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Vertical carbon-14 profiles for resolving spatial variability in recharge in arid environments

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SUMMARY

Groundwater age tracers are often measured to help constrain estimates of groundwater recharge, especially in arid environments where other methods are unsuitable. However multiple processes can influence the shape of vertical tracer profiles in an aquifer including (1) variation in tracer input concentrations from the unsaturated zone, (2) the role of diffusion in transporting tracer into the aquifer when fluxes are low and (3) spatial variability in recharge. This study demonstrates the influence of spatially variable recharge and spatially variable carbon-14 (^{14}C) activities in the unsaturated zone on vertical ^{14}C profiles in groundwater. Through groundwater flow and solute transport modelling, we demonstrate that recharge estimated from single point measurements of ^{14}C may be wrong more than an order of magnitude when unsaturated zone ^{14}C activities and recharge vary spatially. We then present a case study from the Ti Tree Basin in arid central Australia, where detailed profiles of ^{14}C activity in unsaturated zone gas and groundwater have been measured, and spatial variability in unsaturated zone ^{14}C is observed (ranging from 54 to 106 pMC above the watertable). Through modelling our data, we show that when unsaturated zone ^{14}C activities are known, measurement of the ^{14}C profile can help constrain estimates of recharge and its spatial variability. This approach improves our understanding of groundwater flow in the Ti Tree Basin, by showing mountain front recharge to be an important mechanism.

1. Introduction

Environmental tracers have been widely used to estimate groundwater age and residence time, and can potentially provide more accurate estimates of recharge and flow velocity than traditional hydraulic methods (Cook and Bohlke, 2000). Vertical profiles of groundwater age can be especially useful as indicators of spatial variability in groundwater recharge. For example, Robertson and Cherry (1989) measured vertical profiles of groundwater age using tritium in an aquifer in Ontario. They found that the depth at which 'bomb-peak' tritium was measured in the aquifer varied spatially, and used this to infer spatial variability in groundwater recharge.

In arid environments low recharge rates lead to longer residence times and tracers that can provide estimates of age over longer time scales are needed. Carbon 14 (^{14}C) dating of total dissolved inorganic carbon (TDIC) in groundwater can provide estimates of age over time scales of $\sim 10^2$–10^4 years, making ^{14}C useful for estimating recharge in arid zones (Herczeg and Leaney, 2011). For example Harrington et al. (2002) measured ^{14}C activities in groundwater in the Ti Tree Basin in arid central Australia. Harrington et al. (2002) used an analytical approach to demonstrate that recharge varied spatially, with many of the higher recharge rates lying on flowlines originating near ephemeral surface water features that are subject to flooding after rare, high intensity rainfall events. Carbon 14 may additionally be used to help calibrate groundwater flow models in arid and semi-arid environments (Sanford, 2011).

Many factors can complicate the interpretation of ^{14}C in aquifers. For example, carbonate mineral weathering can dilute the ^{14}C activity of groundwater compared to that of the atmosphere (Fontes and Garnier, 1979; Gillon et al., 2009). In low recharge environments, diffusion and dispersion can significantly enhance transport of ^{14}C in the aquifer, and this may influence the assessment of groundwater fluxes (Walker and Cook, 1991; Castro and Goblett, 2005). Production of 'old' (low ^{14}C) CO_2 from oxidation of organic matter or calcite precipitation in the unsaturated zone may significantly reduce the ^{14}C activity of soil CO_2 (Keller and

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Bacon, 1998; Walvoord et al., 2005; Gillon et al., 2009), which influences the ^{14}C activity of infiltrating groundwater. This may lead to misinterpretation of ^{14}C activities when estimating groundwater age (Bacon and Keller, 1998). Wood et al. (2014) showed how variation in watertable depth may lead to spatial variation in ^{14}C activities above the watertable (i.e. ^{14}C activity that recharging water would equilibrate with) when a source of old CO_2 is present in the unsaturated zone. In this regard estimates of groundwater age from ^{14}C may differ significantly from estimates provided by other methods (e.g. estimates of age provided by other environmental tracers or advective ages determined by groundwater modelling).

While previous studies have considered these processes (diffusion, dispersion, production of old CO_2 in the unsaturated zone) and their influence on ^{14}C activities in groundwater separately ((Walker and Cook, 1991; Castro and Goblett, 2005; Bacon and Keller, 1998), their potential combined effects, along with spatial variability in recharge on groundwater age profiles have not been assessed. The aim of this study is to investigate the influence of spatially variable ^{14}C activities at the watertable and spatially variable recharge on vertical ^{14}C profiles in an unconfined aquifer, firstly in a theoretical study, then secondly using a field example. We focus on low recharge environments where diffusion and dispersion are also significant. Through theoretical modelling we show how estimated ^{14}C age profiles under different recharge and ^{14}C input (^{14}C activity at the watertable) scenarios compare with those determined from particle tracking (i.e. advective ages, which are not influenced by diffusion and dispersion). We then use measured ^{14}C profiles from an unconfined aquifer in arid central Australia (Ti Tree Basin), and measurements of unsaturated zone ^{14}C activity, along with groundwater flow and solute transport modelling to inform the likely magnitude and spatial variability of recharge along three inferred flow lines.

2. Theoretical modelling

2.1. Methods

To evaluate the influence of spatial variability in recharge and ^{14}C input on ^{14}C derived age profiles, we present four scenarios using a 2D transect model constructed using the MODFLOW-2005 code. MODFLOW is a widely used code which solves the partial differential groundwater flow equation using a finite differencing approach (Harbaugh, 2005). The model represents an unconfined aquifer 50 km long and 65 m deep based approximately upon the length and thickness of the unconfined aquifer in the Ti Tree Basin, Northern Territory, Australia. The domain is discretised into cells approximately 250 m long and 0.5 m thick. The left hand boundary and bottom boundary are no-flow boundaries, while the right-hand boundary is a constant head boundary set to maintain saturated thickness at 65 m. Relevant model parameters (hydraulic conductivity, porosity) are listed in Table 1. Recharge rates used for individual scenarios are discussed later.

Table 1
Model parameters used in simulations where * are based on values in Gelhar et al. (1992) for the aquifer scale modelled; ** are from Cook and Herczeg (2000); *** is from Li and Gregory (1974).

Parameter	Value
Horizontal hydraulic conductivity ($K_{x,y}$)	100 m d^{-1}
Vertical hydraulic conductivity (K_z)	10 m d^{-1}
Porosity	0.2
Longitudinal dispersivity*	100 m
Transverse dispersivity*	10 m
^{14}C decay constant**	1.21×10^{-4} y^{-1}
^{14}C diffusion coefficient in water***	3.15×10^{-2} m^2 y^{-1}

Carbon 14 transport was simulated using MT3D, which couples the groundwater flow model with the advection–dispersion/diffusion equation to simulate solute transport (Zheng, 2010). The unsaturated zone was not modelled in this exercise. Rather diffusion of ^{14}C into the aquifer from the unsaturated zone is considered by assigning a concentration boundary at the watertable (which varies spatially in scenarios three and four). Parameters for diffusion, dispersion and radioactive decay are given in Table 1. We ignore the potential influence of geochemical reactions on ^{14}C dilution in order to assess the influence of ^{14}C input activity and spatial variability in recharge in isolation of any other complicating factors (eg. carbonate weathering).

Advective ages were modelled using the post processing particle tracking code MODPATH (Pollock, 2012), with particles being assigned to the cells where recharge is applied. This method of modelling groundwater age ignores the role of diffusion and dispersion which the solute transport model takes into account. The particles were ‘forward tracked’ so that the time taken to reach a particular cell of interest (i.e. a cell where age is calculated from ^{14}C concentrations) could be determined. These ages were compared with apparent ^{14}C ages, calculated from simulated ^{14}C activities using the equation:

$$t = \frac{1}{\lambda} \cdot ln\left(\frac{A_0}{A}\right) \quad (1)$$

where t represents age (time since recharge in years), A_0 is the initial activity (100 pMC for this exercise), A is the activity simulated in a particular model cell and λ is the decay constant for ^{14}C (Table 1).

Recharge rates are often estimated from measurements of groundwater age using simplified analytical solutions (Cook and Bohlke, 2000). Vogel (1967) presents such a solution, which describes the vertical age profile in an unconfined aquifer of constant thickness and porosity for a given recharge rate. This method can be used to estimate recharge in unconfined aquifers via:

$$R = \frac{H\theta}{t} ln\left(\frac{H}{H-z}\right) \quad (2)$$

where R is the average recharge rate over the interval between where the groundwater was recharged and the sampling location, H is the aquifer thickness and z is the depth at which groundwater age (t) is measured. Here we compare our modelled recharge rates with those that would be calculated based on application of Eq. (2) to individual points (discrete depths).

Four scenarios were considered in the modelling. In Scenario 1 a recharge rate of 1 mm y^{-1} was applied across the entire top of the model domain, representing low diffuse recharge in an arid environment. The ^{14}C input activity is 100 pMC (approximating the atmospheric value). Scenario 2 considers all the recharge to be focused in a small area at the start of the transect, representing preferential recharge or ‘mountain front recharge’ at an aquifer margin. Mountain front recharge may be a significant source of recharge in arid and semi-arid climates. It can occur if a basin has an uplifted boundary (eg. a mountain), and large rainfall events in the uplifted area generate runoff that can lead to focused recharge at the mountain front (the basin margin) (Wilson and Guan, 2004). The ^{14}C input activity was again 100 pMC across the whole model domain.

Focused recharge at the aquifer margin was applied in Scenario 3 but in this case the ^{14}C activity at the watertable varies spatially from 50 pMC to 100 pMC. Wood et al. (2014) measured unsaturated zone ^{14}C activities at multiple sites in the Ti Tree Basin in central Australia, and found consistent trends in ^{14}C dilution at most sites, with variation in unsaturated zone thickness leading to variation in ^{14}C activities at the watertable (lower ^{14}C when the unsaturated zone is thicker, data is presented later in Section 3.2). The transition