CASE STUDY: Helium and radon in the Fitzroy River, Western Australia

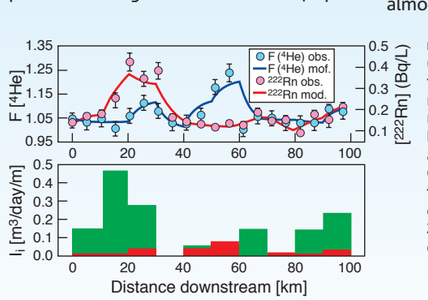
A study of groundwater discharge to a river in north-western Australia is one of the first of its kind to combine a number of tracers in a remote unregulated tropical river.

These systems are characterized by extremely high seasonal stage variations due to monsoonal rainfall and extended dry periods. Development of water resources in such regions relies to a large extent on groundwater. At the same time the entire source of water in river discharge at this time of the year is supported by groundwater discharge. Therefore understanding the sources and location of groundwater discharge to tropical rivers is important to simultaneously manage sustainable groundwater development and protection of river ecosystems.

Hydrograph methods in such remote, dynamic and low gradient systems present several logistical and practical difficulties. Estimating river discharge as a function of stage, the difficulties in installing and sampling piezometric networks and the very low hydraulic gradient in the groundwater systems make traditional methods impractical. Sampling of environmental tracers such as noble gases along in-stream transects are used because of the ability to distinguish and integrate groundwater processes on long time scales rather than simply a snapshot in time.

In this example the focus is on the combined use of the noble gases helium-4 and radon-222 in a tropical river. Helium in deep groundwater tends to be much higher compared with groundwater derived from shallow circulation due to the longer residence time and opportunity for helium accumulation. On the other hand, 'excess' radon-222 represents near field processes because its short half life means it only persists for about 3 weeks in the absence of groundwater input. Using helium-4 and radon-222 can be used in tandem to distinguish groundwater sources to rivers that are deep relative to near stream sources. This has implication for allocating groundwater protection zones for streams to protect dry season baseflow.

The example in Figure 4 is from the Fitzroy River of Western Australia. A transect of radon-222 and helium-4 concentrations during the dry season shows a series of peaks and troughs in concentration (Top panel). The first point to note is that even the baseline concentration for both helium and radon are above that which would be expected from equilibrium with the atmosphere or support from radioactive decay within the stream sediments. This is strong evidence that there is groundwater input along almost the entire 100 km length of the river. The peaks in radon and helium represent zones of higher groundwater discharge with the former indicative of all locations of groundwater input and the latter represent only deep groundwater sources. An analytical model using estimates of groundwater radon and helium concentrations and groundwater fluxes to fit the data can then be used to extract groundwater discharge along specific reaches of the entire transect (bottom panel). While the flux from deep groundwater sources is relatively small compared with total groundwater input from local or phreatic groundwater, for the middle portion of the river it represents almost the entire baseflow contribution.



LEFT **Figure 4. Top panel: Helium-4 (expressed as mixing fraction compared to solubility equilibrium with air) and radon-222 along a transect of the Fitzroy river, WA. Also shown are model values based on groundwater concentrations and groundwater inflow. Bottom panel: Open bars represent total groundwater input and closed bars representing deep groundwater input (from Gardner *et al.*, 2011, Water Resour. Res., Vol 47, W06523, doi:10.1029/2010WR010276).**

FURTHER READING AND LINKS

IAEA (2013): Isotope Methods for Dating Old Groundwater (available at: http://www-pub.iaea.org/books/IAEABooks/8880/Isotope-Methods-for-Dating-Old-Groundwater).

Solomon, DK, 2000. Helium-4 in groundwater. In Ch. 14, PG Cook and AL Herczeg [eds] Environmental Tracers in Subsurface Hydrology. Kluwer Academic Press.

Stute M and Schlosser P, 2000. Atmospheric noble gases In: Ch. 11. PG Cook and AL Herczeg [eds] Environmental Tracers in Subsurface Hydrology. Kluwer Academic Press.

Torgersen T and Stute M, 2013, Helium (and other Noble Gases) as a Tool for Understanding Long Timescale Groundwater Transport in Ch.8. A. Suckow, P. K. Aggarwal and L. J. Araguas-Araguas [eds] Isotope Methods for Dating Old Groundwater. International Atomic Energy Agency

Noble Gas Laboratories:

CSIRO: http://www.clw.csiro.au/services/isotope/

Lamont-Doherty Earth Observatory: http://www.ldeo.columbia.edu/environmental-tracer-group

University of Utah: http://www.earth.utah.edu/noble-gasses/index.php

CONTACT US
t 1300 363 400
+61 3 9545 2176
e enquiries@csiro.au
w www.csiro.au

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FOR FURTHER INFORMATION
Megan LeFournour
Isotope Analysis Service
CSIRO Land and Water
t +61 8 8303 8747
e megan.lefournour@csiro.au
w www.csiro.au

Helium–4 and noble gases as groundwater tracers

Helium is produced naturally in aquifers as a by-product of decay of uranium and thorium that occur in minerals. As the lightest of the noble gases, it is unreactive in the environment. Because it accumulates in groundwater over time, helium-4 concentrations in groundwater vary over several orders of magnitude.

It is a very useful tracer to assess groundwater on time scales from several centuries to millions of years and to study water movement through low permeable layers. The other noble gases (Ne, Ar, Kr, Xe) occur in groundwater through equilibration with the soil atmosphere. Their concentrations may be used to establish the mechanism of recharge and also to estimate parameters such as recharge temperature, salinity and altitude.

Background

The noble gases are the elements helium, neon, argon, krypton, xenon and radon. Because they form no compounds with other elements, their inert nature makes them excellent candidates as groundwater tracers. There are fundamentally two ways in which noble gases are used:

1. Helium-4 is produced by decay of uranium and thorium in aquifer minerals and accumulates in groundwater over time. Its concentration in "old" groundwater is proportional to the contact time of groundwater with the aquifer matrix and the uranium and thorium concentration in the aquifer material.
2. The atmospheric noble gases (Ne, Ar, Kr, Xe) dissolve in water in the unsaturated zone according to their solubility. This solubility is dependent on recharge temperature and salinity. Therefore they are most frequently used to estimate recharge temperatures that can be used to confirm the existence of fossil groundwater recharged under different climatic conditions. A second component of noble gas concentration in groundwater is a super-saturation that occurs when groundwater level moves up and down in response to recharge events. This is often termed "excess air" and can also be used to understand episodic recharge processes.

How can we use He-4 to constrain time scales in a groundwater flow system?

In simple terms the concentration of ^4He in groundwater is directly proportional to its contact time with the aquifer matrix because helium builds up over time due to release by decay of uranium and thorium in aquifer minerals. To use ^4He in groundwater one needs to have some estimate of the production rate, which in the simplest approximation is assumed to be constant over time as it traverses through the aquifer. The release of helium into the groundwater is determined by the amount of U and Th in aquifer minerals, as well as grain size and porosity or fracture connectivity. It is also assumed that all the ^4He that is released is retained by the water. There is no general upper limit of helium concentration in groundwater, so in contrast to radioactive tracers there is no upper end for its time scale. Helium shows a linear behavior in mixtures, which makes the application easier than for radioactive tracers that can be "contaminated" by small admixtures of young water. Instead of trying to directly estimate the production rate, it may be calibrated against another radioactive tracer such as radiocarbon or ^{36}Cl. Then helium can extend the time frame beyond that possible with conventional radioactive tracers.

What do noble gases tell us about groundwater?

The atmospheric noble gases Ne, Ar, Kr and Xe all partially dissolve in water and their concentrations in groundwater are a record of the temperature and salinity conditions at the time of recharge. Because each of the noble gases has different solubility-temperature relationships it can reveal two major attributes. The first is the temperature at the time of recharge, which approximates the mean annual temperature. Studies in many parts of the world have generally found that recharge temperatures were several degrees cooler than that of the more recent ten thousand years. This confirms inferences made using stable isotope data that fossil groundwater recharged during more temperate climate regimes about 10,000–15,000 years ago were common in many arid and semi-arid parts of the world. The implication for groundwater sustainability is that these are no longer being replenished in the current climate regime. Similar data are not yet readily available in Australian groundwater systems, though similar trends are likely.

A second observation is that the absolute noble gas concentrations in groundwater are often higher than that possible from simple passive equilibration with the soil atmosphere. This so-called "excess air" arises by processes at or near the water table that essentially pump air into the water by floods or rises and falls in the water table. This "excess air" component

CSIRO's Isotope Analysis Service supports the research projects of CSIRO and provides an effective and readily-accessible service for hydrology and environmental communities, including Government and private sectors, to obtain measurements of environmental isotopes and trace gases. The service undertakes approximately 3000 analyses each year for Australian and international clients. The results of these analyses guide informed natural resource management decisions.

is thought to be proportional to the speed of water table rise that in turn can be used to estimate recharge rates.

Sampling procedure

Helium and noble gases are sampled in one of two ways (Figure 1). The easiest option is deployment of diffusion cells into the well and left for at least 24 hours to equilibrate in the well screen or spring. The dissolved gases diffuse through a semi-permeable membrane and the diffusion cells are retrieved and sealed for return to the laboratory. The advantage of using this method is the relative simplicity of the sampling procedure and greater convenience for subsequent laboratory analysis. In some circumstances deployment of diffusion cells is not possible. Instead water samples are collected and returned to the laboratory in gas-tight copper tubes. This sampling technique is also straight forward, taking less than half an hour, but the laboratory gas extraction and purification is a bit more cumbersome and therefore more expensive than the gas diffusion method.

Also at present only He and Ne can be measured reliably from copper tubes.

Applications

The use of helium and noble gases has been around since the 1960s, though its application to hydrological problems in Australia has been restricted by lack of local analytical capability. The first application using helium measurements in groundwater was as a tool in mineral exploration for identifying possible occurrences of hidden uranium deposits.

HELIUM-4

Perhaps one the of most well known studies of helium in groundwater is of the Great Artesian Basin which covers about one fifth of Australia and was initiated for the purpose of understanding the deep circulation and fluid transport processes. The example from a study done in the early 1980s (Figure 2) shows the relationship between helium-4 concentration and groundwater 'age'. The data show a steady increase in helium concentration up to about 400,000 years, where this age in Figure 2 was estimated up to 50,000 years from radiocarbon and over the whole age range consistent with estimates from Darcy's law. The rate of helium emanation up to 50,000y was consistent with the U and Th content in the aquifer material. The very large increase in ^{4}He concentration is still subject to some conjecture; one interpretation is that there is an external source of helium in the interior part of the basin, possibly from crustal degassing. Other interpretations are that it is from passive accumulation from local sources. In any case, this highlights some of the potential insights from helium-4 distributions from large and slow-moving groundwater systems.

Subsequent studies in this and other large groundwater basins have included other long-lived tracers such as ^{36}Cl and ^{81}Kr and these are consistent with the ^{4}He results. This highlights the benefit gained in improving the reliability of system understanding through confirmation with two or more different tracers. The main advantage of ^{4}He is that it is relatively simple to implement and inexpensive compared with ^{14}C, ^{36}Cl or ^{81}Kr and can be extended over a much longer timeframe.

NOBLE GASES

The application of noble gases to the study of groundwater now extends to several places throughout the world. Often these studies look at changes to the hydrological balances due to variations in climate during the last 40,000 years. An example from the Stampriet aquifer of southern Africa, a large sedimentary aquifer similar in climate and vegetation to parts of central Australia, is shown in Figure 3 below.

The noble gas temperature, which is an estimate of recharge temperature is shown to average ~26.5 °C over the past 10,000 years. Groundwater older than about 15,000 years is ~5.3 °C cooler reflecting the more temperate climate in this region during the last glacial maximum period. The benefit of using noble gases in this example is that the δ^{18}O record reflects changes in moisture sources over the same time period rather than temperature changes, as is often observed in continental aquifer systems in low and mid-latitude regions. Some very high fractions of excess air (reflected by ΔNe up to 200%) are indicative of rapid fluctuations in the water table in the recent past indicating a transition between dryer and wetter climate regimes. The change in excess air here occurs later than the change in temperature.

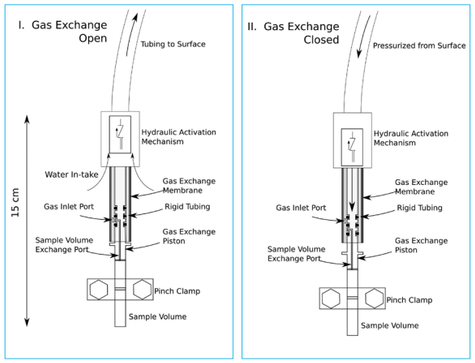
Advantages and limitations

The noble gases are fundamentally inert and form no compounds with other elements. Once their signal is imparted at the time of recharge, it is not modified over time during transit within the aquifer.

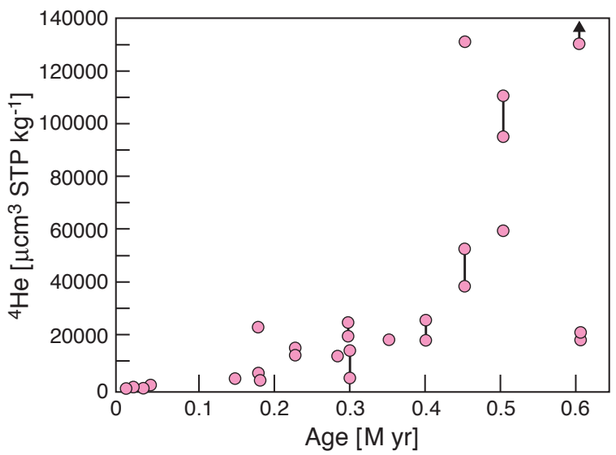
For helium-4 to be used as a quantitative estimate of flow rates and time, the rate of production of helium-4 needs to be estimated or measured. There are a number of ways to do this, such as comparison with other tracers (^{14}C, ^{36}Cl) or laboratory helium release experiments in sealed stainless steel canisters. The disadvantage of the latter method is that it is time consuming and requires access to aquifer solid material.

External sources of helium-4 such as fluid transport from deeper basins can disturb the simple relationship of helium concentration versus time. Also local differences in uranium and thorium concentration in aquifer rocks can change the accumulation rates of helium.

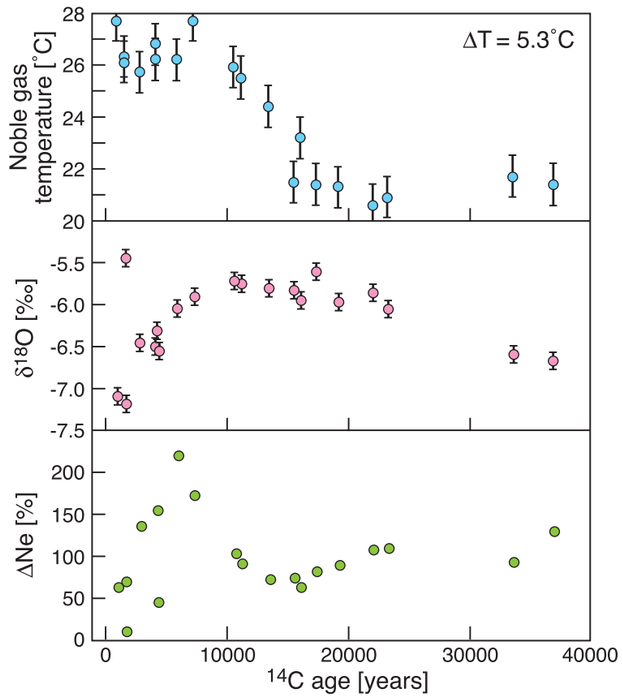
One of the limitations of the method is that relatively few laboratories are capable of analyzing the full suite of noble gases. At this stage CSIRO can measure ^{4}He, Ne and Ar but not the heavy noble gases Kr and Xe. A new facility to measure all noble gases is under construction.



LEFT **Figure 1. The left panel shows a diffusion cell sampler and the right panel shows filling of a copper tube at a well head for noble gas sampling.**



ABOVE **Figure 2. Relationship between ^{4}He concentration and age for the Great Artesian Basin (Torgersen and Clark, 1985, Geochim. Cosmochim Acta, 49, 1211-1218.). In the older samples very high ^{4}He concentrations are thought to be due to crustal degassing.**



ABOVE **Figure 3. Noble gas temperature, δ^{18}O and ΔNe% (excess air) in groundwater from the Namibian portion of the Stampriet aquifer (modified from Stute and Talma, 1998, Isotope techniques to study past and current environmental change in the hydrosphere and atmosphere, 307-318, IAEA, Vienna).**

FAQs

Q: How are samples collected?

A: CSIRO's isotope analysis laboratory will supply pinch-off clamps and either diffusion cells or copper tube for sampling of gas or water respectively. It is important to contact the laboratory to discuss the circumstances and project particulars to get the appropriate sampling gear. If copper tubes are used, it is critical to use a submersible pump and to maintain high water pressure during sampling to ensure no bubble formation occurs, which alone can make results worthless. The groundwater being sampled must not have any contact with air. See http://www.clw.csiro.au/services/isotope/ for more detailed sampling instructions.

Q: How soon can I expect results?

A: The CSIRO laboratory endeavours to have results within 6 weeks. However, as the only laboratory in Australia capable of analysing noble gases in groundwater samples, a backlog of samples may occur, delaying your results. Please contact us if turnaround time is an issue for your project.

Q: What can I do if the sample is contaminated?

A: Contamination usually is the result of entrainment and partial dissolution of air bubbles into copper tubes at the time of sample collection. If small this is indiscernible from excess air as a signal in groundwater. For large bubbles enclosed in the copper tube the results cannot be interpreted. There are no anthropogenic contaminants of the noble gases to be concerned about.

Q: How much does it cost?

A: For actual price information see **http://www.clw.csiro.au/services/isotope/** Usually a loan fee is charged for the sampling devices and reimbursed if the equipment is returned unused.