sulfur content of the samples. Sulfur-36 ions follow along a similar path as chlorine-36 ions in the accelerator, thus hindering accurate chlorine-36 analysis. The following procedure is intended for groundwater or other samples where small sample quantities are not the major concern. Small samples can be bolstered by addition of known amounts of dead chloride (carrier), but frequently centrifuge techniques are employed to reduce filtering steps (e.g. Conard et al., 1986, The chemical preparation of AgCl for measuring Cl-36 in polar ice with accelerator mass spectrometry, Radiocarbon, V. 28, p556-560.)

Care must be taken during the purification process to avoid contamination. Samples should be covered, at least with watch glasses, whenever possible, even when in filter funnels. The entire process is conducted on a laboratory fume hood. The AgCl precipitate is photo-sensitive and containers should be covered with foil hoods or similar shading devices. Disposable poly gloves should be worn during the entire process and changed often. All equipment should be washed and treated before any use or reuse. Laboratory squeeze bottles of distilled deionized (DD) water, dilute HNO_3, and dilute NH_4OH are useful for chemical treating of equipment. Glass and plasticware should first be washed with laboratory soap and water, and rinsed with distilled water. Steam cleaning is desirable. Next, it should be rinsed with dilute HNO_3 followed by DD water, then rinsed with NH_4OH, followed by several rinses with DD water.

A different subsample should be maintained for chloride concentration determination. If the volume of the subsample used for the Cl-36 analysis is noted, the recovery of the lab procedure can be calculated, based on the initial concentration and the resultant precipitate weight.

Two different options are presented for precipitating AgCl from a NH_4OH solution. One evaporates the NH_4OH using a hot plate, and the other neutralizes the NH_4OH by HNO_3 addition. The hot plate method uses less reagents and results in stronger crystal formation. The acid addition method is easier and requires less equipment.

1) Add reagent grade 0.1 N $AgNO_3$ to water sample in an amount sufficient to precipitate at least 200 mg AgCl (usually 50 ml or so). Let stand for 24 hours in the dark.

2) Unless sample quantities are limited, decant and discard the supernatant. Filter the AgCl precipitate to near dryness in a filter funnel, with 0.45 micron filter paper, using a vacuum pump or vacuum line. Wash the precipitate thoroughly, in the filter funnel, with DD water, and discard solution.

3) Transfer the filter funnel to and armed flask with a 25x200mm test tube inside (lower and raise test tube into and out of the flask with treated plastic forceps). Dissolve the precipitate by adding 25-50ml reagent grade NH_4OH to the filter funnel. Allow sufficient time for the precipitate to dissolve and gravity filter. Only if necessary, gently draw the solution into the test tube with the vacuum. Use a squeeze bottle of reagent grade NH_4OH to rinse and dissolve any precipitate that may stick to the sides of the funnel. After noting the material captured, remove filter funnel and discard used filter with any remaining precipitate.

4) Transfer solution from test tube to a treated 200ml beaker. Carefully add 1 ml $Ba(NO_3)_2$ to solution in beaker, as sputtering may occur. Cover beaker with parafilm and allow to sit overnight. [To make the $Ba(NO_3)_2$ solution, place a good amount of solid $Ba(CO_3)_2$ in a flask. Add sufficient HNO_3 to dissolve some of the $Ba(CO_3)_2$, but leave some in solid form in the bottom of the flask (assures saturation). When using the $Ba(NO_3)_2$ solution, draw off the liquid from the top.]

5) Filter solution into a test tube and transfer solution to a treated 400ml beaker.

6) Option 1--Lay a glass stirring rod across the top of the beaker and cover with a chemical watch glass (concave side up). Evaporate the NH_4OH and reprecipitate the AgCl by heating the beaker at 50°C-65°C for 1-½ to 3 hours. Add small amounts of DD water (from squeeze bottle) during the heating process to buoy up the precipitate and prevent it from sticking to the bottom of the beaker.

6) Option 2--Precipitate the AgCl by neutralizing the NH_4OH with small, careful additions of HNO_3 to the solution. Care should be taken <u>not</u> to add the acid in a sudden manner to the basic solution.

7) Using DD water as needed, rinse the precipitate from the beaker into the filter apparatus. Wash the precipitate thoroughly with DD water and filter it to near dryness.

8) Transfer filter funnel to an armed flask with a test tube setup. Redissolve the AgCl precipitate by adding 25-50ml reagent grade NH_4OH to the filter funnel. Again allow sufficient time for the precipitate to dissolve and gravity filter. Only if necessary, draw the solution into the test tube with the vacuum pump. Use a squeeze bottle of reagent grade NH_4OH to rinse and dissolve any precipitate that may stick to the sides of the funnel. After noting material collected, remove the filter funnel and discard the used filter paper.

9) Transfer solution to a 400ml beaker and repeat steps 6 and 7. If sulfur contamination is suspected (eg. if solution has color, or there was heavy filter residue removed, or known problem), repeat steps 8 and 9. During final filtering process, try to gather precipitate from filter funnel sides onto micropore filter using DD water.

10) Crumple and then flatten a blue filter-cover paper (found between individual 0.45 micron filters), and lay it on a treated watch glass (concave up). Using treated forceps, place the filter paper with the AgCl precipitate on top of the blue filter-cover paper. Place the watch glass in an oven allowing the precipitate to dry overnight at 45°C (if time is critical, a drying time of 2 hours at 65°C should be sufficient).

11) Weigh a treated and dried sample vial. Transfer the dry powder to the vial, and reweigh to obtain sample weight. Vial should be well sealed (perhaps with parafilm), labeled, and wrapped with aluminum foil (and relabeled) for storage.

Sample is ready for loading into sample holders used for AMS analysis.

APPENDIX B: CLSEC COMPUTER FILES

Appendix B.1. Listing of FORTRAN code for the program CLSEC.

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C
C   PROGRAM CLSEC (VER 1.3) BY B.G. JONES
C
C   DESCRIPTION-- CODE TAKES CHEMISTRY AND DENSITY INFO FOR A MEDIA
C      AND DETERMINES THE NEUTRON FLUX AND NEUTRON ABSORPTION PER
C      KG OF MEDIA.  THESE VALUES ARE USED TO CALCULATE A SECULAR
C      EQUILIBRIUM RATIO FOR 36CL/CL IN THE PORE WATER OR ROCK.
C
C   INPUT-- CONTROL AND SAMPLE DATA IS READ FROM AN INPUT
C      FILE AS DESCRIBED FOLLOWING.  RIGHT JUSTIFY INPUT IN COLUMNS.
C
C   1) CONTROL CARD (4I5)
C
C     COL.  1-5   NSAM:    NUMBER OF SAMPLES TO BE READ FROM INPUT FILE
C           6-10  ICONV:   PARAMETER INDICATING IF CHEM ARRAY OXIDE DATA
C                          IS TO BE OPTIONALLY READ AS ELEMENTAL %;
C                          = 1 IF YES
C                          = 0 IF NO
C          11-15  IPRN:    PARAMETER INDICATING IF SAMPLE DATA IS TO
C                          BE ECHOED IN OUTPUT FILE FOR ERROR CHECKING;
C                          = 1 IF YES
C                          = 0 IF NO
C          16-20  ISTAT:   PARAMETER INDICATING IF SIMPLE STATISTICS ARE
C                          TO BE CALCULATED FOR CALCULATED RATIOS.
C                          =1 IF YES
C                          -0 IF NO
C
C   SAMPLE CARDS, REPEAT FOR EACH SAMPLE
C   2) SAMPLE NAME CARD (A80)
C
C      COL. 1-60  STITLE:  SAMPLE TITLE TO BE ECHOED ON OUTPUT
C
C   3) POROUS MEDIA PARAMETERS (4F8.2)
C
C     COL.  1-8   POR:     MEDIA POROSITY. SET = 0 TO CONSIDER ONLY ROCK
C           9-16  SPWTR:   SPECIFIC WEIGHT OF ROCK (G/CM3)
C          17-24  SAT:     WATER SATURATION INDEX (0 TO 1)
C          25-32  CCLW:    CHLORIDE CONCENTRATION IN PORE WATER (MG/L)
C
C   4-6) ROCK CHEMISTRY CARDS (3 LINES, 9F8.2)
C
C     COL.  1-8   CHEM(J): ROCK CHEMISTRY DATA FOR COMPONENT J, WHERE J
C           9-16  J=1, 27  IS, IN ORDER, GROUPED BY LINE;
C                          1) SIO2,AL2O3,FE2O3,FEO,MGO,CAO,NA2O,K2O,TIO2
C                          2) MNO,P2O5,H2O,CO2,LI,RB,SR,MN,U
C                          3) TH,CL,SM,GD,B,F,BE,N,S; RESPECTIVELY.  THE
C                          OXIDES ARE GIVEN IN WT% AND THE ELEMENTS ARE
C                          IN PPM.  H2O REFERS TO ROCK OXIDE WT%.  IF A
C                          VALUE FOR MNO IS GIVEN, THAT FOR MN IS
C                          IGNORED.  OXIDES AS ELEMENTAL % IF ICONV=1.
```

```
      IMPLICIT REAL*8 (A-H,O-Z)
      IMPLICIT INTEGER*4 (I-N)
      REAL NWCL,NWH2O,MEAN,MOLE(27),MW(28)
      DIMENSION CHEM(28),CONV(13),XSEC(28),CM(28),RSE1(10),RSE2(10)
      CHARACTER*80 STITLE
      CHARACTER*12 FNAMI,FNAMO
      CHARACTER*1 AIND1,AIND2,AIND3
      DATA AIND1/'N'/,AIND2/'N'/,AIND3/'N'/
C ESTABLISH INDEXING OF CHEMISTRY ARRAYS (FOR EQUATION READABILITY)
      DATA ISI,IAL,IFE2,IFE,IMG,ICA,INA,IK,ITI/1,2,3,4,5,6,7,8,9/
      DATA IMNO,IP,IH2O,ICO2,ILI,IRB,ISR,IMN/10,11,12,13,14,15,16,17/
      DATA IU,ITH,ICL,ISM,IGD,IB,IFL,IBE/18,19,20,21,22,23,24,25/
      DATA INIT,ISUL,IOXY/26,27,28/
C MOLE ARRAY IS CONSTENT, CONVERTS WT% AND PPM TO MOLES/100G ROCK
      DATA MOLE/60.09,50.98,79.85,71.85,40.31,56.08,30.99,47.1,79.9,
     #70.94,70.97,9.01,44.01,6.94E4,85.47E4,87.62E4,54.94E4,237.98E4,
     #232.0E4,35.45E4,150.35E4,157.25E4,10.81E4,18.998E4,9.01E4,
     #14.01E4,32.06E4/
C MW ARRAY IS THE MOLECULAR WEIGHT FOR 28 ELEMENTS
      DATA MW/28.086,26.98,55.847,55.847,24.312,40.08,22.9898,39.102,
     #47.9,54.938,30.9738,1.00797,12.01115,6.94,85.47,87.62,54.94,
     #237.98,232.0,35.45,150.35,157.25,10.81,18.998,9.01,14.01,32.06,
     #15.9994/
C CONV ARRAY CONVERTS THE DATA GIVEN OPTIONALLY AS ELEMENTAL % TO
C  WT% OXIDE.
      DATA CONV/2.1393,1.8895,1.382,1.2865,1.6581,1.3992,1.3480,
     #1.2046,1.6680,1.2912,2.2914,8.9364,3.6641/
C XSEC ARRAY IS THE THERMAL CROSS-SECTION FOR EACH OF THE 27
C    INPUT ELEMENTS + OXYGEN (FROM MUGHABGHAB, DIVADEEN, & HOLDEN,
C    V1 1981, V2 1984, BROOKHAVEN LABS DATA PUB. BY ACADEMIC PRESS;
C    VALUES FOR SM AND GD ARE ADJUSTED FOR WESTCOTT FACTORS 1.7 AND
C    0.85 FROM FABRYKA-MARTIN, 1988, TABLE D-4)
      DATA XSEC/0.171,0.231,2.56,2.56,0.063,0.43,0.53,2.15,6.09,
     #13.3,0.172,0.3326,0.0035,70.5,0.38,1.28,13.3,7.6,
     #7.4,33.5,9639.,41556.,767.,0.0096,0.0092,1.9,0.52,0.00019/
C AVOGADRO'S NUMBER
      AV=6.02252E23
C SPECIFIC WEIGHT OF PORE WATER (G/CM3)
      SPWTW=1.0
C SPECIFIC WEIGHT OF AIR (G/CM3, USED FOR UNSATURATED POROSITY)
C     SPWTA=.001--  NOT CURRENTLY USED
C CONVERSION FROM BARNES TO CM2
      BARCM2=1.E-24
C
   99 WRITE(*,210)
  210 FORMAT(/5X,'Enter input filename: ',$)
      READ(*,220) FNAMI
  220 FORMAT(A12)
      WRITE(*,230)
  230 FORMAT(/5X,'Enter output filename: ',$)
      READ(*,220) FNAMO
      OPEN(UNIT=8,FILE=FNAMI,STATUS='OLD',ERR=99)
      OPEN(UNIT=9,FILE=FNAMO,STATUS='UNKNOWN')
```

```
C  WRITE INPUT FILENAME TO OUPUT FILE
      WRITE(9,900)FNAMI
 900  FORMAT(/3X,'PROGRAM CLSEC VER 1.3          INPUT FILE : ',A12)
C  DETERMINE NUMBER OF ANALYSES TO READ (MAX=99), CONV AND PRINT OPTION
      READ(8,800)NSAM,ICONV,IPRN,ISTAT
 800  FORMAT(5I5)
      IF(NSAM.GT.99)NSAM=99
      IF(ICONV.GT.0)AIND1='Y'
      IF(IPRN.GT.0)AIND2='Y'
      IF(ISTAT.GT.0)AIND3='Y'
      WRITE(9,910)NSAM,AIND1,AIND2,AIND3
 910  FORMAT(/3X,'NUMBER OF SAMPLES :',I3,//3X,
     # 'CONVERT ELEMENTAL % TO WT% : ',A1,5X,'PRINT INPUT : ',A1,5X,
     # 'DO STATS : ',A1)
      SRSE=0.0
C
C  READING DATA FOR EACH SAMPLE
      DO 10 I=1,NSAM
         ABSUM=0.0
         WRITE(9,920)I
 920     FORMAT(/3X,28(1H*),' SAMPLE # ',I2,' ',28(1H*))
C  READ SAMPLE NAME
         READ(8,805) STITLE
 805     FORMAT(A80)
         WRITE(9,925)STITLE
 925     FORMAT(/3X,'SAMPLE: ',A80)
C  READ POROUS MEDIA DATA: POROSITY, SPEC. WT. OF ROCK MATERIAL,
C   SATUATION INDEX, CHLORIDE CONCENTRATION IN MG/L WATER.
         READ(8,810)POR,SPWTR,SAT,CCLW
 810     FORMAT(9F8.0)
         IF(IPRN.GT.0)WRITE(9,930)POR,SPWTR,SAT,CCLW
 930     FORMAT(/3X,'POROSITY =',F5.3,3X,'SPECIFIC WT ROCK =',F5.3,
     #    /3X,'SATURATION =',F5.3,3X,'CL IN WATER =',F6.2)
C  READ CHEM ARRAY DATA FOR SAMPLE
         READ(8,810)(CHEM(J),J=1,27)
C  ZERO OUT THE PPM MN VALUE IF AN OXIDE MN VALUE IS GIVEN
         IF(CHEM(IMNO).GT.0.0)CHEM(IMN)=0.0
C  CONVERT ELEMENTAL % TO OXIDE WT% IF REQUESTED
         IF(ICONV.GT.0)THEN
            DO 110 J=1,13
               CHEM(J)=CHEM(J)*CONV(J)
110         CONTINUE
         ENDIF
C  PRINT GIVEN CHEM DATA IF REQUESTED
         IF(IPRN.GT.0)WRITE(9,940)(CHEM(J),J=1,27)
 940     FORMAT(/7X,'SIO2',3X,'AL2O3',3X,'FE2O3',5X,'FEO',5X,'MGO',5X,
     #    'CAO',4X,'NA2O',5X,'K2O',4X,'TIO2',/,3X,9F8.2,//8X,'MNO',4X,
     #    'P2O5',5X,'H2O',5X,'CO2',6X,'LI',6X,'RB',6X,'SR',6X,'MN',7X,
     #    'U',/,3X,9F8.2,//9X,'TH',6X,'CL',6X,'SM',6X,'GD',7X,'B',7X,
     #    'F',6X,'BE',7X,'N',7X,'S',/,3X,9F8.2)
C  DRY BULK DENSITY
         BLKD=(1-POR)*SPWTR
C  TOTAL WET POROUS MEDIA MASS PER VOLUME
```

```
C        DENS=POR*(1-SAT)*SPWTA+POR*SAT*SPWTW+BLKD--WITH DENSITY OF AIR
         DENS=POR*SAT*SPWTW+BLKD
C  VOLUME OF WATER (L) PER KG WET POROUS MEDIA (NOTE: EQ. 14 OF
C   BENTLEY ET AL., 1986, CHAPTER 10, HANDBOOK OF ISOTOPE GEOCHEMISTRY
C   DOES THIS SLIGHTLY WRONG BY CALCULATING VOLW=POR/BLKD)
         VOLW=POR*SAT/DENS
         WRITE(9,945) VOLW
 945     FORMAT(1P,/13X,'CALCULATED WATER VOLUME (L/KG MEDIA) =',
     #    E11.4,0P)
C  MASS OF DRY ROCK (KG) PER KG WET POROUS MEDIA
         RMASS=BLKD/DENS
C        AMASS=POR*(1-SAT)*SPWTA/DENS-- MASS OF DENSE AIR
C  CONVERT WATER CL CONCENTRATION FROM MG/L TO MOLES/L
         CCLW=CCLW/35450.
C  MOLES OF CL IN WATER PER KG WET MEDIA
         NWCL=VOLW*CCLW
C  ASSOCIATED NEUTRON ABSORPTION
         ABWCL=NWCL*XSEC(ICL)*BARCM2*AV
C  MOLES OF WATER MOLECULES IN PORES PER KG WET MEDIA
         NWH2O=VOLW*1000/18.015
C  ASSOCIATED NEUTRON ABSORPTION
         ABWH2O=NWH2O*(2*XSEC(IH2O)+XSEC(IOXY))*BARCM2*AV
         WRITE(9,950) ABWH2O,ABWCL
 950     FORMAT(1P,10X,'ABSORPTION BY PORE WATER (CM2/KG MEDIA) =',
     #    E11.4,/4X,'ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) =',
     #    E11.4,0P)
C
C  DO LOOP CALCULATES SUM OF NEUTRON ABSORPTION IN ROCK IN
C   MOLES-CM2/100G AND THEN ATOM-CM2 PER KG ROCK
         DO 20 J=1,IOXY
            IF(J.NE.IOXY)THEN
C   CONVERT INPUT TO MOLES, MULTIPLY BY THERMAL XSEC, AND ADD TO SUM
               CM(J)=CHEM(J)/MOLE(J)
               ABSUM=ABSUM+CM(J)*XSEC(J)
            ELSE
C   CALCULATE MOLES OF OXYGEN BASED ON ELEMENTAL OXIDES
               CM(IOXY)=CM(ISI)*2+CM(IAL)*3/2+CM(IFE2)*3/2+CM(IFE)
     #          +CM(IMG)+CM(ICA)+CM(INA)/2+CM(IK)/2+CM(ITI)*2+CM(IMNO)
     #          +CM(IP)*5/2+CM(IH2O)/2+CM(ICO2)*2
               ABSUM=ABSUM+CM(IOXY)*XSEC(IOXY)
C   CONVERT MOLES-BARNES TO ATOMS-CM2, AND PER 100 G TO PER KG ROCK
               ABSUM=ABSUM*BARCM2*AV*10.
C   CONVERT ABSORPTION SUM (ABSUM) TO PER KG WET POROUS MEDIA
               ABSUM=ABSUM*RMASS
C   NOW THAT OXYGEN IS ESTABLISHED, COMBINE FE2 AND FE
               CM(IFE)=CM(IFE)+CM(IFE2)
            ENDIF
 20      CONTINUE
         CHEM(IOXY)=CM(IOXY)*MW(IOXY)*10000.
         WRITE(9,960)CHEM(IOXY),RMASS*CHEM(IOXY),ABSUM
 960     FORMAT(1P,20X,'CALCULATED PPM OXYGEN IN ROCK =',E11.4,/10X,
     #    'PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK =',E11.4,/14X,
     #    'NEUTRON ABSORPTION BY ROCK (CM2/KG) =',E11.4,0P)
```

```
C   CALCULATE NEUTRON FLUX IN N/KG MEDIA.
C   FLUX EQUATIONS FROM FABRYKA-MARTIN, 1988, PhD THESIS, UNIV. OF AZ
C    FLUX DUE TO SPONTANEOUS FISSION OF U238 (EQ. 3-16)
        U238=.9927*CM(IU)*AV*10
        SFU=U238*8.49E-17*2.0
C    FLUXES FROM U ACTIVATION OF ROCK ELEMENTS AND PORE WATER (TB. C-3)
        RFLX1=(454.*0.69*CM(ISI)*MW(ISI)+444.*5.1*CM(IAL)*MW(IAL)+351.
     #   *0.19*CM(IFE)*MW(IFE)+461.*5.8*CM(IMG)*MW(IMG)+456.*14.5
     #   *CM(INA)*MW(INA)+414.*0.45*CM(IK)*MW(IK)+561.*0.45*CM(ICO2)
     #   *MW(ICO2)+548.*21.1*CM(ILI)*MW(ILI)+527.*62.3*CM(IB)*MW(IB)
     #   +472.*30.8*CM(IFL)*MW(IFL)+529.*265.*CM(IBE)*MW(IBE)+527.
     #   *0.23*CM(IOXY)*MW(IOXY)+74.7*.077*CM(IU)*270.03)
        RFLX2=(454.*CM(ISI)*MW(ISI)+444.*CM(IAL)*MW(IAL)+351.*CM(IFE)
     #   *MW(IFE)+461.*CM(IMG)*MW(IMG)+456.*CM(INA)*MW(INA)+414.
     #   *CM(IK)*MW(IK)+561.*CM(ICO2)*MW(ICO2)+548.*CM(ILI)*MW(ILI)
     #   +527.*CM(IB)*MW(IB)+472.*CM(IFL)*MW(IFL)+529.*CM(IBE)*MW(IBE)
     #   +527.*CM(IOXY)*MW(IOXY)+74.7*CM(IU)*270.03+433.*CM(IP)*MW(IP)
     #   +439.*CM(ISUL)*MW(ISUL)+428.*CM(ICA)*MW(ICA)+375.*CM(ITI)
     #   *MW(ITI))
        RFLXU=RMASS*CHEM(IU)*(RFLX1/RFLX2)
        WATWF2=NWH2O*527.*MW(IOXY)
        WATWF1=WATWF2+NWH2O*2.*1563.*MW(IH2O)
        PMFLXU=RMASS*CHEM(IU)
     #   *((10.*RMASS*RFLX1+0.23*WATWF2)/(10.*RMASS*RFLX2+WATWF1))
C    FLUXES FROM TH ACTIVATION OF ROCK ELEMENTS AND PORE WATER (TB C-3)
        RFLX3=(454.*0.335*CM(ISI)*MW(ISI)+444.*2.6*CM(IAL)*MW(IAL)
     #   +351.*0.205*CM(IFE)*MW(IFE)+461.*2.6*CM(IMG)*MW(IMG)+456.
     #   *6.8*CM(INA)*MW(INA)+414.*0.305*CM(IK)*MW(IK)+561.*0.18
     #   *CM(ICO2)*MW(ICO2)+548.*9.6*CM(ILI)*MW(ILI)+527.*19.2*CM(IB)
     #   *MW(IB)+472.*11.8*CM(IFL)*MW(IFL)+529.*91.2*CM(IBE)*MW(IBE)
     #   +527.*0.079*CM(IOXY)*MW(IOXY)+74.7*.027*CM(IU)*270.03)
        RFLXTH=RMASS*CHEM(ITH)*(RFLX3/RFLX2)
        PMFLXT=RMASS*CHEM(ITH)
     #   *((10.*RMASS*RFLX3+0.079*WATWF2)/(10.*RMASS*RFLX2+WATWF1))
C  SUM OF NEUTRON FLUXES
        SFFLX=RMASS*SFU
        TRNFLX=SFU+1000.*(RFLXU+RFLXTH)/RMASS
        RNFLXU=1000.*RMASS*RFLXU
        RNFLXT=1000.*RMASS*RFLXTH
        RNFLX=SFFLX+RNFLXU+RNFLXT
        SNFLX=SFFLX+1000.*(PMFLXU+PMFLXT)
        WNFLX=((PMFLXU+PMFLXT)-RMASS*(RFLXU+RFLXTH))*1000.
C    #   -AMASS*(PMFLXU+PMFLXT))*1000.
        WRITE(9,970)SFFLX,RNFLXU,RNFLXT,TRNFLX,RNFLX,WNFLX,SNFLX
 970    FORMAT(1P,/5X,'SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA =',
     #   E11.4,/10X,'ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA =',
     #   E11.4,/9X,'ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA =',
     #   E11.4,/7X,'TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK =',
     #   E11.4,' (TRNFLX)',/6X,'TOTAL ROCK NEUTRON FLUX (N/YR) PER KG',
     #   ' MEDIA =',E11.4,' (RNFLX)',/3X,'NET PORE WATER ADJUSTMENT',
     #   ' TO FLUX PER KG MEDIA =',E11.4,/11X,'TOTAL NEUTRON FLUX',
     #   ' (N/YR) PER KG MEDIA =',E11.4,' (SNFLX)',0P)
C  DETERMINE THE SECULAR EQUILIBRIUM OF 36CL/CL (RSE), EQ 17 FROM
```

```
C   BENTLEY ET AL., 1986, CHAPTER 10, HANDBOOK OF ISOTOPE GEOCHEMISTRY
         D=((2.236E-6)*(ABSUM+ABWCL+ABWH2O))
         RSER=(0.7577*TRNFLX*43.3*BARCM2)/D
         RSE1(I)=(0.7577*RNFLX*43.3*BARCM2)/D
         RSE2(I)=(0.7577*SNFLX*43.3*BARCM2)/D
         WRITE(9,980)RSER,RSE1(I),RSE2(I)
 980     FORMAT(1P,/9X,'SECULAR EQUILIBRIUM RATIO (USING TRNFLX) =',
     #    E11.4,/10X,'SECULAR EQUILIBRIUM RATIO (USING RNFLX) =',
     #    E11.4,/10X,'SECULAR EQUILIBRIUM RATIO (USING SNFLX) =',
     #    E11.4,0P)
         SRSE=RSE2(I)+SRSE
 10   CONTINUE
      IF(NSAM.LT.2)GOTO 1000
      IF(ISTAT.LE.0)GOTO 1000
      WRITE(9,985)
 985  FORMAT(/5X,'************ SAMPLE STAT SUMMARY *************')
C  MEAN IS THE MEAN SECULAR EQUILIBRIUM OF 36CL/CL
      MEAN=SRSE/NSAM
      SMSQ=0.0
      DO 40 N=1,NSAM
         DIF2=(RSE2(N)-MEAN)**2
         SMSQ=SMSQ+DIF2
  40  CONTINUE
C  SDEV IS THE STANDARD DEVIATION
      SDEV=SQRT(SMSQ/(NSAM-1))
      WRITE(9,990) MEAN,SDEV
 990  FORMAT(1P,/3X,'MEAN RATIO (USING SNFLX) = ',E10.3,3X,
     #  'STD DEVIATION= ',E10.3,0P)
 1000 STOP
      END
```

Appendix B.2.a. Listing of input file used to verify the CLSEC program.

```
   5    1    1
Test case 1, Porosity=0, Granite
   0.00    2.70    1.0     0.0
   31.5    8.23    2.97    0.00    0.94    2.54    2.85    2.53    0.34
   .054    0.09    0.00    0.03    24.1    110.4   442.    0.0     3.01
   8.53    130.5   8.83    8.83    9.03    522.    2.0     20.1    301.
Test case 2, Saturation=0, Granite
   0.01    2.70    0.0     0.0
   31.5    8.23    2.97    0.00    0.94    2.54    2.85    2.53    0.34
   .054    0.09    0.00    0.03    24.1    110.4   442.    0.0     3.01
   8.53    130.5   8.83    8.83    9.03    522.    2.0     20.1    301.
Test 3, Saturated Granite, mod. from Fabryka-Martin, 1988
   0.01    2.70    1.0     0.0
   31.5    8.23    2.97    0.00    0.94    2.54    2.85    2.53    0.34
   .054    0.09    0.00    0.03    24.1    110.4   442.    0.0     3.01
   8.53    130.5   8.83    8.83    9.03    522.    2.0     20.1    301.
Test 4, Saturated Clay/shale, mod. from Fabryka-Martin, 1988
    0.1    2.6     1.0     0.0
   25.0    11.0    3.50    0.0     1.41    2.66    .069    2.40    0.47
   .071    .081    0.0     1.05    63.     210.    474.    0.0     3.37
  11.58    168.    6.8     6.8     105.    526.    3.2     63.     3157.
Test 5, Saturated Sandstone, mod. from Fabryka-Martin, 1988
    0.2    2.6     1.0     0.0
   40.3    2.7     1.07    0.0     .77     4.29    .362    1.17    0.16
   .005    .019    0.0     .033    16.4    66.     22.     0.0     0.493
   1.86    11.     11.     11.     38.     296.    0.55    22.     263.
```

Appendix B.2.b. Listing of verification case output file.

```
PROGRAM CLSEC VER 1.3          INPUT FILE : fbm.in

NUMBER OF SAMPLES :  5

CONVERT ELEMENTAL % TO WT% : Y     PRINT INPUT : Y     DO STATS : N

**************************** SAMPLE #  1 ****************************

SAMPLE: Test case 1, Porosity=0, Granite

POROSITY = .000   SPECIFIC WT ROCK =2.700
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   67.39   15.55    4.10     .00    1.56    3.55    3.84    3.05     .50

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .07     .21     .00     .11   24.10  110.40  442.00     .00    3.00

      TH      CL      SM      GD       B       F      BE       N       S
    8.53  130.50    8.83    8.83    9.03  522.00    2.00   20.10  301.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 0.0000E+00
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 0.0000E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 4.7993E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.7993E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 6.4621E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.2797E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.5501E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 4.7838E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 9.6137E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6137E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 0.0000E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6137E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.1829E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 2.1829E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 2.1829E-14

**************************** SAMPLE #  2 ****************************

SAMPLE: Test case 2, Saturation=0, Granite

POROSITY = .010   SPECIFIC WT ROCK =2.700
SATURATION = .000   CL IN WATER =   .00
```

```
   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  67.39   15.55    4.10     .00    1.56    3.55    3.84    3.05     .50

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .07     .21     .00     .11   24.10  110.40  442.00     .00    3.00

     TH      CL      SM      GD       B       F      BE       N       S
   8.53  130.50    8.83    8.83    9.03  522.00    2.00   20.10  301.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 0.0000E+00
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 0.0000E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 4.7993E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.7993E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 6.4621E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.2797E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.5501E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 4.7838E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 9.6137E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6137E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 0.0000E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6137E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.1829E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 2.1829E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 2.1829E-14

**************************** SAMPLE #  3 ****************************

SAMPLE: Test 3, Saturated Granite, mod. from Fabryka-Martin, 1988

POROSITY = .010   SPECIFIC WT ROCK =2.700
SATURATION =1.000   CL IN WATER =    .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  67.39   15.55    4.10     .00    1.56    3.55    3.84    3.05     .50

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .07     .21     .00     .11   24.10  110.40  442.00     .00    3.00

     TH      CL      SM      GD       B       F      BE       N       S
   8.53  130.50    8.83    8.83    9.03  522.00    2.00   20.10  301.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 3.7272E-03
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 8.2909E-02
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 4.7993E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.7815E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 6.4380E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.2749E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.5237E+03
```

```
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 4.7482E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 9.6137E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.5469E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA =-5.2734E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.5416E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.1632E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 2.1482E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 2.1470E-14

**************************** SAMPLE #  4 ****************************

SAMPLE: Test 4, Saturated Clay/shale, mod. from Fabryka-Martin, 1988

POROSITY = .100   SPECIFIC WT ROCK =2.600
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   53.48   20.78    4.84     .00    2.34    3.72     .09    2.89     .67

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .09     .19     .00    3.85   63.00  210.00  474.00     .00    3.30

      TH      CL      SM      GD       B       F      BE       N       S
   11.58  168.00    6.80    6.80  105.00  526.00    3.20   63.00 3157.00

            CALCULATED WATER VOLUME (L/KG MEDIA) = 4.0984E-02
         ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 9.1165E-01
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                  CALCULATED PPM OXYGEN IN ROCK = 4.5413E+05
         PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3552E+05
              NEUTRON ABSORPTION BY ROCK (CM2/KG) = 9.7699E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.3500E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.0028E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 5.0509E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.0164E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.4037E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA =-7.0850E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.3329E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 1.3963E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.2918E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.2820E-14

**************************** SAMPLE #  5 ****************************

SAMPLE: Test 5, Saturated Sandstone, mod. from Fabryka-Martin, 1988

POROSITY = .200   SPECIFIC WT ROCK =2.600
SATURATION =1.000   CL IN WATER =   .00
```

```
  SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
 86.21    5.10    1.48     .00    1.28    6.00     .49    1.41     .17

   MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
   .01     .04     .00     .12   16.40   66.00   22.00     .00     .49

    TH      CL      SM      GD       B       F      BE       N       S
  1.86   11.00   11.00   11.00   38.00  296.00     .55   22.00  263.00

         CALCULATED WATER VOLUME (L/KG MEDIA) = 8.7719E-02
      ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 1.9513E+00
ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.1521E+05
      PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.7002E+05
          NEUTRON ABSORPTION BY ROCK (CM2/KG) = 6.0467E+00

 SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.9069E+02
      ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 2.4751E+02
     ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 4.3010E+02
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.0232E+03 (TRNFLX)
  TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 8.6830E+02 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 2.3307E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 8.7063E+02 (SNFLX)

     SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 1.8771E-15
      SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.5930E-15
      SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.5972E-15
```

Appendix B.3.a. Listing of input file containing Morrison formation CLSEC runs.

```
  20    1    1
USGS SSJB DATA, U=3, TH=4.5
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       3.
    4.5    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=5, TH=7
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       5.
     7.    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=4
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
     4.    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS PLUTO DATA, U=8, TH=4
   0.30    2.65     1.0     0.0
   40.0    3.50    1.08    0.00    0.20    0.11    0.78    2.40     0.13
 0.0113   0.016    0.00    0.00    13.1     93.    72.3     0.0       8.
    4.0    100.      4.      4.    25.8     0.0    0.66     0.0     325.
USGS SSJB DATA, U=8, TH=12
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
    12.    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS PLUTO DATA, U=8, TH=12
   0.30    2.65     1.0     0.0
   40.0    3.50    1.08    0.00    0.20    0.11    0.78    2.40     0.13
 0.0113   0.016    0.00    0.00    13.1     93.    72.3     0.0       8.
    12.    100.      4.      4.    25.8     0.0    0.66     0.0     325.
USGS SSJB DATA, U=8, TH=12, porosity=.2
   0.20    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
    12.    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=12, porosity=.4
   0.40    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
    12.    100.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=12, Cl=200 ppm
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
    12.    200.      4.      4.    25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=12, Cl=50 ppm
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00    0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14    13.1     93.     94.     0.0       8.
```

```
    12.     50.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=12, Gd & Sm=8 ppm
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0       8.
    12.    100.      8.      8.     25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=12, Gd & Sm=8, Cl=200 ppm, porosity=.35
   0.35    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0       8.
    12.    200.      8.      8.     25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=8
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0       8.
     8.    100.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=8, TH=14
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0       8.
    14.    100.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS SSJB DATA, U=18, TH=9.8
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0      18.
    9.8    100.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS PLUTO DATA, U=18, TH=9.8
   0.30    2.65     1.0     0.0
   40.0    3.50    1.08    0.00     0.20    0.11    0.78    2.40     0.13
 0.0113   0.016    0.00    0.00     13.1     93.    72.3     0.0      18.
    9.8    100.      4.      4.     25.8     0.0    0.66     0.0     325.
USGS SSJB DATA : U=100, TH=30, SATURATED
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0     100.
    30.    100.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS SMITH LAKE DATA : U=100, TH=30, SATURATED
   0.30    2.65     1.0     0.0
   40.0    5.50    1.70    0.00     0.14    0.40    0.81    2.70     0.10
 0.0082   0.016    0.00    0.14     13.1     93.     94.     0.0     100.
   30.0    100.      4.      4.     25.8     0.0    0.65     0.0    4900.
USGS SSJB DATA : U=1000, TH=100, SATURATED
   0.30    2.65     1.0     0.0
   40.0    3.60    0.77    0.00     0.17    0.40    0.63    2.41     0.10
  0.017   0.016    0.00    0.14     13.1     93.     94.     0.0    1000.
   100.    100.      4.      4.     25.8     0.0    0.65     0.0      88.
USGS SMITH LAKE DATA : U=1000, TH=100, SATURATED
   0.30    2.65     1.0     0.0
   40.0    5.50    1.70    0.00     0.14    0.40    0.81    2.70     0.10
 0.0082   0.016    0.00    0.14     13.1     93.     94.     0.0    1000.
  100.0    100.      4.      4.     25.8     0.0    0.65     0.0    4900.
```

Appendix B.3.b. Listing of output file for Morrison Formation CLSEC runs.

```
PROGRAM CLSEC VER 1.3          INPUT FILE : usgs-sjb.in

NUMBER OF SAMPLES : 20

CONVERT ELEMENTAL % TO WT% : Y     PRINT INPUT : Y     DO STATS : N

**************************** SAMPLE #  1 ****************************

SAMPLE: USGS SSJB DATA, U=3, TH=4.5

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    3.00

      TH      CL      SM      GD       B       F      BE       N       S
    4.50  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2266E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.1016E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 1.4754E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 1.0288E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 4.6595E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 3.6058E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 5.8039E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 3.6116E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 9.3357E-15
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 7.2246E-15
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 7.2362E-15

**************************** SAMPLE #  2 ****************************

SAMPLE: USGS SSJB DATA, U=5, TH=7

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
```

```
     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    5.00

      TH      CL      SM      GD       B       F      BE       N       S
    7.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2267E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 1.8359E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 2.4591E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 1.6004E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 7.6116E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 5.8954E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.0321E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 5.9057E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 1.5250E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.1812E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.1833E-14

**************************** SAMPLE #  3 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=4

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

      TH      CL      SM      GD       B       F      BE       N       S
    4.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2267E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 9.1452E+02
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 9.9569E+03 (TRNFLX)
```

```
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 7.7865E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 2.5842E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 7.8124E+03 (SNFLX)

     SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 1.9949E-14
      SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.5601E-14
      SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.5653E-14

**************************** SAMPLE #  4 ****************************

SAMPLE: USGS PLUTO DATA, U=8, TH=4

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.61    1.49     .00     .33     .15    1.05    2.89     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .01     .04     .00     .00   13.10   93.00   72.30     .00    8.00

      TH      CL      SM      GD       B       F      BE       N       S
    4.00  100.00    4.00    4.00   25.80     .00     .66     .00  325.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0159E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3176E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2872E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 4.0518E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 9.4254E+02
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.0153E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 7.9318E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.6022E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 7.9479E+03 (SNFLX)

     SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.0175E-14
      SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.5762E-14
      SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.5793E-14

**************************** SAMPLE #  5 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
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    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
  12.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2268E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.7436E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6156E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.5478E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6311E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.4895E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.9265E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.9296E-14

**************************** SAMPLE #  6 ****************************

SAMPLE: USGS PLUTO DATA, U=8, TH=12

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.61    1.49     .00     .33     .15    1.05    2.89     .17

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .01     .04     .00     .00   13.10   93.00   72.30     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
  12.00  100.00    4.00    4.00   25.80     .00     .66     .00  325.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0159E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3176E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2874E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 4.0518E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.8276E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2697E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.8169E+03 (RNFLX)
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NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.8109E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.8187E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.5230E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.9507E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.9511E-14

**************************** SAMPLE #  7 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, porosity=.2

POROSITY = .200   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
  12.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

         CALCULATED WATER VOLUME (L/KG MEDIA) = 8.6207E-02
      ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 1.9176E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
               CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
      PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.6183E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.4871E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 3.1184E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 4.4340E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 3.0918E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 1.0644E+04 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA =-1.7517E+00
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 1.0642E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.8466E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 2.4385E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 2.4381E-14

**************************** SAMPLE #  8 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, porosity=.4

POROSITY = .400   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
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    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
  12.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 2.0101E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 4.4712E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.0381E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 3.9234E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.7266E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.3899E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.3638E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 8.4803E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 4.8083E+01
         TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 8.5284E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.1718E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.4823E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.4907E-14

**************************** SAMPLE #  9 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, Cl=200 ppm

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
  12.00  200.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2758E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.7436E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6156E+03 (RNFLX)
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NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.5478E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6311E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.4729E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.9137E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.9168E-14

**************************** SAMPLE # 10 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, Cl=50 ppm

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

      TH      CL      SM      GD       B       F      BE       N       S
   12.00   50.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2023E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.7436E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6156E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.5478E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6311E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.4978E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.9330E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.9361E-14

**************************** SAMPLE # 11 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, Gd & Sm=8 ppm

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
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     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

      TH      CL      SM      GD       B       F      BE       N       S
   12.00  100.00    8.00    8.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.9078E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.7436E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6156E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.5478E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.6311E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.2777E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.7626E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.7655E-14

**************************** SAMPLE # 12 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=12, Gd & Sm=8, Cl=200 ppm, porosity=.35

POROSITY = .350   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

      TH      CL      SM      GD       B       F      BE       N       S
   12.00  200.00    8.00    8.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.6888E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.7566E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.2005E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.7859E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.8363E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.6680E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.5577E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2425E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.0619E+03 (RNFLX)
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NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 2.9716E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.0917E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.1342E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.5565E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.5616E-14

**************************** SAMPLE # 13 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=8

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

    MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
    .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

     TH      CL      SM      GD       B       F      BE       N       S
   8.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2267E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 1.8290E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.1191E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 8.7011E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 2.0660E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 8.7217E+03 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.2422E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.7433E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.7474E-14

**************************** SAMPLE # 14 ****************************

SAMPLE: USGS SSJB DATA, U=8, TH=14

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

   SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
  85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
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   MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
   .02     .04     .00     .51   13.10   93.00   94.00     .00    8.00

    TH      CL      SM      GD       B       F      BE       N       S
 14.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2268E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 2.9375E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 3.9345E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 3.2008E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.3042E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 1.0073E+04 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 1.2887E+01
         TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 1.0086E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.6131E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 2.0181E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 2.0207E-14

**************************** SAMPLE # 15 ****************************

SAMPLE: USGS SSJB DATA, U=18, TH=9.8

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

  SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
 85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

   MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
   .02     .04     .00     .51   13.10   93.00   94.00     .00   18.00

    TH      CL      SM      GD       B       F      BE       N       S
  9.80  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2269E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 6.6094E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 8.8526E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.2406E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 2.2650E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 1.7703E+04 (RNFLX)
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NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 5.7112E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 1.7760E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 4.5379E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 3.5467E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 3.5582E-14

**************************** SAMPLE # 16 ****************************

SAMPLE: USGS PLUTO DATA, U=18, TH=9.8

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =    .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.61    1.49     .00     .33     .15    1.05    2.89     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .01     .04     .00     .00   13.10   93.00   72.30     .00   18.00

      TH      CL      SM      GD       B       F      BE       N       S
    9.80  100.00    4.00    4.00   25.80     .00     .66     .00  325.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0159E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3176E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2875E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 6.6094E+03
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 9.1165E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.3092E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 2.3099E+04 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 1.8035E+04 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 3.4632E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 1.8070E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 4.5898E-14
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 3.5837E-14
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 3.5906E-14

**************************** SAMPLE # 17 ****************************

SAMPLE: USGS SSJB DATA : U=100, TH=30, SATURATED

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =    .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17
```

```
      MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
      .02     .04     .00     .51   13.10   93.00   94.00     .00  100.00

       TH      CL      SM      GD       B       F      BE       N       S
    30.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2286E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 3.6719E+04
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 4.9181E+04
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 6.8588E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.1829E+05 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.2758E+04 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 3.4909E+02
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.3107E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.3694E-13
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.8580E-13
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.8650E-13

**************************** SAMPLE # 18 ****************************

SAMPLE: USGS SMITH LAKE DATA : U=100, TH=30, SATURATED

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =   .00

     SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
    85.57   10.39    2.35     .00     .23     .56    1.09    3.25     .17

      MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
      .01     .04     .00     .51   13.10   93.00   94.00     .00  100.00

       TH      CL      SM      GD       B       F      BE       N       S
    30.00  100.00    4.00    4.00   25.80     .00     .65     .00 4900.00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.2716E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.5377E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.6599E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 3.6719E+04
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 5.4981E+04
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 7.7618E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2734E+05 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.9462E+04 (RNFLX)
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NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 4.9865E+02
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.9961E+04 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.4088E-13
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.8815E-13
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.8909E-13

**************************** SAMPLE # 19 ****************************

SAMPLE: USGS SSJB DATA : U=1000, TH=100, SATURATED

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =    .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57    6.80    1.06     .00     .28     .56     .85    2.90     .17

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .02     .04     .00     .51   13.10   93.00   94.00     .00 1000.00

      TH      CL      SM      GD       B       F      BE       N       S
  100.00  100.00    4.00    4.00   25.80     .00     .65     .00   88.00

           CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                 CALCULATED PPM OXYGEN IN ROCK = 5.0540E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.3504E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.2447E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 3.6719E+05
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 4.9174E+05
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.2859E+04
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.1211E+06 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 8.8179E+05 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 3.7634E+03
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 8.8555E+05 (SNFLX)

      SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.2406E-12
       SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.7624E-12
       SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.7699E-12

**************************** SAMPLE # 20 ****************************

SAMPLE: USGS SMITH LAKE DATA : U=1000, TH=100, SATURATED

POROSITY = .300   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =    .00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   85.57   10.39    2.35     .00     .23     .56    1.09    3.25     .17
```

Appendix B.4.a. Listing of input file for Great Artesian Basin data comparison.

```
    1    0    1
Test case 5, GAB
    0.20     2.65      1.0    100.0
    68.9     13.8     4.43      0.0     1.01     4.12     1.57     1.61      1.27
     0.0       0.       0.     1.32     31.3     55.3     191.     418.      1.66
    6.09       0.      4.5      4.3      30.       0.       0.       0.        0.
```

```
     MNO    P205     H20     CO2      LI      RB      SR      MN       U
     .01     .04     .00     .51   13.10   93.00   94.00     .00 1000.00

      TH      CL      SM      GD       B       F      BE       N       S
  100.00  100.00    4.00    4.00   25.80     .00     .65     .00 4900.00

           CALCULATED WATER VOLUME (L/KG MEDIA) = 1.3921E-01
        ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 3.0967E+00
  ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 0.0000E+00
                  CALCULATED PPM OXYGEN IN ROCK = 5.2716E+05
        PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.5377E+05
            NEUTRON ABSORPTION BY ROCK (CM2/KG) = 4.6760E+00

   SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 3.6719E+05
        ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 5.4974E+05
       ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.5869E+04
     TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 1.2034E+06 (TRNFLX)
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 9.4280E+05 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA = 5.1324E+03
          TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 9.4793E+05 (SNFLX)

       SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 2.2718E-12
        SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 1.7798E-12
        SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 1.7895E-12
```

Appendix B.4.b. Listing of output file for Great Artesian Basin data comparison.

```
PROGRAM CLSEC VER 1.3          INPUT FILE : test2.in

NUMBER OF SAMPLES :  1

CONVERT ELEMENTAL % TO WT% : N     PRINT INPUT : Y     DO STATS : N

**************************** SAMPLE #  1 ****************************

SAMPLE: Test case 5, GAB

POROSITY = .200   SPECIFIC WT ROCK =2.650
SATURATION =1.000   CL IN WATER =100.00

    SIO2   AL2O3   FE2O3     FEO     MGO     CAO    NA2O     K2O    TIO2
   68.90   13.80    4.43     .00    1.01    4.12    1.57    1.61    1.20

     MNO    P2O5     H2O     CO2      LI      RB      SR      MN       U
     .00     .00     .00    1.32   31.30   55.30  191.00  418.00    1.60

      TH      CL      SM      GD       B       F      BE       N       S
    6.09     .00    4.50    4.30   30.00     .00     .00     .00     .00

          CALCULATED WATER VOLUME (L/KG MEDIA) = 8.6207E-02
       ABSORPTION BY PORE WATER (CM2/KG MEDIA) = 1.9176E+00
 ABSORPTION BY CL IN PORE WATER (CM2/KG MEDIA) = 4.9062E-03
                 CALCULATED PPM OXYGEN IN ROCK = 4.8213E+05
       PPM OXYGEN IN MEDIA CONTRIBUTED BY ROCK = 4.4057E+05
           NEUTRON ABSORPTION BY ROCK (CM2/KG) = 5.6347E+00

  SPONTANEOUS FISSION FLUX (N/YR) PER KG MEDIA = 6.2368E+02
       ROCK FLUX FROM U (ALPHA,N) PER KG MEDIA = 1.1959E+03
      ROCK FLUX FROM TH (ALPHA,N) PER KG MEDIA = 2.1593E+03
    TOTAL ROCK NEUTRON FLUX (N/YR) PER KG ROCK = 4.7006E+03 (TRNFLX)
   TOTAL ROCK NEUTRON FLUX (N/YR) PER KG MEDIA = 3.9789E+03 (RNFLX)
NET PORE WATER ADJUSTMENT TO FLUX PER KG MEDIA =-3.5544E+01
        TOTAL NEUTRON FLUX (N/YR) PER KG MEDIA = 3.9433E+03 (SNFLX)

     SECULAR EQUILIBRIUM RATIO (USING TRNFLX) = 9.1266E-15
      SECULAR EQUILIBRIUM RATIO (USING RNFLX) = 7.7252E-15
      SECULAR EQUILIBRIUM RATIO (USING SNFLX) = 7.6562E-15
```