



## Review papers

# Comparison of hydraulic and chemical methods for determining hydraulic conductivity and leakage rates in argillaceous aquitards



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## SUMMARY

The capacity of argillaceous aquitards to effectively separate fresh water aquifers from lower quality waters has been widely studied for several decades using both hydraulic and chemical methods. Hydraulic methods, namely laboratory, field and regional groundwater flow modelling, are used to estimate hydraulic conductivity ( $K$ ), whereas chemical methods, namely aquitard porewater and aquifer groundwater chemistry, are used to estimate either leakage rates or porewater velocity ( $V$ ). We reviewed a total of 40 formations where either  $K$  or  $V$  (or both) have been estimated. Typical vertical hydraulic conductivity ( $K_z$ ) values in argillaceous materials estimated using hydraulic methods are  $10^{-12}$ – $10^{-9}$  m s<sup>-1</sup>. Usual  $V$  values, estimated using chemical methods, ranges between 0.01 and 1 mm y<sup>-1</sup>, although the range is as wide as  $10^{-4}$ – $10^3$  mm y<sup>-1</sup>, when inferred from hydraulic  $K_z$  measurements. Based on a Péclet number of 1, we calculated the lower limits of porewater velocity that can be reliably identified for different tracers, in most cases ranging from <1 mm y<sup>-1</sup> for <sup>2</sup>H, <sup>18</sup>O and Cl<sup>-</sup> to >100 mm y<sup>-1</sup> for temperature. Despite the limited number of sites where both methods were applied, comparison between hydraulic and chemical-derived values showed a reasonable correlation, although the range of  $K_z$  and  $V$  estimated using hydraulic methods is larger than that obtained using chemical methods. Methods applied to the whole aquitard thickness (some field hydraulic methods, regional groundwater flow modelling, aquitard porewater and aquifer chemistry) consistently result in lower  $K_z$  values, most probably indicating the presence of very low  $K_z$  layers within the aquitard, likely to be missed while using field hydraulic methods that test only a section of the entire thickness.  $K_z$  was observed to decrease with depth, presumably due to an increase in consolidation resulting in loss of porosity and smaller aperture of fractures and joints. Multi-disciplinary studies involving the use of hydraulic and chemical methods at different scales, combined with geophysical techniques to locate fractures at local and regional scales, are highlighted as promising avenues for the study of inter-aquifer leakage.

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## 1. Introduction

Quantification of inter-aquifer leakage, or flow through aquitards, is critically important for a number of water resources management purposes. There is growing interest to capture atmospheric carbon and store it underground, and this requires knowledge of seal integrity to avoid displacement of saline fluids (Birkholzer et al., 2009; Zhou et al., 2008), acidification of adjacent aquifers and possible heavy metal mobilisation (Kharaka et al., 2010; Mehlhorn et al., 2014) and bacterial proliferation (Oppermann et al., 2010). Confining layers in multi-layered shallow aquifer systems play an important role in isolating fresh water aquifers from deep low-quality waters. Exploitation of unconventional coal seam and shale gas involves injection of hydraulic fracturing fluids into, and groundwater extraction from deep formations, leading to concerns of depletion of adjacent aquifers (Moore, 2012) and mobilisation of natural salts (Brinck and Frost, 2007) and fracking fluids (Batley and Kookana, 2012; Gordalla et al., 2013) to shallower aquifers exploited. In the last two decades several sites have been proposed as underground repositories for radioactive waste (Bonin, 1998; NWMO, 2011), and these sites often rely on argillaceous rocks to host and isolate the waste. All of these activities require detailed and extensive characterization of the “sealing” capabilities of argillaceous rocks.

Approaches used to study inter-aquifer leakage can be grouped into two categories: (1) hydraulic methods and (2) chemical or tracer (henceforth, chemical) methods. Hydraulic methods are used to estimate hydraulic conductivity ( $K$ ), while chemical methods are applied to estimate flux or porewater velocity ( $V$ ). Hydraulic conductivity is related to flux through the hydraulic gradient and to velocity through hydraulic gradient and porosity. Methods applied to estimate aquitard porosity are described by Patriarche et al. (2004), Savoye et al. (2006) and van der Kamp et al. (1996). Vertical hydraulic conductivity ( $K_z$ ) measured in argillaceous formations can range from as low as  $10^{-17}$  m s<sup>-1</sup> (Luffel et al., 1993) to as high as  $10^{-5}$  m s<sup>-1</sup> (Eaton et al., 2007; Jørgensen et al., 1998; Nilsson et al., 2001; Sidle et al., 1998), with higher values usually related to the presence of fractures and weathered shallow tills.  $V$  rates estimated using chemical methods range between  $10^{-3}$  and  $5.6$  mm y<sup>-1</sup> (Gardner et al., 2012). Reviews on  $K$  of argillaceous rocks have been previously presented (Brace, 1980; Neuzil, 1986, 1994), but these reviews focus exclusively on bulk  $K$  measurements obtained using hydraulic methods applied at different scales.  $K_z$  is of main interest for inter-aquifer leakage, and chemical approaches can represent different spatial and temporal scales than hydraulic methods, and involve different assumptions. Thus, a comparison between hydraulic and chemical methods to estimate  $K_z$  may assist in identifying uncertainties of the methods, and would provide a scientific basis for determining which method is most appropriate for a given scale of application and study objective.

By applying Darcy's Law and the assumption of steady-state flow,  $K_z$  values obtained using hydraulic methods ( $K_{z\text{-hydraulic}}$ ) can be converted to leakage rates or porewater velocities ( $V_{\text{hydraulic}}$ )

using vertical head gradients and porosity, and can be compared to velocities obtained using chemical methods ( $V_{\text{chemistry}}$ ). Similarly,  $V_{\text{chemistry}}$  can be converted to  $K_z$  values ( $K_{z\text{-chemistry}}$ ), and compared to  $K_{z\text{-hydraulic}}$ . In this review, we present a brief description of the hydraulic and chemical methods, summarize their drawbacks and advantages, and review  $K_z$  values and  $V$  rates for different consolidated and unconsolidated argillaceous aquitards. We compare reported values of  $K_z$  and  $V$  in those formations where both methods have been applied, and analyse the literature data in terms of its scale of application and limits of resolution. Crystalline rocks and consolidated argillaceous formations are both used to host underground wastes, but only unconsolidated argillaceous formations overlain and isolate shallow aquifers utilised for water supply. Because of this, in this review we focus on both consolidated and unconsolidated argillaceous aquitards, but not crystalline rocks.

## 2. Methods to estimate inter-aquifer leakage

### 2.1. Hydraulic methods for estimating $K$

Although several laboratory and field hydraulic techniques exist, here we only discuss the most commonly used. A number of papers describing these methods already exist, and the reader is referred to these for detailed descriptions.

#### 2.1.1. Laboratory-based methods

Laboratory methods such as consolidation tests and permeameters can be used to determine  $K_z$  on small cores obtained from the aquitard matrix. Consolidation tests involves the use of an oedometer, an apparatus that applies increments of pressure to a sample core in order to measure the change of its volume and its consolidation coefficient, from which  $K_z$  is calculated (Keller et al., 1989). Permeameters force a liquid or gas to flow through the core, while the rate of flow and the difference of pressure at the two ends of the core are measured to estimate  $K_z$  from Darcy's Law (Hendry, 1982; Phillips et al., 1989; Timms et al., 2014).

Two main issues are of potential concern for laboratory tests: method reliability and scale of application. The reliability of these methods depends on a number of variables such as preparation and size of the cores, sample deterioration (i.e.: desiccation and microfracturing; Neuzil, 1993; Neuzil and Provost, 2014), equipment configuration and measurement uncertainties, applied stresses and whether tests are performed in steady-state or transient flow fields (Timms et al., 2014). Uncertainty introduced by the method used can be only quantified if  $K_z$  estimates are obtained by applying different methods to the same core at the same depth. Otherwise, differences between methods can simply be attributed to spatial heterogeneity. Timms et al. (2014) compared  $K_z$  obtained from a custom designed centrifuge permeameter and standard gravity column permeameter applied to samples of the same core and depth, and found differences up to one order of magnitude. These discrepancies were attributed to differences in test setup, such as core diameter size and the use of deionized water instead

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