The location of old groundwater in hydrogeologic basins and layered aquifer systems

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[1] The age of groundwater, the time since the water recharged the subsurface, is a fundamental characteristic of groundwater that impacts diverse geologic processes and practical applications. The distribution of groundwater age depends on many factors including permeability, recharge rate, aquifer geometry, and topography. Seminal work simulated topography-driven regional groundwater flow with various topographies, localized high-permeability zones, and more recently with permeability decreasing with depth, but the role of layered aquifer systems which are common in both consolidated and unconsolidated sediments has not been systematically explored. Here we show that high age zones with predictable locations occur in layered geologic systems across a wide range of hydraulic gradients, basin geometries, and permeabilities. Numerical simulations of a generic three-layer aquifer system indicate that high age zones consistently form in the low-permeability layer near the middle of the basin. The zones of older groundwater result from low groundwater velocities in the low-permeability layer and the rejuvenation of the groundwater through mixing of different flow paths near discharge zones. The high age zones are not hydraulic stagnation points but are associated with areas of low velocity. Formation and location of zones of high groundwater ages in low-permeability units are important as these units are targeted for radioactive waste disposal and shale gas extraction. High age zones are also likely to affect geologic processes that depend on groundwater or solute fluxes and may serve as archives of past hydrological or climatological conditions. **Citation:** Gassiat, C., T. Gleeson, and E. Luijendijk (2013), The location of old groundwater in hydrogeologic basins and layered aquifer systems, *Geophys. Res. Lett.*, *40*, doi:10.1002/grl.50599.

1. Introduction

[2] Groundwater age plays an important role in a wide range of geologic processes [*Alley et al*., 2002; *Bethke and Johnson*, 2002; *Freeze and Cherry*, 1977; *Fyfe et al*., 1978; *Garven*, 1995; *Ingebritsen et al*., 2006; *Tóth*, 1999]. Knowledge of the age of groundwater derived from radiogenic isotopes determines the timescales of subsurface processes and contaminant transport rates and is a key indicator for the renewability of groundwater resources [*Bethke and Johnson*, 2008; *Cook and Bohlke*, 2000; *Kazemi et al*., 2006]. Groundwater age is defined as the time that has passed since the water crossed the water table [*Alley et al*., 2002; *Kazemi et al*., 2006]. *Tóth* [1963] demonstrated that areas of stagnant groundwater form at the intersection of local, intermediate, and regional flow systems (Figure 1a). Such stagnant zones, with extremely high groundwater ages and low velocity, are important because they accumulate solutes and contaminants transported by groundwater [*Tóth*, 1999]. The location of stagnation points is sensitive to the basin geometry, the surface topography [*Tóth*, 1963], and the decrease of permeability with depth [*Jiang et al*., 2011]. Previous modeling studies have simulated groundwater ages in basins with a gradual decrease of permeability with depth [*Jiang et al*., 2011; *Jiang et al*., 2010]. *Jiang et al*. [2009] showed how decreasing permeability with depth limits regional groundwater flow, while *Jiang et al*. [2010] importantly showed that different groundwater flows lead to both simultaneous rejuvenation and aging of groundwater.

[3] However, most aquifer systems and hydrogeologic basins consist of layered sediments or rocks, in which small changes in material properties such as grain size [*Koltermann and Gorelick*, 1995] or clay content [*Revil and Cathles*, 1999] can generate orders of magnitudes of changes in permeability. Lenses or layers of high- or low-permeability materials have been shown to greatly modify regional groundwater flow [*Freeze and Witherspoon*, 1967], and groundwater age in specific basins has been simulated [*Sanford et al*., 2004]. *Goode* [1996] and *Sanford* [2011] showed that overturned groundwater ages (younger ages under older ages) can exist where substantial vertically layered heterogeneity is present, regardless of the upper boundary condition. Field evidence from two basins indicates that these overturned ages exist in layered systems, sometimes due to groundwater inflow from adjacent basins and mountains [*Sanford et al*., 2001; *Sanford et al*., 2002; *Sanford et al*., 2004]. However, previous authors have not named these "high age zones," documented their occurrence in a wide variety of hydrogeological conditions, examined the processes and hydrogeological conditions that control their formation, or discussed their importance to a variety of Earth processes.

[4] Hydrogeologic basins and aquifer systems commonly have laterally extensive low-permeability layers. Laterally extensive low-permeability layers are particularly common in basins that form due to flexural or thermal subsidence, such as foreland and sag basins. In basins created by extensional or strike-slip tectonics, faulting often limits the continuity of strata [*Ingersoll*, 1988]. In unconsolidated aquifer systems, laterally extensive low-permeability units such as glacial tills, lacustrine and marine clays, and silts are common.

Additional supporting information may be found in the online version of this article.

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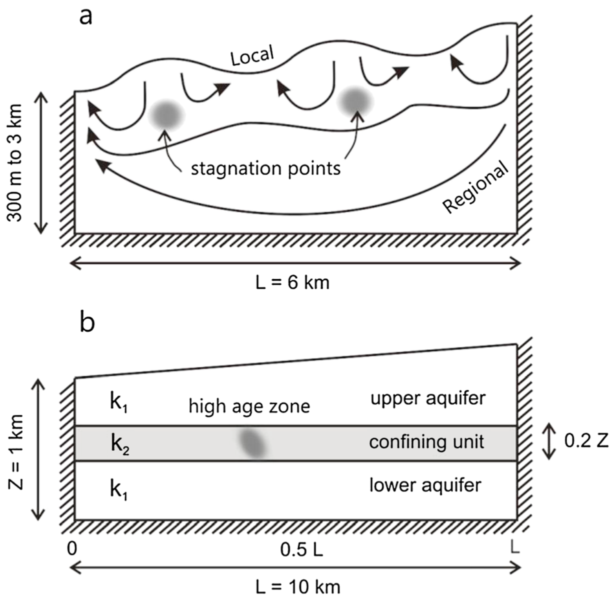

Figure 1. Stagnation points and zones of high groundwater age in regional-scale groundwater flow. (a) Theoretical model and results described in *Tóth* [1963]. Grey areas represent stagnation zones that form at the intersection of local, intermediate, and regional flow systems. (b) Theoretical model used in this study with dark grey area showing the high age zone. The hatched lines indicate a no-flow boundary.

In unconsolidated aquifer systems consisting of fluvial, glaciofluvial, alluvial, and colluvial deposits [*LaBolle et al.*, 2006], units are less continuous. The heterogeneous permeability field in such discontinuous aquifer systems has been successfully simulated with geostatistical methods [*Marsily et al.*, 2005]. Simulating the distribution of groundwater ages in such heterogeneous permeability field is computationally expensive and limited to small scales (hundreds of meters) at sites with detailed hydrogeological data [*LaBolle et al.*, 2006].

[5] Our objective is to scrutinize the processes and conditions that lead to the formation of high groundwater age zones at regional scales (Figure 1b). Therefore, we simulate a simplified layered and heterogeneous system in order to simulate the process of high age formation in a wide variety of hydrological and geological settings, while acknowledging that hydrogeologic basins and layered aquifer systems often have multiple low-permeability zones and crosscutting faults. We numerically simulated groundwater age in a wide range of steady state, regional-scale (one to hundreds of kilometers) groundwater systems (see "Methods" and Table S1 in the supporting information for the range of parameter values). We simulate mean groundwater age mass as a tracer subject to advection, diffusion, and dispersion using a yearly production of one unit of conservative solute within the fluid [*Bethke and Johnson*, 2002; *Goode*, 1996; *Voss and Wood*, 1994]. We define "high age zone" as a zone within the low-permeability layer where the maximum groundwater age is at least twice that of surrounding water (see "Methods" in the supporting information for more details).

2. Where High Age Zones Form

[6] For a range of hydraulic gradients and permeability values that represent typical value of sedimentary rocks ($k_1 = 10^{-13}$ m^2) or unconsolidated sediments ($k_1 = 10^{-11}$ m^2) [*Gleeson et al.*, 2011], simulations show that distinct zones of high groundwater ages develop in the low-permeability unit (Figure 2). The location and width of this high age zone vary systematically with permeability contrast and hydraulic gradient. Three model simulations illustrate the location and width of the high age zone for different permeability contrasts between the low-permeability layer and the surrounding medium (Figures 2a–2c). At low permeability contrast ($k_2 = 10^{-14}$ m^2), the high age zone contains groundwater that is a little older than twice the age of the surrounding groundwater. The zone is spread out over a width of 0.3 of the total length of the basin (L), and the maximum age is located at 0.35 L (Figure 2a). At moderate permeability contrast ($k_2 = 10^{-15.4}$ m^2), the ages in the high age zone are more than 30 times older than in the surrounding area. The maximum age within the low-permeability unit is located near the midline of the basin, and the high age zone is relatively narrow (Figure 2b). At higher permeability contrast ($k_2 = 10^{-16}$ m^2), the high age zone is wider and the age contrast less pronounced (Figure 2c). In all cases, the maximum groundwater age is located between 0.2 and 0.5 L. When the permeability of the low-permeability unit (k_2) is lower than 10^{-16} m^2, the high age zone does not form (Figure 2e). In most simulations, high groundwater ages are also found near the downgradient boundary condition as previous authors have shown [*Jiang et al.*, 2009; *Jiang et al.*, 2010].

[7] The location of the high age zone depends on the depth, thickness, and continuity of the low-permeability layer and the aspect ratio of the basin. High age zones shift toward the downgradient part of the low-permeability layer for high values of layer depth or thicknesses (Figure 3). For both low and high values of continuity, the high age zone is located close to the midline of the aquifer. For moderate continuities, the high age zone is located further downgradient. This change in behavior is due to the transition from a single flow system (with one recharge area and one discharge area) to a more complex flow system with multiple recharge and discharge areas at a layer continuity of ~0.7. The aspect ratio of the basin also impacts the location of a high age zone. In shallow, wide basins, the high age zone is located closer to the discharge zone with low to moderate permeability contrasts ($\log(k_1) - \log(k_2) \leq 2$). For basins with high-permeability contrasts, the high age zone is independent of the aspect ratio. The location of high age zones is largely independent of the basin length (Figure 3d).

3. Why High Age Zones Form

[8] Our results suggest that two mechanisms lead to the formation of a zone of high groundwater ages. First, high age zones are caused by mixing of groundwater from flow paths that travel through only the upper two layers and flow paths that travel through all three layers. For a wide range of permeabilities of the low-permeability unit (k_2), the groundwater velocity in the lower aquifer is high enough for the groundwater to travel faster than the groundwater flowing only through the overlying low-permeability unit. Upward flow of the younger groundwater near the discharge zone leads to rejuvenation of groundwater age within the low-permeability unit. High age zones are formed upgradient of the intersection of flow paths going through the lower aquifer and the low-permeability unit, and through the low-permeability unit only. Second, the

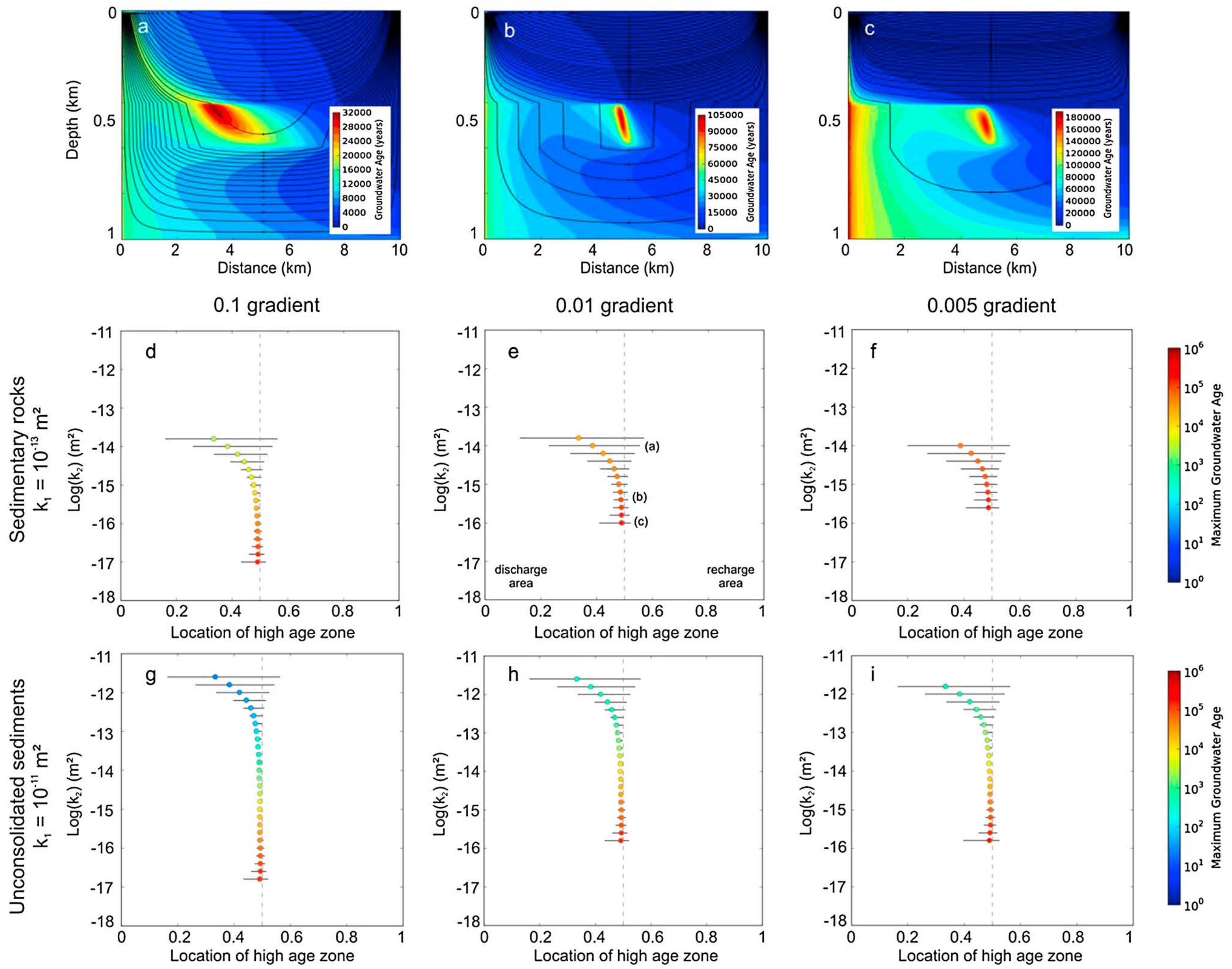

Figure 2. Zones of high groundwater age are common across a wide range of hydraulic gradients and permeability contrasts. Distribution of groundwater age within the aquifer under a 0.01 hydraulic gradient for sedimentary rocks ($k_1 = 10^{-13}$ m^2) and a layer permeability of (a) $k_2 = 10^{-14}$ m^2, (b) $k_2 = 10^{-15.4}$ m^2, and (c) $k_2 = 10^{-16}$ m^2. Location of high age zone versus k_2 for sedimentary rocks ($k_1 = 10^{-13}$ m^2) under a gradient of (d) 0.1, (e) 0.01, and (f) 0.005. Location of high age zone versus k_2 for unconsolidated sediments ($k_1 = 10^{-11}$ m^2) under a gradient of (g) 0.1, (h) 0.01, and (i) 0.005. Circles represent the location of the maximum groundwater age; lines represent the area where groundwater ages are higher than 50% of the maximum age. For each case, simulations have been run for values of k_2 from k_1 to 10^{-18} m^2. Values not specified mean that a high age zone does not form for this set of parameters.

high age zones are controlled by the flow velocities in the low-permeability unit. The lowest velocities are found near the middle of the basin and coincide with the location of the high age zone.

[9] Which of these two mechanisms is dominant controls the location of the high age zone and is predictable from the permeability contrast between the aquifer and the low-permeability unit. At low-permeability contrasts, the velocity is relatively constant over most of the low-permeability unit, and the high age zone is mainly controlled by the rejuvenation of groundwater below the discharge zone due to upward flow of groundwater from the lowest aquifer. At high-permeability contrast, the location of the high age zone follows the narrow zone of low flow velocities that is located in the middle of the basin. Model simulations confirm that high age zones are not necessarily stagnation points, zones where groundwater velocity is zero or/and flow is in opposite directions [*Jiang et al.*, 2011]. In our model simulation, flow velocities maintain values of 10^{-4} m/yr or higher. However, high age zones develop in areas of low velocities.

4. High Age Zones Form in a Variety of Basins

[10] High age zones form in hydrogeologic basins and aquifer systems with a range of permeability contrasts (Figure 2). The permeability contrast ($k_1 - k_2$) in which high age zones form in hydrogeologic basins and unconsolidated sediments is ~3–4 and ~5–6 orders of magnitude, respectively. At high-permeability contrast, the whole low-permeability lens contains groundwater that is older than the surrounding area, but no distinct high age zone forms within the unit. In fact, the permeability contrast ($k_1 - k_2$) for which a high age zone develops is predictable (Figure 4). For example, for simulations with a 0.01 gradient, high age zones form when the permeability of the low-permeability unit (k_2) is higher than 10^{-16} m^2 and 1 order of magnitude lower than

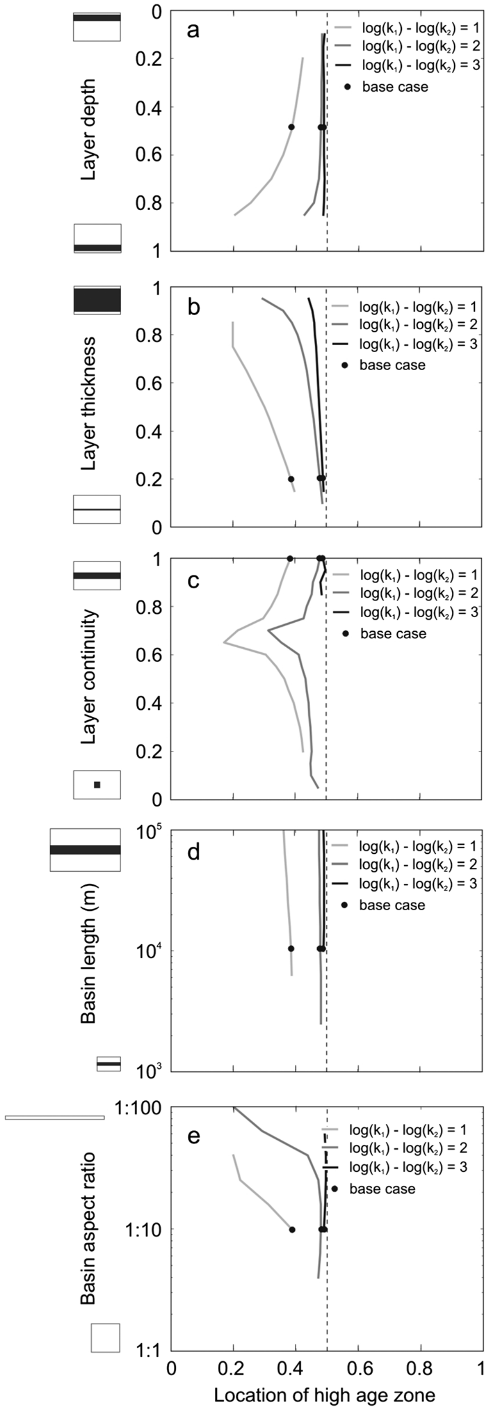

Figure 3. Zones of high groundwater age are common in diverse basin stratigraphies and geometries. Location of high age zone for sedimentary rocks ($k_1 = 10^{-13}$ m^2) with permeability contrasts from 1 to 3 and under a 0.01 gradient depends on (a) layer depth, (b) layer thickness, (c) layer continuity, (d) basin length, and (e) basin aspect ratio. Black dots represent the basin geometry of the base case (Figure 1b). Values not specified mean that a high age zone does not form for this set of parameters. Layer depth and thickness are specified as fractions of the total depth and thickness of the basin. Layer continuity is specified as fraction of the total length of the basin, and the layer is cut off both at the recharge and discharge ends.

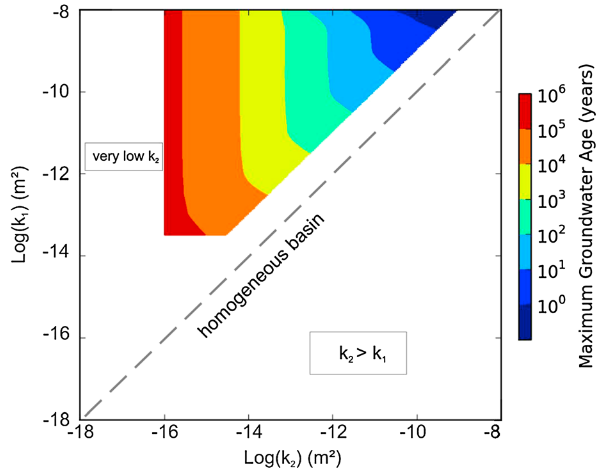

Figure 4. The permeability of the low-permeability unit controls the groundwater age. Values of k_1 and k_2 allow the formation of a high age zone under a 0.01 gradient. The colored area represents the couple (k_1, k_2) for which a high age zone forms. The color indicates the value of the maximum age within the low-permeability layer. Simulations were run for $k_1 = 10^{-8}$ to 10^{-17} m^2 and $k_2 = 10^{-8.5}$ to 10^{-20} m^2.

the permeability of the rest of the aquifer k_1. The maximum groundwater age in the low-permeability unit depends on the layer permeability but not on the permeability of the aquifer, except when the permeability contrast is small. The formation of high age zones is thus primarily controlled by the permeability of the low-permeability layer. Moreover, in both hydrogeologic basins and aquifer systems, high age zones do not form when the permeability of the low-permeability unit is less than approximately 10^{-16} m^2.

[11] Model simulations show that high age zones develop for a wide range of depth, thickness, and continuity of the low-permeability unit and basin geometries (Figure 3). High age zones form in layer depths (normalized to total thickness) of 0.1 to 0.9 (Figure 3a). Similarly, high age zones form in normalized layer thicknesses of 0.1 to 0.9 (Figure 3b). For low-permeability contrasts ($\log(k_1) - \log(k_2) < 3$), high age zones form in basins with a range of continuity values (Figure 3c). However, for higher permeability contrasts, high age zones form only in basins with nearly continuous low-permeability layers (normalized continuity > 0.8). However, high age zones do not develop in relatively small basins (<10^3–10^4 m length). High age zones form in basins with a variety of aspect ratio from 1:10 to >1:50 (Figure 3e).

5. Conclusions and Implications

[12] Numerical simulations clearly show that zones of high groundwater age should be expected in regional (>2 km length) layered hydrogeologic basins and aquifer systems over a wide range of naturally occurring permeability contrasts, hydraulic gradients, and basin geometries. Narrow zones of groundwater with ages up to a million years form in low-permeability units downgradient of midline of typical hydrogeologic basins and aquifers systems. These high age zones form due to low groundwater velocities in the low-permeability layer and the rejuvenation of the groundwater

through mixing of different flow paths near discharge zones. Simulations suggest that these high age zones likely form if the permeability of the low-permeability layer is at least 1 order of magnitude lower than the flanking units but greater than ~10^{-16} m^2. The lower limit corresponds to the regional-scale permeability of fine-grained sedimentary rocks [*Gleeson et al.*, 2011] or the highest permeability values for argillaceous material [*Neuzil*, 1994]. Therefore, these high age zone are expected in many hydrogeologic basins and aquifer systems, although to our knowledge they have rarely been observed in the field. This is likely because groundwater age studies tend to focus on permeable units, and low-permeability units are rarely sampled in sufficient detail to reveal lateral age gradients. High age zones may have been observed in a part of the Pannonian Basin in Hungary with a high density of ^{14}C age data. Several wells show a decrease of ^{14}C age with depth, which cannot be explained by groundwater flow in a homogenous anisotropic aquifer [*Sanford et al.*, 2001; *Sanford et al.*, 2002]. We suggest that they may instead be explained by the existence of high age zones. Our model results are a testable hypothesis for a systematic sampling campaign of groundwater ages within a regional-scale low-permeability unit. The simplified layered system developed in this study leads to a relatively complex distribution of groundwater age, and therefore, we expect a more complex pattern in a field study. Additionally, it is uncertain how three-dimensional flow in layered hydrogeologic basins and aquifer systems could affect the development and processes of high age zones.

[13] The strong lateral variations in groundwater ages in low-permeability layers found in this study have implications for a range of geological processes and Earth science research fields. Water-rock interactions that depend on groundwater and solute fluxes, such as the precipitation or dissolution of carbonates, (de-)dolomitization, and quartz cementation [*Bjørlykke*, 1994; *Ingebritsen et al.*, 2006; *Machel*, 1999], are likely to be limited in high age zones. This may cause lateral variations in porosity, permeability, and mechanical properties of low-permeability materials, which to our knowledge have not previously been considered. High age zones may inhibit the subsurface transport of microbes by groundwater [*Walvoord et al.*, 1999] and the resulting biodegradation of oil and gas [*Head et al.*, 2003], which could have implications for the extraction of hydrocarbons from low-permeable strata. Similar to stagnation points [*Tóth*, 1999], high age zones can form favorable locations for the storage of radioactive waste, as the risk of the spreading of contaminants is relatively low. High age zones are likely to impact the interpretation of groundwater age tracers, which are key tools for characterizing natural flow systems and quantifying groundwater recharge and sustainable extraction rates [*Alley et al.*, 2002; *Cook and Bohlke*, 2000; *Kazemi et al.*, 2006; *Scanlon et al.*, 2002]. Finally, the observed lateral age variations in low-permeability units are also likely to have an impact on studies that use environmental tracers to reconstruct past hydrological [*Beyerle*, 1998] or climatological conditions [*Weyhenmeyer*, 2000]; knowledge of their location allows targeted sampling of old groundwaters.

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