

Potential application of oxygen-18 and deuterium in mining effluent and acid rock drainage studies

M. M. Ghomshei · D. M. Allen

Abstract Oxygen-18 (^{18}O) and deuterium (D, or ^2H) are routinely used in hydrologic, climatologic and geothermal studies. In hydrology, stable isotopes provide information on the type and topology (altitude and latitude) of the recharge waters and the historical effects on water, related to such physical processes as evaporation (in ponds), melting (of snow or ice), condensation, evapotranspiration and mixing. In geothermal studies, stable isotopes provide key information related to recharge and the various temperature-dependent water/rock isotope exchange reactions. The latter is assessed through the oxygen shift in the $^{18}\text{O}/\text{D}$ correlation. At acid rock drainage (ARD) sites, water/rock interactions are primarily controlled by pH and oxidation potential. Using the isotopic characteristics of the rocks and the recharge waters as a basis, the relative oxygen shift of the ARD effluent can provide information on: (1) the residence time, (2) the rate of water/rock reactions, and (3) the actual pH at the rock/water interface. This paper offers a methodology for conducting oxygen and hydrogen isotope studies related to ARD and other mineral effluent problems. The methodology is based on: (1) comprehensive sampling of regional waters, ARD effluent and major contributing minerals and rocks, (2) isotopic and elemental analysis, and (3) data interpretation on the basis of a zero-dimensional (mass balance), multi-component mixing model.

Key words Stable isotopes · Acid rock drainage · Mining effluent · Geochemical mixing

Background

Different isotopes of the same element differ slightly in chemical, and more significantly, in physical properties because of their mass differences. These mass differences are more pronounced in light elements, therefore, isotopes of light elements are commonly used to characterize the origin and cycle of different materials in environmental systems. The nuclear stability and the relative abundance of isotopes of oxygen and hydrogen have been widely documented (Table 1). Among these isotopes, ^{18}O , ^{16}O , ^1H , ^2H (D) and ^3H are routinely used in hydrological, climatological and geological studies.

Oxygen and hydrogen isotopes are important in environmental studies because they are constituents of water and because they are conservative at near-surface temperatures. Oxygen and hydrogen isotopes are often used together in order to characterize the present and past processes in which water is a dynamic component. An important attribute of these isotopes, which makes them relatively unique in isotopic studies, is that their relative abundance in nature is highly variable. Isotopes of light elements (i.e. with low atomic numbers) undergo significant partitioning (enrichment or depletion) during the course of many mass-dependent chemical and physical processes. For example, the relative mass difference between hydrogen isotopes (^1H and D) is two orders of magnitude greater than that of uranium isotopes. Therefore, natural isotopic partitioning (or fractionation) is detectable only in light elements up to potassium and calcium in the periodic table of elements. The most important elements, in which natural variations of the isotopic composition have been observed, include hydrogen, oxygen, carbon, nitrogen and sulfur (Jager and Hunziker 1979).

Stable isotopes are analyzed using a mass spectrometer. ^{18}O and D (Deuterium) are determined by equilibration with carbon dioxide and uranium reduction, respectively. Analytical results are expressed as "permil" differences (denoted by $\delta^{18}\text{O}\text{‰}$ and $\delta\text{D}\text{‰}$ from VSMOW (Vienna Standard Mean Ocean Water) with analytical precision of ± 0.15 and ± 1 permil, respectively. Measurements on VSMOW show that:

$$\text{D}/\text{H} (\text{VSMOW}) = (155.76 \pm 0.05) \times 10^{-6} \quad (1)$$

$$^{18}\text{O}/^{16}\text{O} (\text{VSMOW}) = (2005.2 \pm 0.45) \times 10^{-6} \quad (2)$$

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Table 1
Relative abundance and half-lives of oxygen and hydrogen isotopes

Isotope	Relative abundance	Nuclear stability	Half-life*
^{14}O		Radioactive	Tens of seconds
^{15}O		Radioactive	Tens of seconds
^{16}O	99.759%	Stable	
^{17}O	0.037%	Stable	
^{18}O	0.204%	Stable	
^{19}O		Radioactive	Tens of seconds
^1H	99.985%	Stable	
^2H (deuterium, D)	0.0147%	Stable	
^3H (tritium)		Radioactive	12.26 years

* For radioactive isotopes only

Permil differences from the VSMOW are defined as follows:

$$\delta\text{D} = \left\{ \left[\frac{(\text{D}/\text{H})_{\text{sample}} - (\text{D}/\text{H})_{\text{SMOW}}}{(\text{D}/\text{H})_{\text{SMOW}}} \right] \times 10^3 \right\} \text{‰} \quad (3)$$

$$\delta^{18}\text{O} = \left\{ \left[\frac{(^{18}\text{O}/^{16}\text{O})_{\text{sample}} - (^{18}\text{O}/^{16}\text{O})_{\text{SMOW}}}{(^{18}\text{O}/^{16}\text{O})_{\text{SMOW}}} \right] \times 10^3 \right\} \text{‰} \quad (4)$$

Tritium (^3H) is often used for dating of groundwater because of its short half-life (in geological terms). While naturally-occurring tritium has been rarely reported, tritium fallout derived from hydrogen bomb tests conducted in the 1950s is still detectable in the long-cycled groundwater systems. Detectable concentrations of tritium in groundwater indicates that the groundwater was recharged less than 45 years ago (Taylor and others 1963). Tritium concentrations are determined for non-enriched water samples in tritium units ($1\text{TU} = 1^3\text{H} \times 10^{-18}^1\text{H}$; e.g. Clark and others 1982).

In environmental studies, including mineral industry effluent investigations, the quantification of chemical and physical processes is important. In acid rock drainage (ARD), these processes can be divided into two categories: (1) chemical processes (e.g. oxidation, neutralization, and water/rock elemental and isotopic exchange), and (2) physical processes (e.g. evaporation, condensation, and oxygen transfer). Stable isotopes are fractionated during both physical and chemical processes observed in ARD. Quantification of isotope fractionation will significantly enhance an understanding of the nature and magnitude of the processes involved in the generation of effluent. Unfortunately, isotope tools have not yet been implemented by those involved in mineral industry effluent studies. This is partially due to the fact that mineral industry effluent research is still in its infancy. While there has been a great demand for adopting different analytical and interpretational tools, especially those that have been successfully tested in other environmental fields, stable isotopic analysis has been largely overlooked as a potential tool. This paper aims to introduce the potential application of oxygen and hydrogen isotopes to ARD and other mineral industry effluent problems, and to initiate further interest in stable oxygen and hydrogen interpreta-

tional techniques that can be applied to hydrological and hydrogeological studies at ARD sites.

Oxygen-18 and deuterium in mineral industry effluent

Oxygen and hydrogen isotopes are useful natural tracers in fluid and solid phases that affect the mineral industry effluent. For example, ARD fluids are expected to carry the isotopic signature of both the source waters and the reacting minerals (solid phase).

Isotopic signatures of the solid phase

The minerals taking part in the generation of ARD effluent are dominantly silicates, carbonates and sulphides. Silicates can exchange oxygen with water, especially under low pH conditions in which silicates undergo rapid alteration (e.g. Sherlock and others 1995). Oxygen exchange of the fluid phase with carbonates is important, especially in the case of calcite and dolomite. Sulphides can indirectly affect the oxygen isotopic system in the process of oxidization (Taylor and others 1984) and during isotopic equilibration of the generated sulphates with the fluid phase.

Sulphur isotopes (e.g. Fouillac and others 1993) can be used in order to assess more directly the contribution of sulphides in the ARD process. Sulphur isotopes applicable to ARD and other sulphate-rich effluent are the heavy sulphur (^{34}S) and the common sulphur (^{32}S). The ratio of these two isotopes, reflected in different $\delta\text{‰}$ values (Canyon Diablo Triolite CDT standard), is characteristic of different sulphur sources and fractionation processes. A more rigorous discussion of sulphur isotopes is beyond the scope of this review. However, it should be noted that the combination of $\delta^{18}\text{O}$ and $\delta^{34}\text{S}$ (e.g. Pearson and Rightmire 1980) can be useful in the assessment of ARD rates, especially in the consideration and evaluation of bacterial processes (Pierre 1989).

Considering their relatively fast and easy reaction, it can be reasonably assumed that the major minerals (in solid

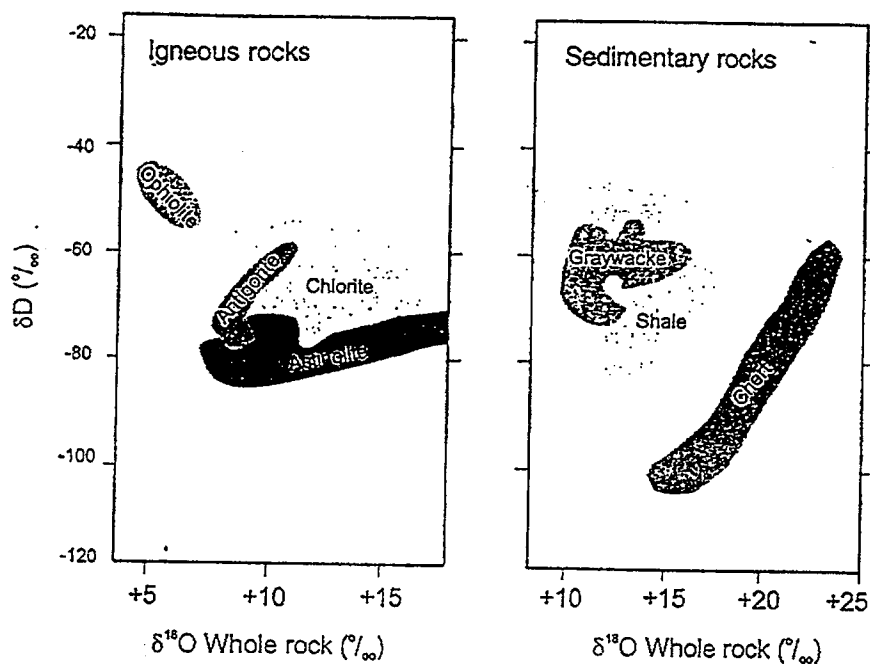


Fig. 1
 δD vs $\delta^{18}O$ for igneous and sedimentary rocks (after Taylor 1977)

phase) affecting the oxygen isotopic signature of ARD are carbonates. Carbonates demonstrate a wide range of isotopic values, depending on the genetic (e.g. biogenic or "mineralogenic") and paleo-environments of their formation. Negative $\delta^{18}O$ values (as low as -12‰) have been reported in many carbonate formations including Jurassic dolomites and calcites from Central Tunisia (Soussi and M'Rabet 1994) and Silurian/Devonian carbonates from the Illinois Basin (Kruger and Simo 1994). Other investigators have reported considerably more enriched carbonates with $\delta^{18}O$ values of $+16\text{‰}$ to $+22\text{‰}$ (Fouillac and others 1993). Therefore, case-specific carbonate isotope values are useful in ARD investigations.

Assessment of isotopic values of silicates can be important in low pH environments, in which silicates may play a significant role in the final chemistry of the fluid phase. Oxygen isotopic values of some igneous and sedimentary

rocks have been compiled by Taylor (1977; Fig. 1). Jager and Hunziker (1979) have compiled the values for parent rock types (Fig. 2).

Isotopic signatures of the fluid phase

Because the isotopic value of atmospheric oxygen ($\delta^{18}O = +14\text{‰}$) is significantly different from that of water, oxidation processes and oxygen transport can be characterized (e.g. Richi 1994; Taylor and others 1984). The isotopic fractionation in pyrite oxidation under laboratory conditions suggests a positive $\delta^{18}O$ shift in the water (Taylor and others 1984). In field applications, the oxygen shift is expected to be influenced further by the presence of carbonates and silicates. The magnitude of the shift would be directly related to the oxidation rate and the residence time (and indirectly to the permeability).

In ARD, the isotopic values of the liquid phase carry two types of information: (1) the isotopic signature of the original water components (groundwater, precipitation, ice water, etc.; Fig. 3), and (2) the isotopic signature of oxidation with respect to the presence of carbonates (and or silicates) involved in the overall chemical process. ARD effluent is commonly derived from a mixture of several types of waters that react, before or after mixing, with the rock (Fig. 4).

The isotopic values for each water type or rock can be defined by a geochemical vector that describes the water in terms of its isotopic composition. In the case of δD and $\delta^{18}O$, this vector is defined in a two-dimensional space (plotting δD vs $\delta^{18}O$). Mixing effects can be mathematically simulated by the addition of two vector compositions. In contrast, water/rock reaction can be simulated by scalar product of two vector compositions.

If one defines the isotope vector compositions, [S], [P], [G], [R], and [ARD] for surface waters, precipitation,

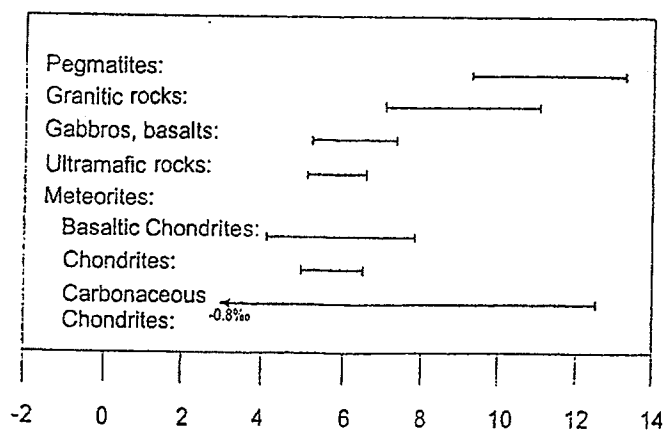


Fig. 2
 Range of $\delta^{18}O$ values of igneous and stony meteorites (after Jager and Hunziker 1979)

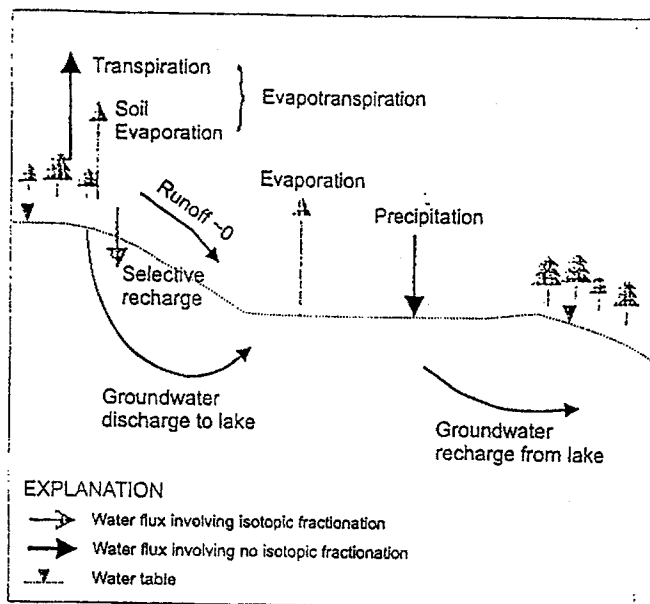


Fig. 3 Schematic diagram of the hydrological components of a groundwater - lake system

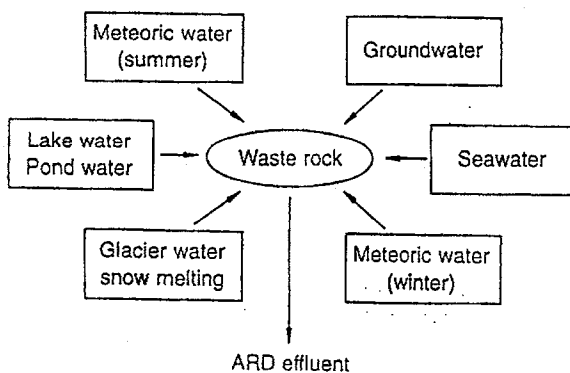


Fig. 4 Water types that may be at the origin of ARD effluent

groundwater, rock and ARD effluent respectively, then isotope information can be obtained from the following equation:

$$[ARD] = \{s[S] + p[P] + g[G]\} \cdot \{r[R]\} \quad (5)$$

In Eq. 5, *s*, *p*, *g* and *r* are the coefficients that quantify the relative proportions of the various isotopic species. Eq. 5 generates two linear equations (one that describes the $\delta^{18}O$ and the other δ^2H) with many unknowns. Therefore, it is necessary to generate additional equations, based on the major ion concentrations, to solve the equation.

In most geochemical situations, the distribution of major ions provides a sufficient number of equations to solve a mixing model without needing the additional two equations obtained from the isotope data. However, in ARD studies, the elemental concentrations of non-ARD me-

teoric waters are commonly very close to each other (close to zero). Thus, there are high analytical errors and insufficient resolution for solving a mixing model using strictly the equations for major ions. It may be instrumental to use the additional isotope equations because the isotopic values of non-ARD (low salinity) waters are significantly different, and thus, can provide meaningful information for the low-salinity waters. The importance of elemental concentrations is only related to the ARD effluent itself and to other high-salinity waters such as sea water. Non-ARD waters may consist of different water types interacting with rock (Fig. 4) before mixing with ARD effluent.

The isotopic values of different water types can be plotted on the same δD vs $\delta^{18}O$ graph, and simple processes (bipolar mixing, evaporation, and water/rock reaction) can be defined as linear trends (Fig. 5).

Water classification based on isotopic composition

Different classes of non-ARD waters include: (1) meteoric waters, (2) seawater, (3) glacier waters, and (4) waters from lakes, basins and ponds (Fig. 4). In some areas, geothermal and mineral waters may also be present. (Geothermal and mineral waters are distinguished from other non-ARD waters by their high elemental concentrations.)

Meteoric waters

$\delta^{18}O$ and δD values of meteoric waters usually fall on a line defined by the general equation:

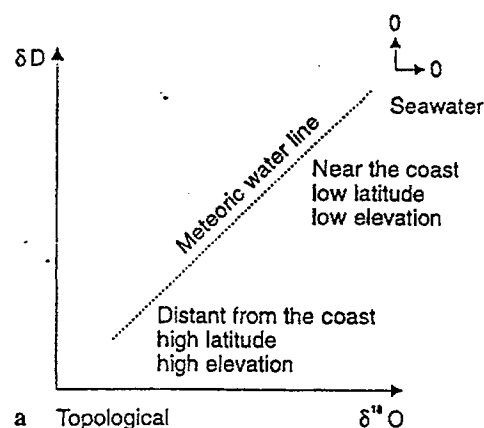
$$\delta D = \delta^{18}O + 10 \quad (6)$$

Different types of meteoric waters and their seasonal variations fall on the global meteoric line (Eq. 6). However, it is advisable to redefine this line more precisely for the area of investigation, because some portions of the area may be represented by "local meteoric lines", which differ slightly from Eq. 6 (e.g. Anati and Gat 1989; Clark and others 1982; Downing and others 1987).

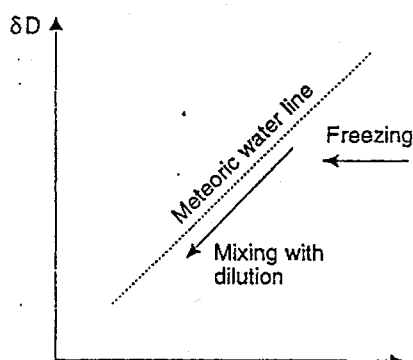
In general, precipitation and surface runoff demonstrate large seasonal variations, while groundwaters demonstrate a much smaller variation (Fig. 6). The high temporal variations of the isotopic values of precipitation mask their spatial variations. For this reason, the geographic variations of isotope values (Fig. 5) can only be assessed for groundwaters. In order to further filter out the temporal variations, deep sampling of aquifers should be undertaken (e.g. Ghomshei and Clark 1993). Spatial variations of isotopes in particular geographic regions are commonly related to (1) latitude (2) distance from the origin of the cloud (i.e. the sea), and (3) altitude (Fig. 5a).

Seawater

In coastal areas, seawater can be one of the water types that contributes to the mineral industry effluent. Seawater-



a Topological parameters



b Mixing freezing

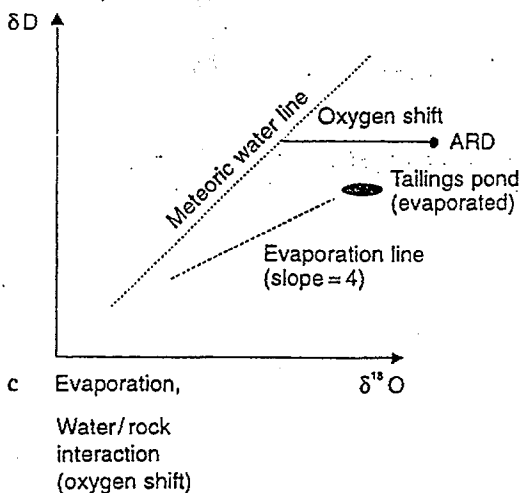


Fig. 5
Qualitative variations of δD and $\delta^{18}O$ in different environmental processes (after Ghomshei and Clark 1993)

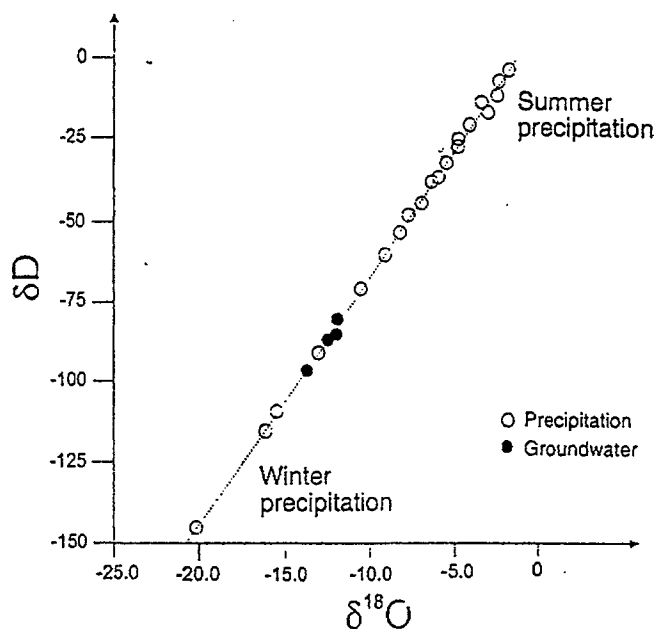


Fig. 6
Schematic graph showing hypothetical compositions of precipitation (wide range) and groundwater (narrow range)

teoric waters are negative. On the δD - $\delta^{18}O$ plot, seawater is commonly the heavy end-member of the local meteoric line (i.e. Craig 1966; Jager and Hunziker 1979; Fig. 5a).

Glacier waters

Glacier waters are distinguished from meteoric waters by their pronounced negative isotopic values. The isotopic composition of glaciers depends primarily on the temperature and elevation at the time of deposition. Systematic measurements of δD and $\delta^{18}O$ have been used therefore to study the flow patterns of glaciers, accumulation rates, and inherent climate variations (e.g. Hartmann 1994; Jager and Hunziker 1979).

Water in lakes, basins and ponds

Lakes and basins are characterized by their evaporation and recharge rates. Closed basins with high evaporation rates are enriched in heavy isotopes. Natural and artificial ponds (e.g. tailings ponds) are also enriched in heavy isotopes, particularly during the high evaporation/low precipitation seasons. Evaporation of the ponds is defined by $\delta D/\delta^{18}O$ slopes between 4 and 5 depending on the temperature (e.g. Craig 1966; Ghomshei and Clark 1993; Fig. 5c). Therefore, oxygen and hydrogen isotopes can be very useful in tracing the seepage from tailings ponds.

ARD effluent

ARD effluent, which is derived from reaction of non-ARD waters with rock, is expected to have the same δD value as the original meteoric waters. This is because rocks commonly lack significant amounts of hydrogen. Furthermore, oxidation processes are not expected to affect significantly the hydrogen isotopes in water (however, hy-

er has a clear chemical signature and is characterized by a high salinity and heavy isotopic signature. The high salinity of the seawater may be similar to some ARD or geothermal waters. However, the isotopic signature of the seawater is distinct from other water types. For example, coastal waters are commonly lighter than seawater. Seawater has isotopic values close to zero while those of me-

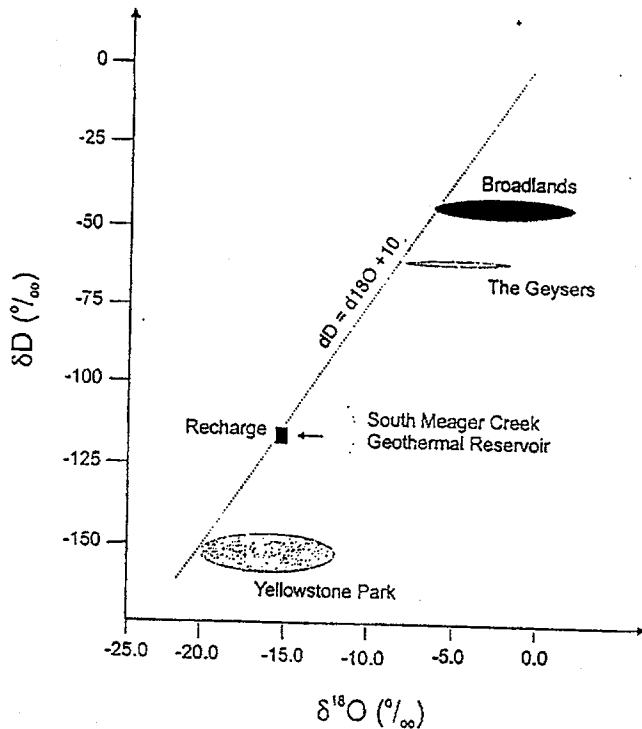


Fig. 7
 $\delta^{18}\text{O}$ and δD compositions of geothermal waters. Prominent oxygen shifts observed (after Trusdell and Hulston 1980; and Ghomshei and Clark 1993). Recharge composition determined by trajectory backtracing

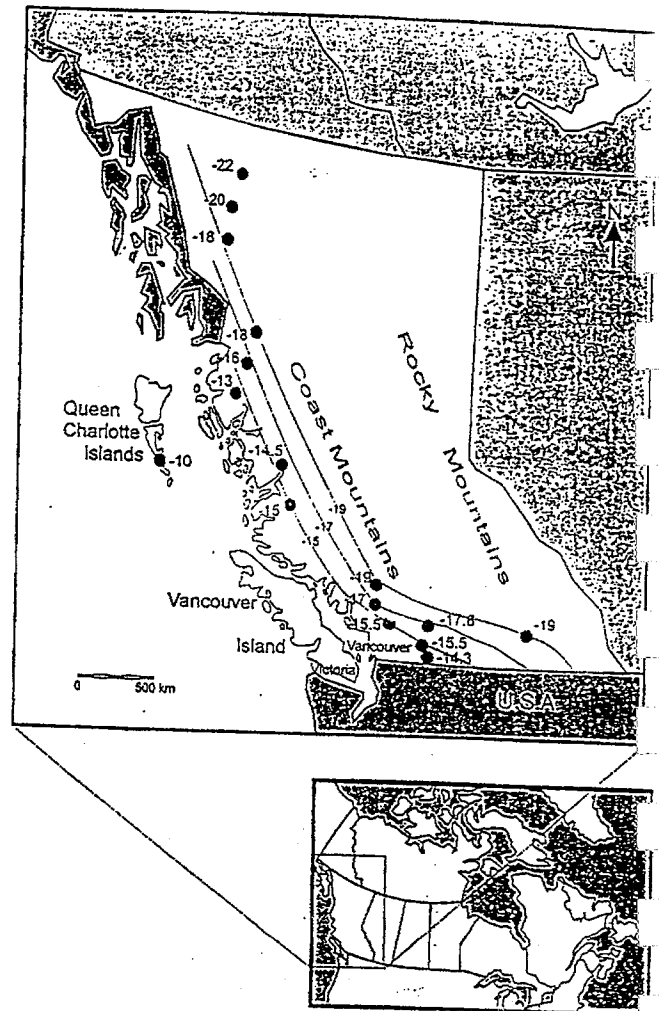


Fig. 8
 Mean $\delta^{18}\text{O}\text{‰}$ for modern meteoric waters in western British Columbia, Canada (after Ghomshei and Clark 1993)

drogen fractionation should be quantified by experimental and field studies). Therefore ARD waters are expected to show only an oxygen shift relative to the original meteoric waters (Taylor and others 1984). The oxygen shift (Fig. 5c) is well understood in the geothermal systems (Fig. 7), where water/rock interaction is controlled by temperature. In ARD, the interaction is controlled mainly by pH.

The isotopic values of the original meteoric waters can be simply obtained on the δD vs. $\delta^{18}\text{O}$ plot by connecting the average endpoint of the [ARD] isotope vector [characteristic of the site and defined by the coordinates ($\delta^{18}\text{O}$, δD)] to the meteoric line by a straight horizontal line (i.e. constant δD ; Fig. 7). This process can be referred to as trajectory backtracing. Identifying the source (or recharge) of the ARD effluent is commonly one of the most instrumental elements in ARD studies. The following concepts can be used in characterization of the original waters:

1. Seasonal variations of the ARD indicates that the original waters are mostly related to precipitation and surface run off. In this case it is possible to identify the season in which the ARD is more active.
2. The absence of large seasonal variations in ARD implies that the source waters are groundwaters, glacier water or seawater. As these waters have distinct isotopic values, the source can be easily identified.

3. The altitude of the original waters can be established in high-relief areas. The sensitivity of isotopes to the altitude has been established by many investigators (e.g. Clark and others 1982).
4. The latitude or the distance from the shore of the original waters (recharge) can be established in areas where original waters may have travelled long distances before reaching the ARD stage. The geographic variations of isotope values have been established by many investigators (e.g. Chitoshi and Minoru 1994). Ghomshei and Clark (1993) presented a contoured isotope map of the Canadian Pacific Coast (Fig. 8). The value of the oxygen shift itself carries convoluted information on: (1) the buffering capacity, (2) the rate of oxidation (and related pH), and (3) the permeability of the rock (affecting the residence time of the water). Knowledge of the permeability and the isotopic values of the participating carbonates will allow estimation of the rate of oxidation.

Field sampling and data analysis

An ARD isotope study should include a comprehensive water sampling program of different types of local waters (creeks, lakes, glacier waters, rain and snow, etc.). Each study should include the following:

1. Precipitation sampling undertaken during two seasons (summer and winter).
2. ARD effluent sampled at different locations (seeps) and also at different seasons, corresponding to collection times for precipitation data.
3. Water samples analyzed for major ions, δD and $\delta^{18}O$. A few samples (depending on the funds available) can also be analyzed for Tritium, in order to estimate ages of waters in the flow systems.
4. A few rock and mineral samples, especially the silicates and carbonates (depending on the availability of the funds and the complexity of the problem) analyzed for δD and $\delta^{18}O$.
5. Conductivity, pH, and temperature measured in the field. Exact sample locations and the altitude should be recorded.

Conclusion

Oxygen-18 and deuterium analysis can be successfully used in solving hydrological/hydrogeological problems similar to those related to mineral industry effluent (particularly ARD). The methodology inherent to stable isotope analysis and interpretation in hydrological and hydrogeological problems is straightforward and well documented. Analysis is based on the distinct isotopic signature of different sources contributing to the problem. Interpretation of isotope data is commonly quantitative and provides a reliable solution to the hydrological/hydrogeological problem.

Application of oxygen-18 and deuterium in ARD will contribute to the understanding of a variety of ARD (and other mineral industry effluent) problems including the source of the ARD effluent (recharge), seasonal-dependency of ARD, oxidation rate, and general hydrogeological problems (e.g. permeability).

In consideration of the importance of the information provided, a stable isotope study can be a cost-effective method for assessment of ARD problems, especially when the problem is dominantly hydrological. In the areas where isotope data on meteoric waters are readily available, additional isotopic study of the ARD effluent is highly recommended.

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