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A simple reason explains why it is so difficult to assess groundwater ages and contamination ages

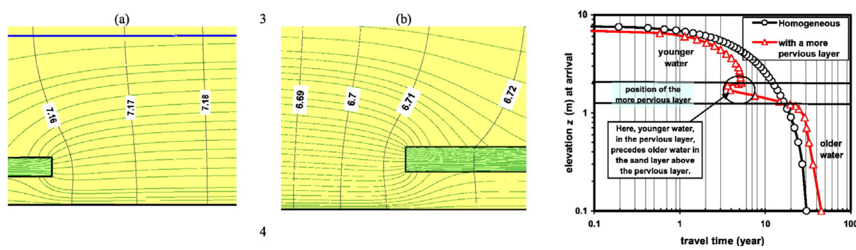
Robert P. Chapuis

Department CGM, École Polytechnique, P.O. Box 6079, Sta. CV, Montreal, QC, H3C 3A7, Canada

HIGHLIGHTS

- Groundwater age is difficult to assess, due to the aquifer heterogeneity.
- A simple case of a single more pervious layer in an aquifer is studied numerically.
- This layer mixes waters, which results in a wide distribution of groundwater age.
- There is no need to introduce diffusion and dispersion to have this age dispersion.
- To clarify this issue, numerical models must respect mathematical convergence rules.

GRAPHICAL ABSTRACT



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ABSTRACT

Several difficulties to assess groundwater age are known, including the aquifer heterogeneity. Strangely, the natural mixing process due to a simple stratification, and only particle tracking without diffusion and dispersion, has never been studied. It is examined here numerically, for a simple case of heterogeneity in which there is a single more pervious layer, for the age that atmospheric water takes in groundwater above, below and within the more pervious layer. Seepage converges to reach this layer and then diverges when it leaves it. In the layer, a target for a monitoring well, waters of different ages are mixed. This simple example of heterogeneity shows that stratification is the basic mechanism explaining dispersion of ages and also of any concentration. It appears that diffusion and dispersion are secondary mechanisms. This explains why it is so difficult to assess groundwater ages and contamination ages. To clarify this issue of basic and secondary mechanisms, a numerical model has to respect rules of mathematical convergence, which take considerable time. These are briefly explained, but are rarely used in groundwater numerical models, which is unfortunate.

1. Introduction

When groundwater is sampled in stratified aquifers, many characteristics fluctuate in time and space, for example concentrations (Kapoor and Kitanidis, 1998; Feenstra, 2003; Guilbeault et al., 2005; Petelet-Giraud et al., 2015), groundwater age (Weissmann et al., 2002; McMahon et al., 2013), and underground transit time for streams (Morgenstern et al., 2010; Manning et al., 2012; Atkinson et al., 2015). The groundwater age is defined as the residence time of water between its entering into the ground as atmospheric water to its discharge to a well or some surface water. It was used in many papers to underline the difficulties to estimate groundwater recharge rate, groundwater velocity, predict the fate of contaminants (e.g., Larocque et al., 2009; Han et al., 2015; McCallum et al., 2014a, 2014b, 2015), or the long-term yield of an aquifer (e.g., Chatton et al., 2016). Fluctuations are caused

E-mail address: robert.chapuis@polymtl.ca.

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by irregular emissions of pollutant or tracer from many sources including atmospheric water (e.g., Darling and Gooddy, 2007; Bauer et al., 2001), fluctuations in groundwater local velocities, physical, chemical and biological reactions, and aquifer heterogeneity (e.g., Lopez et al., 2015), even in laboratory conditions (Brusseau et al., 2000).

Because space and time variability are central, the sampling frequency is critical (Papapetridis and Paleologos, 2012). It is a challenge to assess the age of any groundwater (e.g., Park et al., 2002; Bethke and Johnson, 2002a, 2002b, 2002c; Tonina and Bellin, 2008; Sanford, 2011), and especially the age of contaminated young groundwater (Robertson et al., 2016).

The aquifer heterogeneity and two man-made factors have been pointed out as reasons for the challenge. A few numerical studies of singular or statistically defined heterogeneities have been published, but always considering diffusion, which was viewed as a critical factor (Bethke and Johnson, 2008; Larocque et al., 2009; Sanford, 2011; Molson and Frind, 2012; Rezaei et al., 2016). For example, Bethke and Johnson (2002a, 2002b)) studied how aquitards may contribute to the age of aquifer waters: the journal Geology withdrew their first paper (Bethke and Johnson, 2002a) which had too many errors, but the authors published a revision (Bethke and Johnson, 2002b).

The apparent ages, derived from environmental tracer concentrations, were often compared with numerical ages given by numerical models. Many papers have reported a poor or flawed correlation between the two ages. It has been proposed that flaws occur because age should be viewed not as a single value but as probabilistic value. The mechanisms leading to a discrepancy between the two "ages" include lateral exchanges with aquitards, transverse dispersion, dual porosity effects, immobile zones, and heterogeneity. Several papers have tried to limit the biases by using correction processes, but these seem to depend mostly upon the user and the numerical grid (McCallum et al., 2015).

Other papers have studied the two man-made factors due to incorrect design and installation of monitoring wells. This is a central problem. According to Nielsen and Schalla (2005), at least 2/3 of these wells are incorrectly installed, which has been confirmed by the author's personal experience with legal prosecutions. Waters of different ages may be mixed via two paths. The first mixing occurs in the borehole wall: a damaged and/or poorly sealed wall creates vertical hydraulic short-circuits between aquifer layers (Chapuis and Sabourin, 1989; Chesnaux et al., 2006) and mixes waters down to the screen where water is sampled. The second mixing takes place within the well screen and filter pack, and its probability increases with the filter pack length (Kearl, 1997; Zinn and Konikow, 2007; Berthold and Börner, 2008; Baudron et al., 2014).

This paper does not examine the two man-made processes. It focuses on the natural mixing due to heterogeneity. Currently, when multiple tracers, mostly from atmospheric origin, are used to assess groundwater ages, it is assumed that the water samples result from minimal mixing of waters having different ages. This assumption, which may be erroneous (Suckow, 2014), is examined here. Field studies are repeatedly finding dispersed water ages. The reason for age dispersion could be simple: aquifer heterogeneity. This natural dispersion would be then merely increased by the two man-made processes. However, numerical studies of heterogeneity effects have been carried out using advection plus diffusion and/or dispersion. With such approaches, the numerical results suggest firstly that the theory would need long travel distances and large times to become applicable. They suggest secondly that the dispersion of results may fit the dispersion of field data, but only statistically. These are disappointing results.

Groundwater mixing resulting from stratification is worth studying. It seems that no study has ever been done on the influence of stratification with advection alone, without any diffusion. What could we learn from this study? Could we show that a simple stratification, using particle tracking alone and no diffusion, is a sufficient reason for a large variability in groundwater age?

A simple mixing process is studied here numerically for an unconfined aquifer – the most likely to be polluted – without simplifying the highly non-linear equations for unsaturated and saturated seepage. The numerical convergence rules, as defined in computational fluid dynamics (Roache, 1994, 2009) are respected, but the lengthy convergence studies are not presented here. The results are compared for (i) a homogenous aquifer, for which analytical solutions are available for seepage and age and (ii) the same aquifer but with a single more pervious layer.

To better understand related numerical issues, a well documented example is examined first, an example for which the author was given full access to files, data and interpretations, which became public documents. This situation is noteworthy because most published cases do not give all the key details that were made available for the example below.

2. Example of poor methods and predictions

Murphy et al. (2010) published a case study in which they used ^{3}H—^{3}He data to assess groundwater ages, and compared them to advective ages obtained with a 3D numerical model and particle tracking. The correlation was shown to be poor in Court (Chapuis, 2010a), even after the authors had discarded several data. For example, for an isotopic age of about 30 years, the numerical model gave numerical ages between 3 and 36 years, whereas for a numerical age of about 15 years, the ^{3}H—^{3}He data gave ages between 4 and 34 years.

Several independent proofs were given in Chapuis (2010a) that the groundwater velocities used by Murphy et al. (2010) were at least 3 times too slow, and that all 3D numerical models (with over 10^6 nodes) used for this case study had major flaws and self-contradictory results, due to non-respect of numerical modeling mathematical rules (e.g., size of elements, minimum number of elements per layer, etc.), which yielded, for example, huge errors for protection perimeters (Chapuis, 2010a).

The velocity values of Chapuis (2010a) were supported by results of field tracer tests performed independently by Biogénie (2010) and Dessau (2010) in their field pilot tests to try to reduce the concentrations of chlorinated compounds in groundwater. For designing their pilot test Biogénie (2010) were given groundwater velocities in the 60–150 m/y range, based on numerous previous studies performed by different companies. However, the pilot test had a limited performance. Field tracer tests were carried out by Biogénie (2010) who obtained velocities of 475 to 548 m/y. As a result, the residence time of water within the treatment unit, which would have been 5 months for a groundwater velocity of 60 m/y and 2 months for 150 m/s, was only 20 days as measured in the field. This was a good reason for having a limited performance. Similarly, in reporting their pilot test results, Dessau (2010) explained that they were expecting a groundwater velocity of 60–150 m/y according to the information they had be given. After having designed their pilot test for 80 m/y, they recorded a poor performance. Their own tracer tests gave a groundwater velocity of about 350 m/y, four times faster than anticipated, thus a good reason for the limited performance of their pilot test.

Two points summarize this unfortunate example case: (i) there was a large dispersion of age values, and (ii) it was difficult to understand the reason for this dispersion because the numerical models were incorrectly designed and not verified using a quality control process.

3. Methods

Several methods are available to assess groundwater age. For unconfined aquifers, the water age is often obtained after assuming flow tubes similar to those in a homogenous aquifer (Fig. 1).

This model assumes that a water volume migrates as a closed system along a stream tube, without exchanging molecules with adjacent stream tubes (Fig. 2). This is called piston flow age, advective or