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Investigating the Age Distribution of Fracture Discharge Using Multiple Environmental Tracers, Bedrichov Tunnel, Czech Republic.

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Abstract:	The transit time distribution (TTD) of discharge collected from fractures in the Bedrichov Tunnel, Czech Republic, is investigated using lumped parameter models and multiple environmental tracers. We utilize time series of $\delta^{18}O$, δ^{2}H and ^{3}H along with CFC measurements from individual fractures in the Bedrichov Tunnel of the Czech Republic to investigate the TTD, and the uncertainty in estimated mean travel time in several fracture networks of varying length and discharge. We compare several transit time distributions, including the dispersion distribution, the exponential distribution, and a developed TTD which includes the effects matrix diffusion. The effect of seasonal recharge is explored by comparing several seasonal weighting functions to derive the historical recharge concentration. We identify best fit mean ages for each TTD by minimizing the error-weighted, multi-tracer χ^{2} residual for each seasonal weighting function. We use this methodology to test the ability of each TTD and seasonal input function to fit the observed tracer concentrations, and the effect of choosing different TTD and seasonal recharge functions on the mean age estimation. We find that the estimated mean transit time is a function of both the assumed transit time distribution and seasonal weighting function. Best fits as measured by the χ^2 value were achieved for the dispersive model using the seasonal input function developed here for two of the three modeled sites, while at the third site, the best fit was achieved with the exponential model and our seasonal input function. The average mean transit time for all TTDs and seasonal input functions converged to similar values at each location. The sensitivity of the estimated mean transit time to the seasonal weighting function was equal to that of the transit time distribution. These results indicated that understanding seasonality of recharge is at least as important as the uncertainty in the flow path distribution in fracture networks, and that unique identification of the TTD and mean

	transit time is difficult given the uncertainty in the recharge function. However, the mean transit time appears to be relatively robust to the structural model uncertainty. The results presented here should be applicable to other studies using environmental tracers to constrain flow and transport properties in fractured rock systems.
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Dear Editor,

The paper submitted here describes an investigation of the mean travel time in fracture networks estimated using multiple environmental isotope tracers. The focus is on determining the uncertainty in the derived mean travel time when different transit time distribution and different seasonal recharge weighting functions are used. The resulting analysis will be useful for future studies where environmental isotopes are used to estimate mean ages, and/or mean groundwater residence times are used to calibrate distributed flow and transport models. The data was provided as part of the DECOVALEX-2015 Task C2.

Thank you,

W. Payton Gardner

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(will be inserted by the editor)

Investigating the Age Distribution of Fracture Discharge Using Multiple Environmental Tracers, Bedrichov Tunnel, Czech Republic.

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Abstract The transit time distribution (TTD) of discharge collected from fractures in the Bedrichov Tunnel, Czech Republic, is investigated using lumped parameter models and multiple environmental tracers. We utilize time series of $\delta^{18}O$, δ^{2}H and ^{3}H along with CFC measurements from individual fractures in the Bedrichov Tunnel of the Czech Republic to investigate the TTD, and the uncertainty in estimated mean travel time in several fracture networks of varying length and discharge. We compare several transit time distributions, including the dispersion distribution, the exponential distribution, and a developed TTD which includes the effects of matrix diffusion. The effect of seasonal recharge is explored by comparing several seasonal weighting functions to derive the historical recharge concentration. We identify best fit mean ages for each TTD by minimizing the error-weighted, multi-tracer χ^2 residual for each seasonal weighting function. We use this methodology to test the ability of each TTD and seasonal input function to fit the observed tracer concentrations, and the effect of choosing different TTD and seasonal recharge functions on the mean age estimation. We find that the estimated mean transit time is a function of both the assumed transit time distribution and seasonal weighting function. Best fits as measured by the χ^2 value were achieved for the dispersive model using the seasonal input function developed here for two of the three modeled sites, while at the third site, equally good fits were achieved with the exponential model and the dispersion model and our seasonal input function. The average mean transit time for all TTDs and seasonal input functions converged to similar values at each location. The sensitivity of the estimated mean transit time to the seasonal weighting function was equal to that of the transit time distribution. These results indicated that understanding seasonality of recharge is at least as important as the uncertainty in the flow path distribution in fracture networks, and that unique identification of the TTD and mean transit time is difficult given the uncertainty in the recharge function. However, the mean transit time appears to be relatively robust to the structural model uncertainty. The results presented here should be applicable to other studies using environmental tracers to constrain flow and transport properties in fractured rock systems.

Keywords Environmental Isotopes · Hydrogeology · Isotope Geochemistry · Surface Water

1 Introduction

Flow and transport in fracture networks remains one of the more challenging issues facing hydrogeologists today. Accurate predictions of flow and transport in fracture networks is important in a variety of hydrogeological problems including: nuclear waste repository safety assessment, contaminant transport and remediation, unconventional hydrocarbon production, enhanced geothermal energy production and mine reclamation. Fractured reservoirs are characterized by extreme heterogeneity and data scarcity. Generally, it is not possible to uniquely determine the spatial distribution of fracture network properties and provide a complete description of the system behavior; however, simple models which incorporate field data at the appropriate scale, should be able to provide insight into the salient feature of the system behavior (Neuman, 2005). The transport time distribution is a

powerful description of the flow and transport in a hydrogeologic system; however, little is known about the field scale transit time distribution in fracture networks. Lumped parameter models provide a simplified methodology to investigate the residence time distribution in fracture networks.

Transport in fracture networks is affected by rapid advection in fractures and diffusion into and out of adjacent intact lower-permeability matrix (e.g. Haggerty et al (2001); Maloszewski and Zuber (1990, 1985); Neretnieks (1980); Sudicky and Frind (1982); Tang et al (1981)). The effect of matrix diffusion is to slow tracer transport compared to the advective fluid velocity, dampen concentration variation and produce long tailing of tracer release (Cook and Robinson, 2002; Maloszewski and Zuber, 1985; Neretnieks, 1980, 1981). Tracer transport in fractured networks has been evaluated theoretically and used to interpret tracer transport experiments with a variety of models from analytical models of simple geometry to numerical models of discrete fracture networks. Multiple continuum models allow linear exchange between the advective and immobile porosities for a single rate (Warren and Root, 1963) to multiple mass transfer rates (Haggerty and Gorelick, 1995). Analytical solutions exist for models that account for concentration gradients in the matrix for simple geometries such as a single fracture (Tang et al, 1981), parallel fractures (e.g. Maloszewski and Zuber (1990, 1985); Sudicky and Frind (1982)) and a distribution of different matrix block shapes and sizes (e.g. Haggerty et al (2000)). Discrete fracture network models have been used to develop fully distributed reactive flow and transport models (e.g. Therrien and Sudicky (1996)) as well as produce fluid velocity fields for particle tracking schemes which can be post processed to account for matrix interactions (e.g. Painter and Cvetkovic (2005); Painter et al (2008); Roubinet et al (2013)). The parameters used in these models are a strong function of the spatial and temporal scale of interest (Neuman, 2005; Shapiro, 2001), thus observations used to estimate fracture parameters should be made at similar length and time scales to the desired predictions.

Environmental tracers can provide a rich dataset which can be used to understand fracture flow and transport over a wide range of spatial and temporal scales. Environmental tracer concentrations have been used to develop conceptual models of tracer transport, and constrain fracture network parameters in a variety of fractured rock systems. Cook and Robinson (2002) develop an analytical model of environmental tracer transport in parallel fractures and show that tracer concentrations could not uniquely determine fracture network properties, but were capable of providing some constraint fracture spacing, aperture and recharge. Cook et al (2005) use this model to constrain the recharge rate in a fractured, porous aquifer in South Australia, and show that this simple model was a reasonable representation of a more complex system and could be used prediction concentration breakthrough.

Groundwater mean age can be used to characterize the residence time distribution in fracture networks. Analytical models of mean groundwater age have been developed in fractured systems (Doyon and Molson, 2012). Groundwater age in the field is often determined using environmental tracer concentrations. However, in fracture networks, environmental tracer concentrations provide the solute residence time, which is not equal to the advective residence time (Neretnieks, 1981). Thus, "apparent" groundwater ages determined from environmental tracers will be older than the advective fluid residence time (Cook and Robinson, 2002), and the use groundwater age in fractured systems must incorporate matrix interactions.

The residence time distribution is a fundamental characteristic of a flow system and provides critical information for determining the parameters controlling flow and transport in the system. Residence time distriubtions have been developed for a variety of simplistic aquifers (Cook and Herczeg, 2000; Maloszewski and Zuber, 1996). These models provide a convenient method to investigate the residence time and flow path distribution in hydrogeologic systems (e.g. Gardner et al (2010, 2011); Solomon et al (2010, 2015)). Lumped Parameter models in conjunction with tracer transport observations provide a means to investigate the residence time distribution and infer flow path characteristics in a reservoir (Danckwerts, 1953). Multiple environmental tracers can be used to identify the best age distribution and mixing models which best fit the observed data (Gardner et al, 2011; Solomon et al, 2010). McCallum et al (2014) show that age tracers have limited ability to uniquely identify the age distribution, but indicate the time series of tracers can help reduce uncertainty. Solomon et al (2010) indicate that the mean ages derived from multiple environmental tracers appears to be robust to choice the transit time distribution. However, these studies were carried out for porous flow aquifers and the application of lumped parameter models in fractured flow systems to estimate the transit time distribution is questionable.

The aim of this paper is to provide some insight into the uncertainty in estimating the mean transit time and the ability to distinguish the transit time distribution in fracture networks discharge to single fractures at the scale of 100's of meters using lumped parameter models and multiple environmental tracer concentrations. We utilize time series of $\delta^{18}O$, $\delta^{2}H$ and ^{3}H along with CFC measurements from individual fractures in the Bedrichov Tunnel of the Czech Republic to investigate the TTD in several fracture networks of varying length and discharge. We compare several transit time distributions, including a developed TTD which includes the effects matrix diffusion. We identify best fit mean ages for each TTD, and compare the ability of each TTD to fit the observed tracer concentrations and the effect of choosing different TTD on the mean age estimation.

2 Theory

Lumped parameter models were developed to investigate residence time distributions in chemical reactors where the true distribution of flow paths is not known (Danckwerts, 1953). The breakthrough curve of a tracer transported through the reactor can be used to provide information on the distribution of flow paths, and the processes affecting transport in the reactor, for example the amount of dead volume, the degree of mixing, and the amount of dispersion in 1-D flow in the reactor (Danckwerts, 1953). The residence time distribution has been used to investigate flow and transport in groundwater systems where the exact distribution of flow paths is unknown for many decades (e.g. Maloszewski and Zuber (1996)). The concentration at time t is given by the convolution integral:

$$C(t) = \int_0^\infty C_{in}(t-t')g(t')e^{-\lambda t}dt', \tag{1}$$

where τ is the residence time, and λ is the decay constant of a radioactive tracer, $C(t-\tau)$ is the historical input at recharge and $g(\tau)$ is the residence time distribution for the the flow paths discharging to the sampling point. Equation 1 along

with observations of tracer concentration in groundwater allows for the investigation of the distribution of flow paths and the processes affecting transport in the "black box" flow system which feeds the discharge point.

2.1 Residence Time Distribution

The residence time distribution contains the flow and transport information for the system upstream of the sampling point and has been derived for several simplified aquifer types (Cook and Herczeg, 2000; Maloszewski and Zuber, 1996). Here we consider two existing transit time distributions which encompass two possible end members of reservoir types, and develop a distribution which incorporates matrix diffusion. The simplest age distribution considered is for water flowing along a single flow path or fracture subject to longitudinal dispersion, termed here as the dispersion distribution which can be written (Maloszewski and Zuber, 1982):

$$g(t') = t'^{-1} \left(4\pi P_e t'/\tau\right)^{-1/2} exp\left[\frac{-\tau * (1 - t'/\tau)^2}{4P_e t'}\right], \tag{2}$$

where $P_e = D/vx$ is the system Peclet number with longitudinal dispersivity D, velocity v and transport distance x, and τ is the mean transit time.

An exponential distribution of residence times is achieved when there is complete mixing of an even distribution of flow paths of all ages, thus represents an end member where many different flow paths converge to the sampled fracture. The exponential distribution is written (Maloszewski and Zuber, 1982):

$$g(t') = \frac{1}{\tau} e^{-\frac{t'}{\tau}}. \tag{3}$$

Equations 2 and 3 were developed for porous media aquifers and do not consider the effect of diffusion into immobile regions on the transit time. In this study, a transit time distribution is developed that includes the effects infinite matrix diffusion for a constant fracture aperture. Here, we assume that the diffusion time to equilibrate matrix blocks with the fracture fluid is long compared to the changes in concentration, and that the fracture system is uniform.

The total transport time distribution ($g_{tran}(t_{tran})$) can be written as the convolution of the advective travel time distribution $g(t')$ and the retention time distribution ($g_{ret}(t_{ret})$) coupled by a velocity dependent transport resistant parameter distribution (Painter et al, 2008):

$$g_{tran}(t_{tran}) = \int_0^{\infty} \int_0^{t_{tran}} g_{ret}(t_{tran} - t'|\beta) f_{\beta|t'}(\beta|t') g(t') dt' d\beta, \tag{4}$$

where $t_{tran} - t' = t_{ret} \geq 0$ is the retention time from matrix diffusion and β is the spatially variable velocity dependent transport resistance parameter with a density distribution of $f_{\beta|t'}(\beta|t')$. If variability in the resistance parameter is neglected (Painter et al, 2008):

$$f_{\beta|t'}(\beta|t') = \delta(\beta - t'\bar{\beta}/\tau) \tag{5}$$

where δ is the Dirac function. In fractured rock applications, the resistance parameter is defined (Painter et al, 2008):

$$\beta(t') = \int_0^{t'} \frac{ds}{b(s)}, \tag{6}$$

where b is the fracture half aperture and s is distance along the flowpath. If a constant aperture is assumed then (Painter et al, 2008):

$$\beta = \frac{t'}{\bar{b}} = t' \frac{\bar{\beta}}{\tau}, \tag{7}$$

where $\bar{b}$ is the effective fracture aperture for the system of interest.

Painter et al (2008) present retention time distributions for a wide variety of subsurface transport processes. For this paper, unlimited matrix diffusion is considered. The retention time distribution for infinite matrix diffusion is:

$$g_{ret}(t_{ret}|\beta) = \frac{\kappa\beta}{2\sqrt{\pi}t_{ret}^{3/2}} exp\left[\frac{-\kappa^2\beta^2}{4t_{ret}}\right], \tag{8}$$

where $\kappa = \theta_{im}\sqrt{D_{im}}$, is a function of the matrix porosity θ_{im} and the matrix diffusion coefficient D_{im}.

The concentration at time t for a system which includes matrix diffusion can now be written:

$$C(t) = \int_0^{\infty} C_{in}(t - t_{tran}) g_{tran}(t_{tran}) dt_{tran}. \tag{9}$$

Inserting equation 4 into equation 9 and assuming constant aperture and no variance in the resistance transport parameter (equations 7 and 5) gives:

$$C(t) = \int_0^{\infty} C_{in}(t - t_{tran}) \int_0^{t_{tran}} g_{ret}(t_{tran} - t'|\beta)(t'/\bar{b}) g(t') dt' dt_{tran}. \tag{10}$$

Equation 10 describes the transit time distribution, and the resulting concentration expected in fracture discharge at time t and is a function of the purely advective travel time, modified by retention from diffusion into the adjacent matrix. Here, flow along a single constant aperture fracture is considered, thus the advective travel time is assumed to the dispersive distribution (equation 2). Equation 10 is then parameterized by the mean advective travel time τ, the longitudnal dispersivity P_e, the fracture aperture $\bar{b}$, matrix porosity θ_{im} and diffusivity D_{im}.

2.2 Seasonal Input Function

The concentration input history $C_{in}(t)$ in equations 1 and 10 is the historical concentration of a given tracer in water recharging the aquifer system. The concentration of these tracers is generally measured in precipitation and/or the atmosphere. The concentration in recharge will be modified from the atmospheric or precipitation concentration by the processes occurring during recharge such as soil flow, evapotranspiration and seasonality of precipitation. When recharge is

constant throughout the year, C_{in} will be equal to the concentration in precipitation. However, in cases where the concentration of the tracer changes seasonally – seasonality in precipitation, infiltration or evapotranspiration can cause C_{in} to differ from the atmospheric or precipitation record. These effects are especially important for stable isotopes of water and tritium. The annual flux weighted average concentration of tracer ($C_{in_{an}}$) is given by:

$$C_{in_{an}} = \frac{\sum_{i=1}^{12} C_i \alpha_i P_i}{\sum_{i=1}^{12} \alpha_i P_i}, \tag{11}$$

where $\alpha_i = R_i/P_i$ the recharge fraction is fraction of precipitation which become recharge for the ith month. In the area of the Bedrichov Tunnel winter is wetter and colder with precipitation predominantly falling as snow, and is a time of low evapotranspiration potential. During the summer months, precipitation falls as rain and there is a high evapotranspiration potential, thus there is a significant potential for seasonality in the recharge function.

To investigate the effects of seasonality we split the year into two seasons - summer and winter - and assume constant constant recharge fractions for summer months (α_s) and winter months (α_w). If the seasonal infiltration ratio $\alpha = \alpha_s/\alpha_w$ is defined, the seasonal recharge can be approximated by (Zuber and Maloszewski, 2001):

$$C_{in_{an}} = \frac{(\alpha \sum_{i=4}^{9} C_i P_i)_s + (\sum_{i=10}^{3} C_i P_i)_w}{(\alpha \sum_{i=4}^{9} P_i)_s + (\sum_{i=10}^{3} P_i)_w}, \tag{12}$$

where the summer months are assumed to be April through October and winter months November through March. The mean stable isotope composition of groundwater can be used to estimate α:

$$\alpha = \frac{(\sum_{i=3}^{10} P_i \delta_i)_w - \delta(\sum_{i=3}^{10} P_i)_w}{\delta(\sum_{i=4}^{9} P_i)_s - (\sum_{i=4}^{9} P_i \delta_i)_s}, \tag{13}$$

where δ is the mean groundwater isotope composition and sums are calculated for all winter and summer months on record.

The long term seasonal infiltration ratio α can now be used to estimate the seasonally weighted composition of recharge. This estimate has been done in variety of manners. The simplest and most common method is to use equation 12 to calculate the seasonally weighted annual average concentration. However, this limits the temporal resolution of the input function to annual steps. Interpretation of the high resolution sampling isotopic composition could provide information at sub-annual timescales, requiring a higher resolution input function. In this paper, seasonally weighted annual average precipitation along with three plausible monthly resolution input functions are investigated. The first and simplest high

resolution input function is simply the precipitation record, which inherently assumes $\alpha = 1$. The second infiltration function, taken from Zuber and Maloszewski (2001), is:

$$\delta_{in}(t) = \bar{\delta} + \alpha_i P_i(\delta_I - \bar{\delta}) / \sum_{i=1}^{12} \alpha_i P_i / n, \tag{14}$$

where $\bar{\delta}$ is the mean input (which must equal the mean output) and $\alpha_i = 1$ when $10 \geq i \leq 3$ and $\alpha_i = \alpha$ otherwise. We also develop another weighting function very close in form to equation 14:

$$\delta_{in}(t) = \bar{\delta} + \alpha_i P_i(\delta_I - \bar{\delta}) / \sum_{i=1}^{12} \alpha_i P_i. \tag{15}$$

The subtle difference between equations 14 and 15 is in the normalization term. Equation 14 is normalized by the average monthly recharge, thus recharge events greater than the average monthly recharge have much larger isotope shifts; however, this results in large fluctuations of the isotope composition when the precipitation is above the monthly average, and probably underestimates mixing in the vadose zone. Conversely, equation 15 is normalized by the total annual precipitation, which means that only months which comprise a significant amount of the total seasonally weighted total annual recharge can cause deviation from the seasonally weighted annual average recharge. This function produces a very smooth infiltration function very close to the seasonally weighted average concentration, but does allow large precipitation events to modify the input signal.

3 Study Area

The study location is a water supply tunnel which connects Josefuv dul reservoir with a water supply tunnel through the Jizera mountains in the northern Czech Republic. A description of the site can be found in *Hokr et al. this volume*. The tunnel is excavated through crystalline granitic rocks of the Krkonose-Jizera Composite Massif, a part of the Bohemian Massif of central Europe. The 2,600m tunnel was excavated in 1980-1981 with a maximum depth 200m below land surface (Figure 1). The tunnel has been utilized as a natural analogue underground laboratory for understanding fracture flow by the Czech radioactive waste disposal research group, RAWRA, since 2003.

Detailed geological history of the massif and characterization of the fracture network intersecting the tunnel are given in Zak et al (2009). The $\sim$1000 km^2 Krkonose-Jizera Plutonic Complex is dominated by the Jazira and Liberec course-grained porphyritic biotite granodiorite to granite. The tunnel lies in the Jaziera granite which is crosscut by two sets of steeply dipping roughly orthogonal fracture sets trending NE-SW and NW-SE. Fractures are roughly planar and subparallel and show little interaction with each other. Fracture spacing ranges from 1 cm to several meters with the most common spacing between 10 cm - 120 cm. Most fractures do not transmit measurable water discharge.

Fracture discharge and isotopic composition data were collected by the Technical University of Liberec and Czech Technical University in Prague respectively from 2010 to 2014 as part of the RAWRA characterization project. These data

were made available as part of the DECOVALEX 2015 project as part of a task to model flow and transport in fractured crystalline systems. Historical isotope composition of precipitation is available by leveraging the Uhlriska experimental watershed isotopes in precipitation database.

4 Field and Analytical Methods

Measurement of fracture discharge, water quality and temperature were at the sampling locations depicted in Figure 1. The irregularly spaced sampling intervals were chosen to be representative of different flow regimes in the tunnel (*Hokr et al. this volume*) (Figure 1). Manual measurements of fracture discharge were made at each site at 14 day intervals starting in 2006. Fracture discharge was measured using V-notch weirs or drip counting depending on the fracture discharge. Automatic measurements of fracture discharge at hourly intervals began in 2009. The locations of automatic measurement have continuously expanded since 2009, thus the density of data and sampling intervals vary for each sampling site. Automated measurements are verified by manual measurements at the 14 day intervals.

Stable isotope composition of fracture discharge at the sampling sites has been measured since 2010. Stable isotope samples were collected at 14 day intervals in 50 ml bottles and analyzed at the IAEA stable isotope laboratory in Vienna, Austria. Dissolved CFC's, tritium and dissolved noble gases were measured at the IAEA Isotope Hydrology Laboratory as part of an IAEA Technical Cooperation Project. Dissolved CFCs were collected in 250 ml glass bottles with metal caps completely submerged in buckets filled with fracture discharge. Dissolved CFC concentrations were measured using purge and trap gas chromatography. Tritium was collected in 1L bottles and analyzed by electrolytic enrichment and counting. Dissolved noble gases were collected using copper tube samples (Weiss, 1968) and analyzed using mass spectrometry.

5 Modelling Methods

The input concentration history $C_{in}(t)$ was estimated at the site for each tracer modeled. Stable isotopes in precipitation near the study site are available from the Uhlriska experimental watershed at monthly internals beginning in 2006. In order to extend the precipitation data set back in time, the Vienna stable isotope in precipitation data set, the closest and longest time series in the IAEA Global Network of Isotopes in Precipitation was used to provide an estimate of the values in precipitation at the site from 1960 to 2006.

The seasonal recharge coefficient α was calculated for each modeled location using the average δD and $\delta^{18}O$ composition of the groundwater and average of Uhlriska dataset and using equation 13 to calculate an α for each isotope. The α used to create the seasonally weighted recharge concentration was taken the average of that calculated with each isotope. Seasonally weighted concentrations were then calculated for tritium, δD and $\delta^{18}O$ using raw historical precipitation and equations 12, 14 and 15 for each sampling site at monthly intervals. The input history for CFCs was calculated using the historical CFC mixing ratios in the atmosphere (Bullister, 2011), the average noble gas recharge temperature

estimate of 4.8 °C from IAEA tritium-helium analyses, a characteristic recharge elevation of 750 m, and zero excess air, to give the CFC concentration at each site in biannual intervals. In order to provide high enough temporal resolution to reduce numerical error the time were resampled at daily intervals using linear interpolation to give the discrete concentration input history $\bar{C_{in}}$.

Equations 1- 10 were simulated using discrete convolution of linearly interpolated concentration input history ($\bar{C_{in}}$) and weighting function vectors ($\bar{g}$). Numerically efficient discrete convolution was accomplished using multiplication of the Fourier transformed discrete vectors. Convolution operations were written in python.

Three of the sampling locations depicted in Figure 1 representative of: shallow high discharge (V6), deep low-discharge fracture discharge (V3), and deep large discharge (V4) were modeled. The concentration in discharge was modeled at each location using equations 1 with the dispersion and exponential weighting functions and 10 with the dispersion advective travel time distribution. At each modeled location, three different concentration input histories were evaluated (equations 12, 14 and 15) using the methods described above.

For each combination of weighting function and historical concentration history, the best fit mean groundwater age at a sampling site was estimated by fitting the observed multi-tracer data at the site. Best fit estimation was accomplished by minimizing error weighted chi-squared residual using a Levenberg-Marquardt algorithm. The error mean age estimate was calculated from the 95% confidence interval of the covariance matrix, using the Jacobian of the error weighted chi-squared residual.

6 Results

The best fit modeled age, uncertainty estimate and χ^2 fit are reported for each precipitation weighting function for each age distribution at all modeled fracture locations in Tables 1-2. At each location, the mean age and standard deviation in estimated age for each age distribution due to the different conceptualization of seasonal recharge function is summarized in Table 4. The mean age and the standard deviation in mean age estimates due to the assumed age distribution is reported for each input function in Table 5. The effect of increasing mean travel time on δ^{18}O signal, along with the observed values at the V6 sampling site, is shown for a dispersion TTD (equation 2), using the annual average weighted precipitation function (equation 11) is shown in Figure 2. As the age increases the variation in the δ^{18}O signal decreases. The corresponding effect on the CFC-12 concentration, along with observed data for the V6 sampling site is shown in Figure 3. During the multi-tracer inversion, the difference between all the observed tracer and the modeled signal is minimized by varying the mean transit time.

Table 1 gives the summarized results for the V6 sampling site. The overall mean age for all age distributions and all recharge weighting functions is 4.58 years with a σ of 1.02 years. For the dispersive age distribution, the mean estimated age over all input functions is 3.4 years with a standard deviation (σ) of 2.6 years. The exponential age distribution gives a mean age of 5.0 years with a σ of 1.28 years for all input functions. For the matrix diffusion model, the mean total transit time is 5.33 years with a σ of 0.99 years for all input functions. In the matrix diffusion

model, the mean advective travel time is 2.55 year, with an average matrix diffusion retention time of 2.78 years. The best fit to the observed data as measured by the χ^2 residual was achieved by a dispersive age distribution with the seasonal input function developed here (equation 15). The observed data and the overall best fit modeled concentration for all tracers is shown in 4. Overall, the dispersion model gives the best fits to the measured data; however, the variance in χ^2 over all the precipitation input functions is high enough to encompass the χ^2 of the matrix diffusion age distribution, indicating that it is difficult to tell the difference between the two age distributions given the uncertainty in the input function (Table 4).

Table 2 summarizes the results for the V2 sampling site. The average mean travel time (τ) over all age distributions and input functions is 7.86 years with a σ of 2.2 years. For the dispersion age distribution, the average mean travel time is 10.3 with σ of 8.49. The exponential age function give an average mean travel time of 6.06 with a σ of 2.1 years. For the matrix diffusion distribution, the mean total transit time is 7.23 years with a σ of 2.3 years, with an average mean advective travel time of 3.3 years and an average mean retention time of 3.9 years. The best fit combination of models is the dispersion age distribution and input function 15. The observed data and overall best fit modeled concentration for all tracers at V2 is shown in Figure 5. Overall best fits are for the dispersion model followed by the exponential and finally matrix diffusion models. The variance in χ^2 due to uncertainty in the seasonal weighting function encompasses the fits of all other travel time distributions, thus it is difficult to uniquely pick any of the travel time distributions as the best (Table 4). For the seasonal weighting functions, the overall best fit is given by the developed seasonal weighting function (equation 15). This weighting function clearly provides the best fits regardless of the travel time distribution used (Table 5).

For the V4 sampling site, the overall average mean travel time is 6.56 years with σ of 1.2 years for all travel time distributions and input functions (Table 3). Average travel time for the dispersion distribution is 5.30 years with a σ of 2.13 years. Average travel time for the exponential age distribution is 6.7 years (σ 3.2 yr), and average travel time for matrix diffusion distribution is 7.74 (σ 3.2 yr). For the matrix diffusion distribution the average advective mean travel time is 3.47 years and the average retention time was 4.3 years. The best fit combination was our seasonal infiltration function (eq 15) with the dispersion equation; however, this combination produces an anomalously low mean travel time of 3.55 years. The observed data and overall best fit modeled concentration for all tracers at V4 is shown in Figure 6. The exponential age distribution fits the data nearly as well with an anomalously high mean travel time of 10.8 years. Over all the seasonal weighting functions, the best fits were given by the exponential model, followed by the dispersion model and finally the matrix diffusion model. At V4, uncertainty in the input function creates enough variance in the χ^2 residual that the average χ^2 values for each TTD are within error (Table 4), thus choosing the best age distribution is difficult given the available data. As in the other models, the input function developed in this paper provides better fits than the other input functions regardless of the travel time distribution (Table 5).

7 Discussion

The results presented here highlight the difficulty in using tracer concentrations to constrain travel time in a fractured network system. At all sampling locations it is difficult to distinguish the best travel time distribution when considering different conceptualizations of the seasonally weighted input function. The inability to distinguish a single best age distribution using measured tracer concentration is consistent with the findings of McCallum et al (2014) and Solomon et al (2010). Thus, the estimated mean age is dependent upon the conceptualization of the system. The standard error of the mean travel time estimate (defined as the standard deviation of mean travel times normalized by the average mean travel time) over all travel time distributions and seasonal weighting input functions ranges from 19% at the V4 sampling site to 22.4% at the V6 sampling site. These results indicate that tracer derived mean travel times converge to a similar value consistent with Solomon et al (2010), and can still be used to provide significant constraint on the flow system.

A significant finding of this study is that the estimated mean travel time is as sensitive to the assumed seasonal input function as the travel time distribution. The effect of seasonality of recharge on the age estimate is largely unconsidered in most studies, and these results show that it is at least as important as the age distribution assumption. Seasonality of recharge may not have as large an effect on travel time estimates which use dissolved gas tracers when the recharge zone is deep enough to keep the recharge temperature constant. However, in situations where the water table is shallow and seasonality is pronounced, recharge temperatures can differ from the mean annual temperature (Thoma et al, 2011) and seasonality will affect dissolved gas tracers. For isotopes of water including tritium and thus tritium-helium age dating, seasonality of recharge will always be important. Mixing of recharging waters during infiltration is a classic and active question in hydrology (e.g. Evaristo et al (2015); Kennedy et al (1986)), and more investigation is need on the effect of seasonality on groundwater age determination. Of the seasonal recharge functions considered in this study, the developed formula consistently fits the observed data better than other functions. This function keeps the recharge concentration close to the annual average concentration, but allows large isotopic deviations in precipitation to effect the recharge concentration. While considerably more investigation is needed to validate this recharge function, the good fits here indicate that at least in this setting it appears give a reasonable approximation to the processes involved.

For two of the three model locations V6 and V2, the observed concentration were best fit by the dispersion travel time distribution. These sampling locations correspond to the discharge from individual fractures intersecting the tunnel at different depths. The shallowest sampling location V6 had the shortest travel time and the deeper sampling location, V2, had a longer travel time. The lumped parameter modeling is conceptually consistent with discharge from these fractures occurring a single fracture network, described a single flow path, rather a collection of multiple flow paths which mix together. For one of the locations, V4, the exponential age distribution fits the data as well as the dispersion model. At this location discharge from a larger fault system was sampled, thus a mixture of different flow paths with a range of ages (which the exponential model represents) is a likely conceptual model. However, uncertainty in the seasonal weighting function

makes it difficult to identify which of these transit time distribution best fits the data.

The fact that the matrix diffusion model does not significantly improve fits indicates that, given the travel times in this fracture network and representative parameters controlling matrix diffusion, matrix diffusion is not a dominant process controlling transport in these fractures. The lack of significant matrix diffusion is also highlighted by the relatively small difference between the mean transport time including matrix diffusion and the mean advective only travel time (Tables 1-2). While the effect of matrix diffusion appears small on the mean travel time in this study, we expect that matrix diffusion would play a much stronger role in controlling the initial arrival and tailing properties of the transit time distribution, which are likely not identifiable using tracer concentrations (McCallum et al, 2014). Additionally, in environments with stronger matrix diffusion (e.g. high matrix porosity and/or longer travel times), the matrix diffusion transit time distribution developed here will be useful in modeling the total transit time.

8 Conclusion

In this study, we investigate the transit time distribution in fracture networks discharging to individual fractures of the Bedrichov Tunnel in the Czech Republic using time series of stable isotopes of water and tritium along with synoptic dissolved CFC concentrations. We use lumped parameter models to compare some likely transit time distributions and determine the effect of transit time distribution and seasonal input choice on the estimated mean age. We compare residence time distribution for a single advective path with longitudinal dispersion, complete mixing (exponential age distribution) and a newly developed a residence time distribution for 1D advective-dispersive transport with infinite matrix diffusion. In order to investigate the effect of seasonal recharge, we compare uniform infiltration, a weighting function developed in Zuber and Maloszewski (2001), and a weighting function developed in this paper. We find that the modeled concentrations are dependent upon the transit time distribution and the seasonal infiltration weighting function, and that the estimated mean travel time is as sensitive to the choice of seasonal weighting function as that of the transit time distribution. Given the uncertainty in the seasonal weighting function, it is difficult to completely identify the best fit transit time distribution. However, regardless of the age distribution or the infiltration model chosen, the best fit mean age converges to a similar value for all three locations modeled here. These results indicate that lumped parameter models along with multiple environmental tracers can be used constrain the mean age, and develop some information on the transit time distribution to help develop conceptual models of flow and transport in fracture networks. The results and methods presented should be applicable in other fractured crystalline environments.

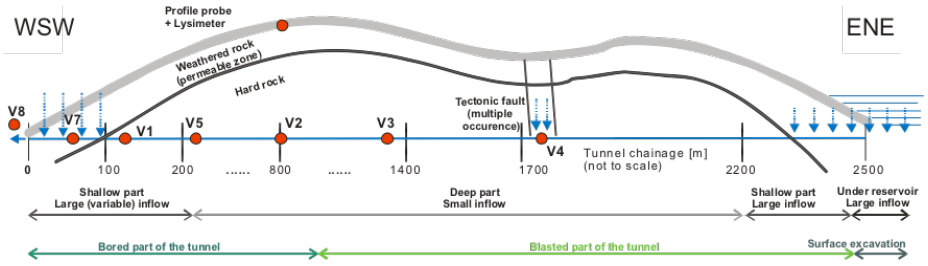

Fig. 1 Schematic cross-section and profile of the Bedrichov Tunnel along with technical and hydrological conditions.

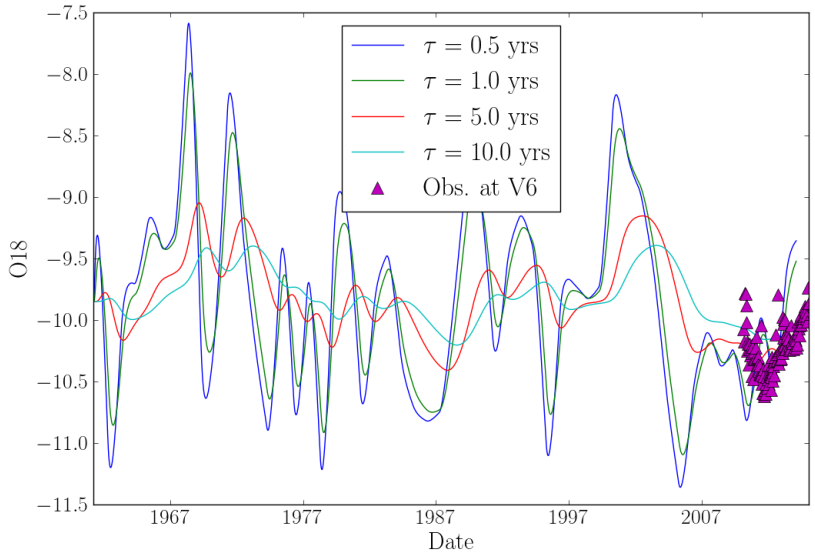

Fig. 2 Effect of increasing mean travel time on the modeled δ^{18}O signal using the annual average weighting function and the dispersion TTD for the V6 sampling site along with the observed δ^{18}O signal.

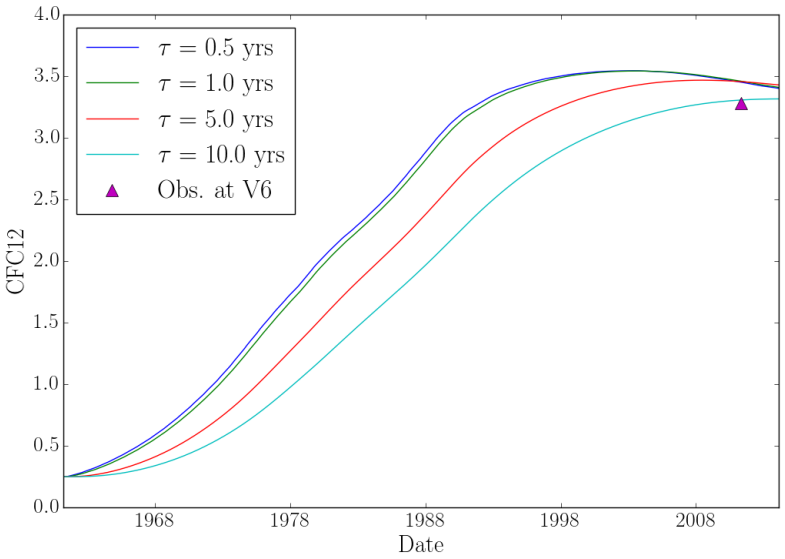

Fig. 3 Effect of increasing mean travel time on the modeled CFC-12 signal using the annual average weighting function and the dispersion TTD for the V6 sampling site along with the observed CFC concentration.

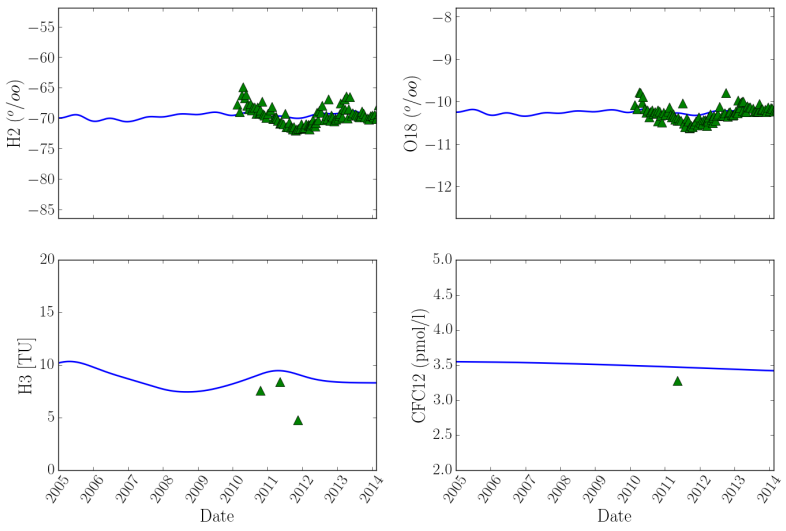

Fig. 4 Tracer data at the V6 sampling location, and the overall multi-tracer best fit lumped parameter model results. For V6 the best fit was achieved for a dispersion TTD using the seasonal input function derived in equation 15.

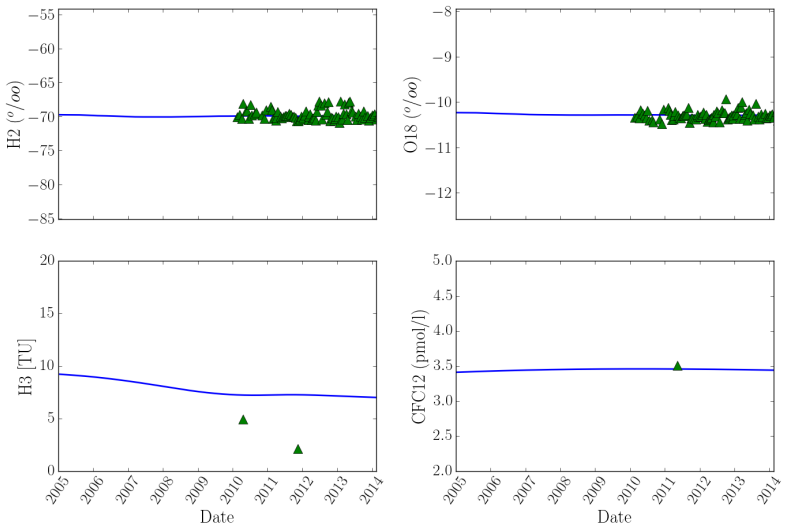

Fig. 5 Tracer data at the V2 sampling location, and the overall multi-tracer best fit lumped parameter model results. For V2 the best fit was achieved for a dispersion TTD using the seasonal input function derived in equation 15.

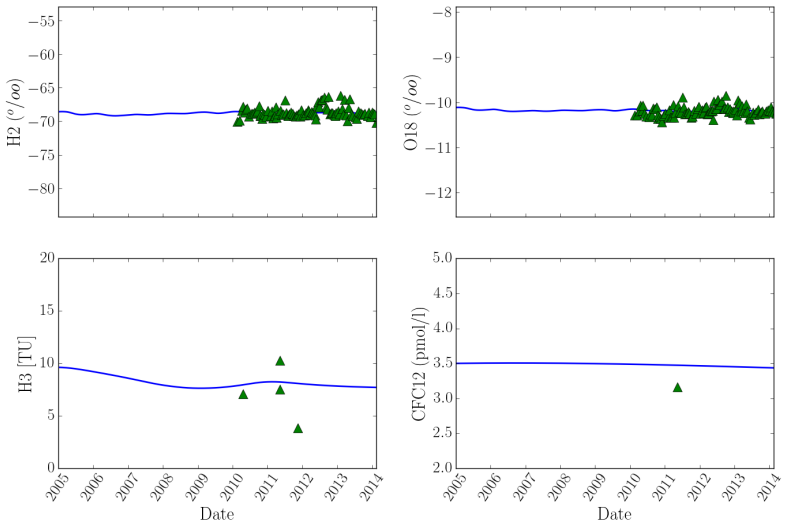

Fig. 6 Tracer data at the V4 sampling location, and the overall multi-tracer best fit lumped parameter model results. For V4 the best fit was achieved for a dispersion TTD using the seasonal input function derived in equation 15.

Table 1 Summary results for V6 sampling site.

Age Distriubtion ($g(t)$)	Infil. Func. ($C_{in}(t)$)	τ_{adv} (yr)	+/- (yr)	χ^2	τ_{tran} yr
Dispersive-Matrix Diffusion	Uniform	2.31	0.09	573	4.73
	eq. 11	3.09	0.31	299	6.67
	eq. 14	2.62	0.15	240	5.49
	eq. 15	2.19	0.65	200	4.45
Dispersive	Uniform	2.65	0.1	343	–
	eq. 11	7.17	0.73	283	–
	eq. 14	2.58	0.15	189	–
	eq. 15	1.27	0.14	177	–
Exponential	Uniform	3.1	0.18	651	–
	eq. 11	5.64	0.44	328	–
	eq. 14	5.78	0.55	382	–
	eq. 15	5.5	0.46	317	–

Table 2 Summary results for V2 sampling site.

Age Distriubtion ($g(t)$)	Infil. Func. ($C_{in}(t)$)	τ_{adv} (yr)	+/- (yr)	χ^2	τ_{tran} yr
Dispersive-Matrix Diffusion	Uniform	2.57	0.13	575	5.37
	eq. 11	4.56	0.32	214	10.6
	eq. 14	2.83	0.2	272	6.03
	eq. 15	3.22	1.95	107	6.99
Dispersive	Uniform	3.13	0.14	445	
	eq. 11	8.22	0.93	202	
	eq. 14	7.32	1.07	250	
	eq. 15	6.65	5.4	108	
Exponential	Uniform	3.07	0.18	466	
	eq. 11	7.72	0.7	226	
	eq. 14	6.66	0.77	303	
	eq. 15	6.81	6	111	

Table 3 Summary results for V4 sampling site.

Age Distriubtion ($g(t)$)	Infil. Func. ($C_{in}(t)$)	τ_{adv} (yr)	+/- (yr)	χ^2	τ_{tran} yr
Dispersive-Matrix Diffusion	Uniform	2.22	0.09	407	4.53
	eq. 11	4.49	0.16	210	10.4
	eq. 14	2.55	0.15	265	5.34
	eq. 15	4.62	0.22	101	10.7
Dispersive	Uniform	3.27	0.18	372	
	eq. 11	7.08	0.76	182	
	eq. 14	7.13	0.97	242	
	eq. 15	3.55	1.3	96.3	
Exponential	Uniform	3.15	0.18	292	
	eq. 11	7.24	0.62	224	
	eq. 14	5.62	0.54	271	
	eq. 15	10.8	1.94	99.7	

Table 4 Average mean age and standard deviation of mean age estimate for all seasonal input functions given a TTD at each sample location.

Model	Input func.	τ_{adv} (yr)	σ (yr)	$\bar{\chi}^2$	σ_χ
V6	Dispersive-Matrix Diffusion	5.33	0.993	328	168
	Dispersive	3.42	2.58	248	79.1
	Exponential	5	1.28	420	157
V2	Dispersive-Matrix Diffusion	7.24	2.31	292	201
	Dispersive	6.31	2.22	251	142
	Exponential	6.06	2.05	276	149
V4	Dispersive-Matrix Diffusion	7.74	3.26	246	127
	Dispersive	5.26	2.14	223	116
	Exponential	6.7	3.21	222	86.1

Table 5 Average mean age and standard deviation of mean age estimate for all TTDs given the seasonal input function at each sample location.

Model	Input func.	τ_{adv} (yr)	σ (yr)	$\bar{\chi}^2$	σ_χ
V6	Uniform	3.49	1.09	522	160
	eq. 11	6.49	0.78	303	22.8
	eq. 14	4.62	1.77	270	100
	eq. 15	3.74	2.2	231	75.1
V2	Uniform	3.86	1.31	495	69.8
	eq. 11	8.83	1.52	214	12
	eq. 14	6.67	0.645	275	26.6
	eq. 15	6.82	0.17	109	2.08
V4	Uniform	3.65	0.764	357	58.9
	eq. 11	8.23	1.85	205	21.4
	eq. 14	6.03	0.963	259	15.3
	eq. 15	8.36	4.17	99	2.43

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