APPLICATION OF STABLE AND RADIOGENIC ISOTOPIC TECHNIQUES TO PROBLEMS OF SURFACE AND GROUNDWATER INTERACTIONS

Alan L Mayo[1], Steve Nelson[2], John Noakes[3], David Tingey[4] and Randy Culp[5]

AUTHORS: [1]Hydrogeologist, [2]Isotope Geochemist, [4]Geochemist, Department of Geology, Brigham Young University, Provo, UT 84602, [3]Isotope Geochemist, [5]Isotope Geochemist, Center for Applied Isotope Studies, University of Georgia, Athens, GA 30602.

Abstract. Stable and radiogenic isotopes of oxygen, hydrogen, and carbon are powerful tools for solving a wide range of hydrogeological problems. The stable isotopes of water, δ^2H and $\delta^{18}O$, can be used to distinguish several waters from each other, evaporation histories, and thermal conditions, regardless of their solute contents. The $\delta^{13}C$ content of a groundwater helps track the carbon history and sources of dissolved carbon and is critical for unraveling carbon-14 ages of water. The tritium (^3H) content helps determine if a component of recharge occurred during the past 50 years and the ^{14}C content allows the determination of the age of groundwater as old as 35,000 years. Carbon-14 is well suited to use in carbonate terrains and carbonate cemented rocks. Three case studies are briefly presented to illustrate the application of isotopes to: 1) evaluation of leakage of surface impoundments, 2) delineation of credible flow paths of groundwater, and 3) developing conceptual models of groundwater flow.

INTRODUCTION

The purpose of this paper is to present three case studies that illustrate the utility of stable and radiogenic isotopes in groundwater to solve a wide range of problems. The isotopes employed are naturally occurring in all waters and act both as self tracers of the water molecule itself, as well as chemical processes that modify the isotopic characteristics of the water. Although none of the case studies presented herein are located in Georgia, the principles and methods illustrated may be applied anywhere. Thus, understanding the potential value of isotopic methods enhances the ability of scientists, engineers, and public policy makers to evaluate and protect the largest source of fresh water on the planet, groundwater.

CASE 1: IDENTIFYING IMPOUNDMENT LEAKAGE – STABLE ISOTOPES OF WATER

Whether by design or otherwise surface water impoundments commonly leak. Depending on the chemical character of the pond, canal, or lake water and the receiving groundwater, and the leakage rate, determining such leakage is often difficult. The stable isotopes of water are often useful for detecting such leakage. Worldwide, the δ^2H and $\delta^{18}O$ of precipitation (rain and snow) generally follow the empirical relationship:

$$\delta^2H = s(\delta^{18}O) + d \ (‰)$$

Where s is the slope and d is the deuterium (hydrogen-2) excess (Merlivant and Jouzel, 1983). Craig (1961) and Dansgaard (1964) have shown that, on the global scale, s approximates 8 and d approximates 10 for coastal meteoric water. The Meteoric Water Line (MWL) is therefore defined as:

$$\delta^2H = 8(\delta^{18}O) + 10 \ (‰)$$

Groundwater recharged during cooler climates, seasons or at higher elevations will have more negative isotopic compositions than groundwater that recharged during warmer climates, season or at lower elevations (Figure 1, line A-B). Groundwaters, which have been heated above about 100°C during deep circulation, will exhibit a positive $\delta^{18}O$ shift relative to the δ^2H composition (line C-E). Surface water that has been subject to excessive evaporation plots along an evaporation trajectory (line C-D) commonly with a slope of 3 to 5.

Here we present a case history of pond leakage into an unconfined groundwater system. At a site in the western U.S. industrial waste water was disposed in a
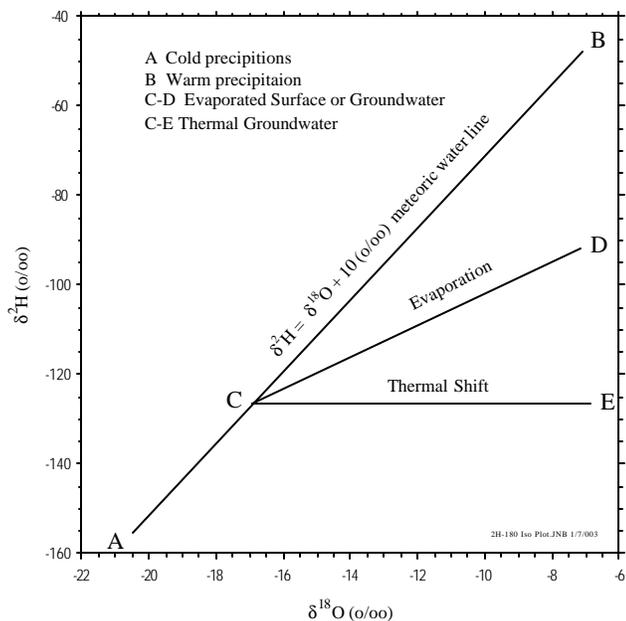
Figure 1. Stable isotopic compositions of surface and groundwater.

Figure 2. Stable isotopic compositions of evaporation pond and monitoring well waters.

evaporation pond. Synoptic solute sampling from 34 shallow monitoring wells and several pond locations were undertaken over several years. Because both pond and receiving groundwater contained elevated TDS contents and similar solute compositions, potential pond leakage could not be verified on the basis of solute compositions alone.

Stable isotopic compositions, collected during a single sampling event, clearly delineated groundwater contaminated with pond water (Figure 2). Background well water plots adjacent to the local MWL. Pond water has a strong evaporation signature.

Contaminated well waters plot in two groups along the evaporation trajectory. One group, consisting of wells located along the periphery of the evaporation pond, have isotopic compositions nearly identical to pond water and are mostly composed of pond water. The second group plots intermediate between background and pond water and are of a mixed origin.

CASE 2: TESTING FLOWPATHS – GROUNDWATER AGES

The question in this example is whether water is transmitted from one hydrographic basin to another through a bedrock aquifer. The study was conducted in Death Valley, for springs of the Furnace Creek area (Fig. 3). Although Pistrang and Kunkel (1964)

concluded that adequate local precipitation occurs to supply the required recharge, numerous other workers (e.g. Winograd and Eakin, 1965; Thomas et al., 1996: and others) suggested that local recharge had little to no impact on spring discharge. Rather, the high-flux springs in the Furnace Creek area result from interbasin flow external to Death Valley (i.e., Ash Meadows, Fig. 3) through fractured carbonate bedrock. Multiple evidences are cited, including similarities in δD and $\delta^{18}O$ between the springs at Ash Meadows and Furnace Creek.

The idea of interbasin transfer was re-evaluated by a combination of solute and isotopic techniques. Mass balance models were employed by examining: 1)
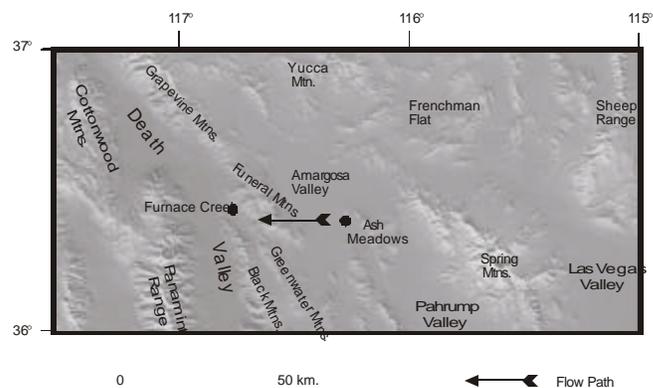
Figure 3. Map of the Death Valley region.

reactions required for precipitation to evolve to the composition of Furnace Creek waters (i.e. local recharge), 2) reactions required for Ash Meadows water to evolve to the composition of Furnace Creek waters (i.e. interbasin flow). The evolution from precipitation to Furnace Creek water requires the dissolution of CO_2 gas, calcite, dolomite, gypsum, halite and illite, all of which are known or can assumed to be present in the alluvium of the Furnace Creek drainage, including evaporite minerals.

On the basis of water-rock reactions alone, Ash Meadows may be easily evolved to Furnace Creek water by the dissolution gypsum, halite, and dolomite, accompanied by calcite precipitation. Both models require an equally plausible set of mass balance reactions. However, considering the similarities in δD and $\delta^{18}O$ values, a hydraulic connection between Ash Meadows cannot be ruled out.

Examination of stable and radiogenic isotopes of carbon (Anderson, 2002, and others), however, rules out the interbasin flow model.

Carbon isotopes in groundwater are modified from initial values acquired in the soil zone by subsequent water-rock interactions. The $\delta^{13}C$ of soil zone CO_2 is expected to be near -25‰ and an initial ^{14}C activity near 100% in regions dominated by C3 plants, which in turn produces a $\delta^{13}C$ of dissolved inorganic carbon (DIC) near -17‰ due to equilibrium fractionation which should remain unmodified in the absence of other carbon reservoirs with which the DIC can interact. However, each mole of acidity acquired in the soil zone by CO_2 dissolution may result in a mole of dissolution of carbonate rock having $\delta^{13}C$ values near 0‰ and 0% ^{14}C activity. Thus, a "modern" groundwater in a carbonate aquifer may have $\delta^{13}C$ values on the order of -8‰ and only 50% modern ^{14}C activities.

The mean $\delta^{13}C$ value for Furnace Creek is –6.36‰ (n=20), whereas the mean for Ash Meadows is –4.66‰ (n=21). A t-test shows the probability that these two samples represent the same population (assuming equal variance) is nearly zero (i.e., 4.2 x 10^{-5}). Similarly, the mean ^{14}C activity for Furnace Creek is 9.6 percent modern carbon (pmc) (n=8), whereas the mean pmc for Ash Meadows is 3.8 (n=21). The probability that these two samples represent the same populations is very small (i.e., 0.001).

Although $\delta^{13}C$ and values ^{14}C activities can change along a flowpath by water-rock interactions, etc., the apparent changes between Furnace Creek and Ash Meadows occur in the wrong sense. Reactions between groundwater and fractured carbonate rocks

would increase, rather than decrease, $\delta^{13}C$ values and ^{14}C activities.

CASE 3: CONCEPTUAL MODELS - STABLE ISOTOPES AND GROUNDWATER AGES

Patterns and mechanisms of groundwater circulation in stratified mountainous terrain are generally not well understood, either conceptually or at the field scale. This occurs because access to deep groundwater is generally limited, the details of subsurface stratigraphic and structural complexities are often poorly defined, and groundwater flow is often fracture controlled. We have used geological, physical, chemical and isotopic data to evaluate interactions between near surface and 300 to 700 m deep groundwater systems in gently dipping stratified bedrock that underlies a 6,500 km^2 mountainous region. Thousands of springs issue from the 700+ m thick, gently dipping, clastic bedrock formations. Deep groundwater samples accessed by mine openings Springs respond to seasonal and climatic variability and the waters contain appreciable ^3H and anthropogenic ^{14}C. In-mine groundwater discharge rates have δ^2H and $\delta^{18}O$ compositions that are distinguishable from near surface groundwater (Figure 4).

Deep groundwaters have more negative isotopic compositions than the near surface counterparts, suggesting cooler climatic recharge conditions.
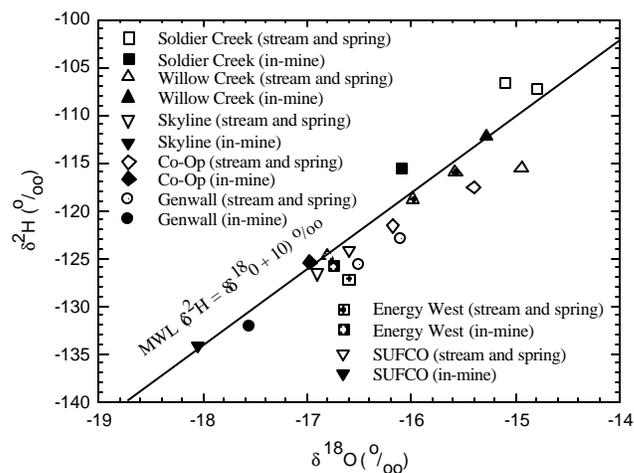
Figure 4. Scatter plot of mean stable isotopic composition of near surface and deep (in-mine) groundwater.
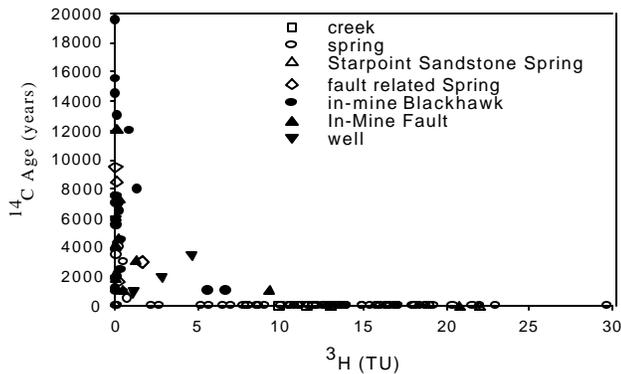
Figure 5. Scatter plot of ^{14}C mean residence times ("age") vs. ^{3}H contents of near surface and deep groundwater.

In general, deep waters have no ^{3}H and have mean ^{14}C residence times of 500 to 20,000 years (Figure 5). Differences in the stable isotopic compositions and mean groundwater ages between near surface and deep groundwaters (combined with other physical and chemical data not presented here) suggest the deep systems waters do not have active hydraulic communication with near surface systems. Many of the deep groundwaters recharged during cooler climatic times.

CONCLUSIONS

Here we have shown how stable and radiogenic isotopes are powerful tools for evaluation of groundwater recharge locations and mechanisms, flow paths, and discharge locations and mechanisms. The three brief case histories highlight only three of the many possible isotopic tools available. The stable isotopes of water can be used to distinguish waters from each other irrespective of intervening changes in solute compositions. Groundwater ages are often indispensable in sorting out actual flow paths and flow histories. Isotopic techniques can be used to supplement physical and chemical data and often provide the critical data needed to solve difficult groundwater problems.

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