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Characterization of a managed aquifer recharge system using multiple tracers

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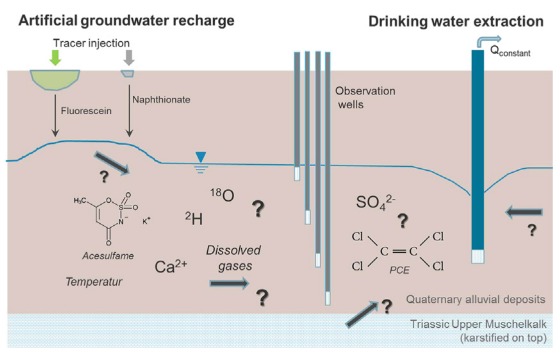
HIGHLIGHTS

- Our study assesses information gained from seven different tracers.
- Tracers to identify mixing are major ions, stable isotopes, and micropollutants.
- Dye and heat tracers were useful to identify travel times of the infiltrated water.
- Time series of hydrocarbons and dissolved gases identify changes of flow system.
- We demonstrate that heterogeneity necessitates application of multiple tracers.

GRAPHICAL ABSTRACT



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ABSTRACT

Knowledge about the residence times of artificially infiltrated water into an aquifer and the resulting flow paths is essential to developing groundwater-management schemes. To obtain this knowledge, a variety of tracers can be used to study residence times and gain information about subsurface processes. Although a variety of tracers exists, their interpretation can differ considerably due to subsurface heterogeneity, underlying assumptions, and sampling and analysis limitations. The current study systematically assesses information gained from seven different tracers during a pumping experiment at a site where drinking water is extracted from an aquifer close to contaminated areas and where groundwater is artificially recharged by infiltrating surface water.
We demonstrate that the groundwater residence times estimated using dye and heat tracers are comparable when the thermal retardation for the heat tracer is considered. Furthermore, major ions, acesulfame, and stable isotopes (δ^2H and $\delta^{18}O$) show that mixing of infiltrated water and groundwater coming from the regional flow path occurred and a vertical stratification of the flow system exist. Based on the concentration patterns of dissolved gases (He, Ar, Kr, N_2, and O_2) and chlorinated solvents (e.g., tetrachloroethene), three temporal phases are observed in the ratio between infiltrated water and regional groundwater during the pumping experiment. Variability in this ratio is significantly related to changes in the pumping and infiltration rates. During constant pumping rates, more infiltrated water was extracted, which led to a higher dilution of the regional groundwater. An infiltration interruption caused however, the ratio to change and more regional groundwater is extracted, which led to an increase in all concentrations. The obtained results are discussed for each tracer considered

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and its strengths and limitations are illustrated. Overall, it is demonstrated that aquifer heterogeneity and various subsurface processes necessitate application of multiple tracers to quantify uncertainty when identifying flow processes.

1. Introduction

Due to increasing demand for water for residential and industrial uses, supplying drinking water in urban areas is challenging (Fletcher et al., 2013). Often, the availability of water is limited due to various groundwater contaminants (Baillieux et al., 2015; Schirmer et al., 2013). Managed aquifer recharge (MAR) is one way to meet water demands and operationally protect sites that produce drinking water (Asano and Cotruvo, 2004; Bouwer, 2002). The infiltration of surface water creates a water surplus and dilutes potentially contaminated groundwater (Dillon, 2005). Typically, physical, chemical, and biological degradation processes improve the quality of this infiltrated water during percolation through the vadose zone (Greskowiak et al., 2005; Henzler et al., 2014). In addition, MAR can be used to build up a local groundwater mound, which can serve as a hydraulic barrier to prevent inflow of contaminated water from areas upstream (Auckenthaler et al., 2010; Franssen et al., 2011; Moeck et al., 2016).

Knowledge about the residence times of artificially infiltrated water and its flow paths is essential to developing adequate groundwater-management and protection schemes (Bekele et al., 2014; Zoellmann et al., 2001). In addition, this information is prerequisite to efficient monitoring and risk assessment. The residence time indicates the travel time of a pollutant from the source to the drinking-water extraction well (Regnery et al., 2015). Short residence times might indicate that groundwater is vulnerable due to limited time for self-purification (Kralik et al., 2014).

Typically, various tracers are used to study residence times and gather information about subsurface processes. Tracers can be grouped into three general categories: 1) artificial tracers, including dye tracers (Ptak and Schmid, 1996; Runkel, 2015); 2) natural tracers, including heat tracers (Becker et al., 2013; Irvine et al., 2015), hydrochemistry data (e.g., major ions (Abou Zakhem and Hafez, 2012, Moeck et al., 2016), stable isotopes (e.g., δ^2H and δ^{18}O (Clark et al., 2004, Demlie et al., 2008, Moeck et al., 2017a), and dissolved atmospheric gases (Aeschbach-Hertig and Solomon, 2013; Clark et al., 2005); and 3) tracers of anthropogenic origin (Massmann et al., 2008), including persistent organic micropollutants (e.g., the artificial sweetener acesulfame (Hillebrand et al., 2015, Moeck et al., 2016) and pollutants present in surface and groundwater (e.g., chlorinated solvents (Urresti-Estala et al., 2015). According to Massmann et al. (2008) and Gasser et al. (2014), the application of multiple tracers is required to estimate the typically wide range of residence times and various subsurface processes that result from subsurface heterogeneity which can typically not be identified using one tracer. For instance, Batlle-Aguilar et al. (2017) show how major ions, isotopic tracers and dissolved gases can be used to study flow processes in semi-confined faulted aquifers. They show the advantages of several environmental tracers covering a wide range of residence times and highlight that in highly complex aquifer systems increasing data density is required to accurately characterize the flow system. Althaus et al. (2009) estimate groundwater travel times, mixing ratios and groundwater origin with ^{3}H/^{3}He dating method, supplemented by ^{85}Kr measurements. Müller et al. (2016) show that using multiple tracers is critical to for the final interpretation of a groundwater system where different tracers are applicable at different sections along the flow direction. Clark et al. (2004) applied stable isotopes of water, tritium/helium dating and gas tracers to investigate groundwater dynamics in the vicinity of an artificial recharge facility. They were able to define flow patterns, however, whether the tracer was distributed vertically throughout the entire aquifer or only in layers, acting as preferential flow paths could not be definitely identified. It can be speculated that using more tracers in addition to the applied tracers would help to overcome the difficulties in the interpretation. After Zuber et al. (2011) and Newman et al. (2010) each tracer has specific constrains and complications. These limitations result from differences in the tracers transport characteristics in groundwater and the unsaturated zone. For instance, exchange with immobile zones and interaction with the aquifer matrix as well as unknown input functions and diffusion exchange of gaseous tracers with the atmosphere through processes in the vadose zone can occur. Moreover, natural production in the subsurface and artificial contaminations can make the use of specific tracers challenging (Cook and Herczeg, 2012; Ekwurzel et al., 1994; Plummer et al., 2001).

The objective of this study was therefore to develop a consistent process understanding and to determine the residence times of artificially infiltrated water at an important drinking-water supply site in northern Switzerland using seven different tracers. To date, the interpretation of residence times and subsurface processes that use all the seven aforementioned tracers (dye tracer, heat tracer, major ions, Acesulfame, ^{2}H and ^{18}O, dissolved gases and chlorinated hydrocarbons) have not yet been conducted and therefore a systematic comparison of the value of the information gained from the applied tracers is missing. It is evaluated whether mixing of artificially infiltrated water and water coming from the regional flow path occurred. Therefore, it was important to assess how the infiltrated water was distributed vertically throughout the aquifer. Knowledge about mixing of infiltrated water and water coming from the regional flow path is essential because the highest concentrations of chlorinated hydrocarbons are detected mainly in sampling locations where regional groundwater is present (Moeck et al., 2017a). In addition to the distribution of various water types, the effects of changes in pumping and infiltration rates on the mixing ratio of artificially infiltrated water and that coming from the regional flow path is investigated. The seven tracers are applied and it is indicated which subsurface processes could not be identified when a certain tracer was omitted. The applied tracers were chosen because they have no or very low background concentrations at the study site, behave mostly conservative (nonreactive and no sorption occurs) in the saturated zone and apart from the dye tracer no artificial injection is required. Furthermore, these tracers are relatively inexpensive. After Clark et al. (2005), tracers which can be economically introduced should be used to ensure a sufficient concentration when artificially injected to taken into account the typically large volume of recharge water, as it is the case in the investigated study site (artificial recharge rate ~3.5 e^7 m^3/a). This is even more the case for naturally occurring tracers that do not require artificial injection. To use the tracers under controlled flow conditions, a pumping experiment was carried out in which groundwater was extracted from an aquifer adjacent to landfills and industrial areas. Using this unique dataset, the value of the information gained from the applied tracers was systematically compared and their similarities and differences are illustrated.

2. Materials and methods

2.1. Study site

The study site is located in northwestern Switzerland and has an area of approximately 10 km^2 (Fig. 1). The annual average precipitation and temperature is 730 mm/a and 11.5 °C, respectively. Highest temperature occurs during the months June–August, whereas the lowest