Supplement: Additional description of methods

Estimating mean transit times using lumped parameter models.

In the lumped parameter models the $^3$H activity in water at the catchment outflow at time $t$ ($C_o(t)$) is related to the input of $^3$H over time ($C_i$) via the convolution integral:

$$C_o(t) = \int_0^\infty C_i(t-\tau) g(\tau) e^{-\lambda \tau} d\tau$$

(S1).

In Eq. (S1), $\tau$ is the transit time, $t-\tau$ is the time when water entered the catchment, $\lambda$ is the decay constant (0.0563 yr$^{-1}$ for $^3$H), and $g(\tau)$ is the response function that describes the distribution of flow paths and tracer concentrations in the aquifer.

Several lumped parameter models implemented in the TracerLPM Excel workbook (Jurgens et al., 2012) were used in this study. The exponential flow model describes the mean transit time in homogeneous unconfined aquifers of constant thickness that receive uniform recharge and where flow paths from the entire aquifer thickness discharge to the stream. The piston flow model assumes linear flow with no mixing within the aquifer, such that all water discharging to the stream at any one time has the same transit time. The exponential-piston flow model describes mean transit times in aquifers that have regions where flow paths have an exponential distribution and regions where flow paths have a linear distribution. For the exponential-piston flow model $g(\tau)$ in Eq. (1) is given by:

$$g(\tau) = 0$$

for $\tau < \tau_m(1-f)$  \hspace{1cm} (S2a)

$$g(\tau) = (f\tau_m)^{1/1-f^{n+1/1-f-1}}$$

for $\tau > \tau_m (1-f)$  \hspace{1cm} (S2b)

where $\tau_m$ is the mean transit time and $f$ is the proportion of the aquifer volume that exhibits exponential flow. Where $f = 1$, Eqs (S1 and S2) describe the distribution of transit times resulting from exponential flow, whereas where $f = 0$, Eqs (S1 and S2) describe the distribution of transit times resulting from piston flow. TracerLPM specifies the ratio of exponential to piston flow as the EPM ratio.
(equivalent to $1/f - 1$). For an EPM ratio of 0, the model is equivalent to the exponential flow model, whereas EPM ratios $>5$ produces a model that is close to piston flow.

The dispersion model is based on the one-dimensional advection-dispersion transport in a semi-infinite medium. The response function for this model is:

$$g(\tau) = \frac{1}{\tau \sqrt{4\pi D_P \tau/\tau_m}} e^{-\left(\frac{(1-\tau/\tau_m)^2}{4D_P \tau/\tau_m}\right)}$$ \hspace{1cm} (S3),

where $D_P$ is the dispersion parameter (unitless), which is the inverse of the more commonly reported Peclet Number. $D_P = D/(v \times x)$, where $v$ is velocity (m day$^{-1}$), $x$ is distance (m), and $D$ is the dispersion coefficient (m$^2$ day$^{-1}$). While the dispersion model is considered to be a less realistic conceptualisation of flow systems, it commonly reproduces the observed distribution of radioisotopes within aquifers (Maloszewski, 2000).

**FEFLOW model.**

The FEFLOW model domain was 10,000 m in the longitudinal ($x$) direction and 25 m in the vertical ($z$) direction (Fig. S1). It was discretised with $10^6$ quadrilateral elements ($\Delta x = \Delta z = 0.5$ m). The initial homogenous model had a uniform porosity of 0.3 a uniform hydraulic conductivity, $K$, of 1 m day$^{-1}$.

The hydraulic boundary conditions were constant flux at the top to simulate 50 mm yr$^{-1}$ recharge and a constant head of 0 m between $z = 0$ and 25 m at $x = 10,000$ m (Fig. S1). All other boundaries are no flow boundaries. The resultant heads at $x = 0$ m range from 60 m (at $z = 0$ m) to 30 m (at $z = 25$ m). The net horizontal hydraulic gradients are 3 to 6 x10$^{-3}$ that are within the range commonly recorded in regional flow systems (Larocque et al., 2009; Irvine et al., 2015).

The geometry of the FEFLOW model and the shape of the flow field resembles that of the exponential lumped parameter model; however, for numerical stability, numerical models of solute transport require that a dispersivity be assigned (which is absent in the exponential lumped parameter model).

A small value of dispersivity (0.5 m) was used, resulting in mechanical dispersion in the numerical simulations being driven primarily by variations in $K$ (McCallum et al., 2014). Similar to the exponential
lumped parameter model, the outlet of the FEFLOW model is the entire aquifer thickness at \( x = 10,000 \) m.

**Fig. S1a.** Conceptual FEFLOW model with dimensions and boundary conditions. **S1b.** Annual \(^3\)H activities for Melbourne rainfall used in the FEFLOW model, data from Tadros et al. (2014).

To assess the impacts of heterogeneity, the output of four sets of 30 simulations with the same average \( K \) (1 m day\(^{-1}\)) but different variances of \( K \) (\( \sigma^2 \ln(K) = 0.3, 1.0, 2.5, 4.0 \)) were compared to a model with homogeneous \( K \). These \( \sigma^2 \ln(K) \) values span the range observed in many natural systems (Larocque et al., 2009; McCallum et al., 2014; Irvine et al., 2015). The \( K \) fields were generated using the Direct Sampling technique (Mariethoz et al. 2010; Mariethoz and Kelly 2011), which uses a Training Image (TI). The simulated \( K \)-fields were produced by sequentially assigning values to the simulated \( K \)-field that reproduce spatial patterns of \( K \) in the TI. The TIs (Fig. S2) were generated using the Sequential Gaussian simulator from GSLIB (Deutsch and Journel, 1998), following a similar process to that used by McCallum et al. (2014). TIs with dimensions of \( 25 \times 25 \) m, and \( \Delta x = \Delta z = 0.5 \) m were used, matching
the discretisation of the numerical model domain. The Tls were generated using a 200 m correlation length in the x direction, and the 5 m in the z direction. The output from GSLIB was converted to $K$ values with $\sigma^2\ln(K) = 0.3$ (Fig. S2a), 1.0 (Fig. S2b), 2.5 (Fig. S2c) and 4.0 (Fig. S2d). Similar ranges have been used elsewhere to investigate the influence of aquifer heterogeneity on transport processes (e.g. Larocque et al. 2009; McCallum et al. 2014; Irvine et al. 2015), and the $\sigma^2\ln(K)$ values span the range reported in the literature (e.g., LeBlanc et al. 1991; Barlebo et al. 2004). In total 30 $K$-fields were generated for each value of $\sigma^2\ln(K)$; in each model $\ln(K)$ is normally distributed.

![Fig S2: Training images (TI) for $\sigma^2\ln(K) = 0.3$ (a), 1.0 (b), 2.5 (c) and 4.0 (d, left column) and one of the resulting $K$-fields (out of 30).](image)

The FEFLOW simulations used time steps of 5 days over 65 years from 1950 to 2015. The relatively short time step ensured model stability and minimised numerical dispersion. The annual $^3$H activities from Melbourne rainfall interpolated onto the time steps of the model (Fig. 3b) were used as the $^3$H activity of recharge. Both $^3$H activities and age were simulated as solutes. Age was simulated by setting a constant age $= 0$ years for the upper boundary and calculating the travel time to the model outlet. Initial conditions were produced by first running the simulations at steady state using age $= 0$ and a pre-1950s $^3$H activity of 2.8 TU at the upper boundary. For all simulations, the mean age from FEFLOW ($F_{age}$) and mean $^3$H activity were calculated using the flux-weighted arithmetic means at the model
outlet (z = 10,000 m). The mean transit time corresponds to the mean age of the water at the model outlet.

References


