



# Geochemical and flow modelling as tools in monitoring managed aquifer recharge



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## ABSTRACT

Due to a growing world population and the effects of anthropogenic climate change, access to clean water is a growing global concern. Managed aquifer recharge (MAR) is a method that can help society's response to this increasing demand for pure water. In MAR, the groundwater resources are replenished and the quality of the recharged surface water is improved through effects such as the removal of organic matter. This removal occurs through mechanisms such as microbial decomposition, which can be monitored by studying the isotopic composition of dissolved inorganic carbon (DIC). Nevertheless, the monitoring can be difficult when there are other factors, like dissolving calcite, affecting the isotopic composition of DIC.

The aims of this study were to establish a method for monitoring the decomposition of organic matter (dissolved organic carbon – DOC) in cases where calcite dissolution adds another component to the DIC pool, and to use this method to monitor the beginning and amount of DOC decomposition on a MAR site at Virttaankangas, southwestern Finland. To achieve this, we calculated the mean residence times of infiltrated water in the aquifer and the fractions of this water reaching observation wells. We conducted geochemical modelling, using PHREEQC, to estimate the amount of DOC decomposition and the mineral reactions affecting the quality of the water.

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## 1. Introduction

Access to clean water is a growing global concern (Gale, 2005). In many countries, a growing population leads to both a higher need for food production and an increased demand for drinking water. These two pressures produce an increased demand for pure water and an increased use of groundwater reserves (Falkenmark and Widstrand, 1992; Shah et al., 2000). Replenishing groundwater reserves with surface water is a viable way of maintaining groundwater resources (Freeze and Cherry, 1979; Asano, 1985; Ma and Spalding, 1997; Gale, 2005). Managed aquifer recharge (MAR) is a general term for any engineering process where surface water is introduced into groundwater (Dillon, 2005; Page et al., 2012; Dillon et al., 2009). The main benefits of MAR are natural subsurface treatment for impurities, storage of water supplies (Grove and

Wood, 1979), and groundwater replenishment, which may help reduce salinity (Dillon et al., 2002).

Untreated surface water is generally not suitable for communal water supply use, often due to its high organic carbon content (Vartiainen et al., 1987; Lindroos et al., 2002; Grünheid et al., 2005; Kortelainen and Karhu, 2006). Organic matter can facilitate unwanted bacterial growth in water supply systems leading to odour and taste problems (Miettinen et al., 1999; Lindroos et al., 2002). Combining dissolved organic carbon (DOC) with chlorine disinfection can lead to the formation of mutagenic or carcinogenic compounds (Lindroos et al., 2002).

MAR is widely used in Europe: for example, in Germany, Finland, Sweden and Hungary to remove DOC from surface water and make it more suitable for human consumption (Sundlöf and Kronqvist, 1992; Hatva, 1996; Van Breukelen et al., 1998; Kuehn and Mueller, 2000; Lindroos et al., 2002; Balderer et al., 2004; Grünheid et al., 2005; Kortelainen and Karhu, 2006; Kolehmainen et al., 2009). In North America, Australia and Asia the benefits of MAR for water quality have also been reported (Herczeg et al.,

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2004; Dillon et al., 2002; Vanderzalm et al., 2006; Moon et al., 2012; Graham et al., 2015). During artificial recharge, surface water is infiltrated into the topsoil of an aquifer by direct infiltration through infiltration ponds (Kortelainen and Karhu, 2006; Kolehmainen et al., 2009) or wells (Vanderzalm et al., 2006; Pavelic et al., 2007), by bank filtration (Grünheid et al., 2005) or by sprinkling (Lindroos et al., 2002). Previous studies have shown that the DOC content of the infiltrated surface water decreases most significantly early on along the flowpath (Frycklund, 1995, 1998; Greskowiak et al., 2005; Kortelainen and Karhu, 2006; Vanderzalm et al., 2006).

During MAR, DOC concentration declines as a function of distance from the infiltration site (Kortelainen and Karhu, 2006; Kolehmainen et al., 2009; Grünheid et al., 2005; Lindroos et al., 2002). This is due to two possible processes: oxidative decomposition due to microbial activity in the aquifer, and sorption on the sediment without decomposing activity (Kortelainen and Karhu, 2006; Kolehmainen et al., 2009). Knowing the amount of oxidative decomposition of DOC, as opposed to DOC removal by sorption to the sediment, is important for understanding the purification process and the effects of MAR to the environment. In previous studies, 40–50% of the total DOC decrease has been attributed to oxidative decomposition (Frycklund, 1995, 1998; Kortelainen and Karhu, 2006; Kolehmainen et al., 2009).

The oxidative decomposition of DOC can be monitored by examining the isotopic composition of dissolved inorganic carbon (DIC) in the MAR water, since the naturally occurring DIC and the DIC produced by the decay of organic material have a different isotopic signal. Decaying organic material adds a significantly lighter component into the DIC pool (Le Gal La Salle et al., 2005; Kortelainen and Karhu, 2006; Kolehmainen et al., 2010). This can be achieved by simple mass balance calculations when there are no external sources of carbon contributing to the DIC pool.

Calcite-rich sediment and limestone are preferable environments for MAR for two reasons. Calcite dissolution is a beneficial process in MAR applications as the higher pH of water prevents corrosion of the water distribution pipes (Kortelainen and Karhu, 2009). Moreover, it has been shown by Pavelic et al. (2007) that calcite dissolution prevents aquifer clogging in MAR systems. Clogging leads to loss in permeability which shortens the lifespan of a MAR site (Vanderzalm et al., 2006; Pavelic et al., 2007). Treating the infiltrated water to prevent aquifer clogging is possible, but costly, and therefore the natural prevention provided by calcite dissolution is a definite asset (Pavelic et al., 2007).

Calcite dissolution complicates the quantification of DOC decomposition as it introduces a third end-member to the carbon budget. The DIC in the infiltrating water is typically derived from the soil or the atmosphere and depleted in  $^{13}\text{C}$  compared to dissolved sedimentary calcite and also enriched in  $^{13}\text{C}$  compared to DOC (Veizer and Hoefs, 1976; Deines, 1980; Vogel, 1993; Schiff et al., 1997; Kortelainen and Karhu, 2009). Therefore, there are two opposing processes contributing to the change in the isotopic composition of DIC. Isotopic evidence can be used in geochemical modelling to constrain these opposing processes and their effect on the composition of DIC. In modelling the MAR system, the contribution of carbon from different sources can be identified if the isotopic compositions of the different sources are known. The model uses the isotopic compositions together with the chemical composition of the source and resulting waters to produce mixing ratios between the two types of carbon.

A MAR site infiltrating surface water from the Kokemäenjoki River located north of the city of Turku was commissioned in 2010 at Virttaankangas. Prior to the startup of the site, thorough studies were conducted on the sedimentological background and water chemistry (Kortelainen et al., 2007; Kortelainen and Karhu,

2009) and a three-dimensional model of the sedimentological and hydrogeological units containing the aquifer was developed (Artimo et al., 2003).

The MAR site meets the main water supply needs of the Turku region with approximately 285 000 inhabitants (Artimo et al., 2003). The Virttaankangas site was selected to host the MAR site because of the vast groundwater reservoir in the aquifer, its proximity to the Turku area and the naturally high pH of the water, which is due to calcite dissolution (Kortelainen and Karhu, 2009).

The main goal of this study was to quantify the oxidative decomposition of DOC and the dissolution of calcite to estimate the effectiveness of the MAR site at Virttaankangas. As a part of this, we had to determine the point in time when DOC decomposition commenced. To achieve these goals, we used a groundwater flow model to determine the flow routes from infiltration areas to the production wells and then the hydrogeochemical modelling program *PhreeqC* (Parkhurst and Appelo, 2013) to determine the onset and the amount of DOC decomposition in the aquifer.

## 2. Study site

### 2.1. Hydrogeological setting

The Virttaankangas esker is part of the glaciofluvial Säkylänharju-Virttaankangas complex, located in south-western Finland (Fig. 1). The area features a boreal climate. The complex was formed during the late Weichselian and early Holocene deglaciation as a meltwater deposit between two sublobes of the retreating Baltic Sea ice-lobe (Punkari, 1980; Kujansuu et al., 1995; Artimo et al., 2003). The esker contains sub-surface kettle hole structures. Because of these structures the travel time cannot be estimated only by the distance and sediment permeability as water entering these kettle hole structures can make the flow take longer than anticipated.

The complex can be divided into three units: The lowermost part is a result of repeated deposition of subaqueous fans containing glaciofluvial sands and gravels (Artimo et al., 2003). The intermediate part is composed of glaciofluvial sand and silt and associated glaciolacustrine silt and clay deposits of a proglacial water body. The uppermost 10–20 m of the complex represent littoral sands and gravels formed by the regression of the water level in the Baltic Basin about 11 000–10 500 years ago (Artimo et al., 2003). The calcite in the Virttaankangas aquifer is located in the two lowermost units. The concentration of calcite is highest in the <0.2 mm fraction, with 3.7–5 wt % calcite, 0.5–1.4 wt % of the total sediment (Kortelainen et al., 2007). The  $\delta^{13}\text{C}$  value of the calcite is  $-4.8 \pm 0.5\text{‰}$  (Kortelainen et al., 2007). Calcite dissolution increases the pH of natural groundwater to 8–9.5 (Kortelainen and Karhu, 2009). Other minerals present in the Virttaankangas sediment are feldspar, quartz, muscovite, talc, chlorite and hematite (Kortelainen et al., 2007).

### 2.2. The MAR system

The MAR site at Virttaankangas consists of seven infiltration areas, each of which comprises of two to four infiltration ponds (Fig. 1). The site can be divided into western and eastern sides. The western side consists of three infiltration areas (IA301, IA303 and IA400), and four production wells (VIK32, VIK33, VIK42 and VIK43), and the eastern side four infiltration areas (IA401, IA500, IA501 and IA503) and three production wells (VIK51, VIK53 and VIK54). The natural groundwater flow is from northwest to southeast, but pumping can reverse the flow direction. Surface water is first treated with poly-aluminium chloride (PACl) to remove some of the DOC before infiltrating into the aquifer.

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