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Carbon-14 as a tracer of groundwater discharge to streams

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The provenance of groundwater discharge to a stream can be determined by measuring the response of multiple groundwater age tracers within the stream across the discharge zone. The sampling interval required to detect groundwater discharge is limited by the rate of equilibration with the atmosphere downstream of the discharge zone, which is determined by the gas transfer velocity. Carbon-14 (^{14}C) equilibration is driven by CO_2 exchange, which is a small component of the dissolved inorganic carbon in most stream systems, and therefore the rate of equilibration is slower than for other gaseous age tracers. In this paper we use a step-wise approach to develop and demonstrate the use of ^{14}C as a tracer in streams receiving groundwater discharge.

Excess carbon dioxide (CO_2) in the emerging groundwater degasses until equilibrium with atmospheric CO_2 is reached; increasing pH and enriching the residual ^{14}C by fractionation. In addition, the ^{14}C gradient between groundwater and the atmosphere drives a slower process of isotopic equilibration. We have measured the rates of this chemical and isotopic equilibration experimentally by exposing 250 L of old groundwater to the atmosphere in an evaporation pan. Chemical equilibrium was achieved within 2 days, during which the ^{14}C increased from 6 to 16 pMC. The influence of fractionation during the initial CO_2 degassing on isotopic equilibrium rates was negligible. Isotopic equilibrium took over 2 months, with ^{14}C in the evaporation pan increasing to 108 pMC over 71 days. This increase in ^{14}C was simulated using a mass balance model with an effective ^{14}C gas transfer velocity of 0.013 m d^{-1}.

Field testing of the method was conducted at two sites. Firstly, we measured the evolution of ^{14}C in dewatering discharge as it flows along an ephemeral creek channel in the Pilbara, Western Australia. Measured ^{14}C increased from 11 to 31 pMC along the 10km reach, which corresponds to a travel time of about 2 days. The measured increase was simulated using a mass balance model with an effective ^{14}C gas transfer velocity of 0.025 m d^{-1}. Secondly, we measured ^{14}C at six sites along the Daly River, Northern Territory. Here we observed a decrease of 7 pMC across a major groundwater discharge zone and were able to simulate the observed ^{14}C using a gas transfer velocity of 0.14 m d^{-1}. We quantify the total groundwater influx using measurements of stream discharge and radon-222, which allows us to estimate the ^{14}C activity of the groundwater discharge at 64 pMC.

The potential for groundwater discharge to be detectable in other streams using ^{14}C depends on the magnitudes of groundwater discharge, dissolved inorganic carbon, and ^{14}C relative to stream values.