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Experimental determination of noble gases and SF_6, as tracers of CO_2 flow through porous sandstone

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ABSTRACT

In order to label the CO_2 injected underground for geological storage and allow it to be differentiated from natural sources, a panoply of additive chemical tracers have been proposed. Yet, the transport of these tracers relative to CO_2 in pore space is currently poorly constrained. This leads to uncertainty as to whether tracers will act as an early warning of CO_2 arrival, or be preferentially retained in the pore space making them ineffective. Here, we present the factors affecting transport of noble gases and SF_6 relative to CO_2 in a porous rock. Using a porous sandstone core, each of the tracers were loaded into a sample loop and injected as discrete gas pulses into a CO_2 carrier stream at five different experiment pressures (10–50 kPag upstream to ambient pressure downstream). Tracer arrival profiles were measured using a quadrupole mass spectrometer. Significantly, our results show that peak arrival times of helium were slower than the other noble gases at each pressure gradient. The differences in peak arrival times between helium and other noble gases increased as the pressure gradient along the system decreased and the curve profiles for each noble gas differ significantly. The heavier noble gases (Kr and Xe) along with SF_6 show an earlier arrival time and a wider curve profile compared to He and Ne curves through the CO_2 carrier gas stream. This shows that Kr and Xe could be substituted for SF_6, a potent greenhouse gas, in tracer applications. For comparison, CO_2 pulses were passed through a N_2 carrier gas resulting in significantly slower peak arrival times compared to those of noble gases and SF_6. Hence, all investigated tracers when co-injected with CO_2 could potentially act as early warning tracers of CO_2 arrival, though we find that Kr, Xe and SF_6 will provide the most robust advance warning. Analysis of our experimental results shows that they cannot be explained by a simple one dimensional flow model through a porous medium. We outline a conceptual model that incorporates different preferential flow paths depending on flow velocities of individual gas streams. This model can explain the observed dataset and shows that the flow of noble gases and SF_6 tracers is influenced by pore-scale heterogeneity.

1. Introduction

Increases in the atmospheric concentration of greenhouse gases and aerosols alter the energy balance of the climate system (IPCC, 2015). CO_2 is the second most abundant greenhouse gas in the Earth's atmosphere after H_2O and a major contributor to radiative forcing. The primary source of the increased atmospheric concentration of CO_2 since the preindustrial period is from the combustion of fossil fuels, a significant source of global energy for decades to come (Haszeldine, 2009). Thus it is imperative that the levels of CO_2 emitted from fossil fuel combustion are significantly reduced, an essential goal to limit the increase in global average temperature to below 2 °C above pre-industrial levels (UNFCCC, 2015).

Carbon Capture and Storage (CCS) is the only currently available technology that can directly reduce the emissions from power generation and industrial point sources. Combined with biomass combustion (BECCS) it also offers the potential of net removal of CO_2 from the atmosphere (Azar et al., 2010). Industrial scale carbon capture and storage technologies rely on the secure long term storage of CO_2 in the subsurface (Miocic et al., 2016). The engineering, long-term safety and social licence to operate a geological storage site is critically dependent on how secure CO_2 retention is assessed and communicated. Hence, there is a need for robust monitoring regimes, which can detect any unplanned migration of CO_2 during storage. The unplanned migration of CO_2 does not necessarily imply that the storage site has failed, thus there is a difference between migration and leakage. Leakage implies that during migration, the CO_2 enters environmentally sensitive regions (e.g. groundwater aquifers, or escaping to the surface) where it may result in damage to the environment, human health or release back into the atmosphere. Migration means the movement of CO_2 out of the

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Nomenclature

μ	dynamic viscosity of the fluid
A	pore area
D	pore diffusion coefficient
D_x	longitudinal apparent diffusion constant
g_f	density of the fluid
i	hydraulic head/gradient
K	hydraulic conductivity
k	intrinsic permeability
L	characteristic length
M_0	mass injected
P	pore perimeter
n_e	effective porosity
q	specific discharge
Re	Reynolds number
t	time
v	advective velocity
v_f	velocity of the fluid
x	flow path length
α	dispersivity
α^*	non-linear flow parameter (Forchheimer equation)
β	turbulence factor (Forchheimer coefficient)
γ	pore geometry
ρ	fluid density

intended subsurface storage site, but that CO_2 is still retained in the subsurface and does not leak to the surface. Monitoring a CO_2 storage site before, during and after injection is crucial part of the regulatory framework for CCS. Continuous monitoring of a site and the surrounding environment minimises the potential risk of leakage of CO_2, as well as improving the opportunity to mitigate any changes that may occur over time, helping with the social licence to operate the storage site (Haszeldine, 2009; Scott et al., 2013).

The term 'chemical monitoring' in application to CO_2 storage refers specifically to a substance (solid, liquid or gas) that can be measured to characterise it before, during and after the monitoring process. A chemical tracer can be any substance used to understand the physical movements of fluid through a system. It has been shown that chemical tracers can be complementary to physical monitoring methods (Myers et al., 2013). The majority of tracer applications within CCS are related to either understanding the subsurface, quantifying the trapping capacity or determining containment and leakage rates for monitoring (Myers et al., 2012). The main advantages of chemical tracers are that they can be used to make measurements that are difficult to access physically and can cover large scales. Monitoring injected CO_2 can be challenging, as it is a reactive compound, highly soluble and its properties are dependent on the pressure and temperature of the system. Thus, the use of non-reactive conservative chemical tracers that have limited interactions with the injection fluids and surrounding reservoir, enable the tracking of fluid pathways. Such tracers include synthetic chemicals such as SF_6 and perfluorocarbon tracers (PFTs), isotopic labelled gases and noble gases. Such tracers have been used to track the movement and fate of injected CO_2 in a variety of CO_2 injection experiments, injecting Frio, Otway and Cranfield (McCallum et al., 2005; Jenkins et al., 2012; Lu et al., 2012).

Noble gases (He, Ne, Ar, Kr and Xe) exist in trace amounts in geological environments and they can be used as tracers within natural CO_2 reservoirs (Gilfillan et al., 2008; Gilfillan et al., 2009; Zhou et al., 2012; Sathaye et al., 2014) and for CO_2 used in EOR projects (Nimz and Hudson, 2005; Györe et al., 2015; Györe et al., 2017). As such, small amounts of noble gas blends can also be intentionally added to the CO_2 being injected for storage or be measured in the captured CO_2 stream and used as tracers to monitor CO_2 movement (Hovorka et al., 2013; Flude et al., 2016; Flude et al., 2017). This distinct mixture of noble gas isotopic compositions can be identified during monitoring as being associated with the injected CO_2. To investigate the potential role of noble gases as tracers for CO_2 storage the physical behaviours of noble gases and CO_2 within the porous media needs to be fully understood.

Furthermore, recent work on the flow of N_2 and brine through a permeable sandstone at subsurface reservoir conditions has shown that Darcy flow conditions are not maintained and instead the flow undergoes 'dynamic connectivity' (Reynolds et al., 2017) emphasising that we do not yet fully understand the complexity of fluid flow through 'real world' heterogeneous porous media.

A common method to observe the behaviour of fluids under laboratory conditions is to carry out one dimensional flow through column experiments; these columns can be built from cores of natural rock or artificially constructed using materials such as packed silica beads (Edlmann et al., 2013; Liu et al., 2014). In this study a specially constructed flow cell was designed and built to investigate how noble gases could be used as tracers for CO_2 migration in storage sites. From this equipment, experimental breakthrough curves for noble gases travelling through a sample of porous sandstone in relation to CO_2 over pressure gradients of 10, 20, 30, 40 and 50 kPa were generated (10–50 kPag upstream to ambient pressure downstream) at laboratory temperature. For comparative purposes the same experiments were conducted for SF_6, another popular fluid tracer. Here, we present the results from these experiments, which were then modelled using a one dimensional advective dispersion transport equation. The outcome of these results is then explained in terms of a preferential pathway system, where the relative contribution of the flow paths depend on comparative flow velocity differences.

2. Experimental methods

2.1. Sample characterisation

A representative sample of porous media was required to characterise the transport behaviours of noble gases, SF_6 and CO_2 under the same experimental conditions. A sample of sandstone was then selected. The sample was quarried at South Charlton, near Alnwick in Northumberland and is commercially referred to as the Hazeldean sandstone (Hutton Stone *Co.* Ltd., 2013). The sandstone is part of the Fell Formation that is Chadian to Holkerian (Carboniferous) in age (British Geological Survey, 2016). The sandstone is massive and it was selected due to its high quartz content (~95%) and the low carbonate content (trace amounts).

The Fell sandstone sample was subjected to analysis prior to the commencement of experiments. When the trace minerals were discounted, XRD results indicate that the Fell sandstone is mostly comprised of quartz (95%), with minor feldspar and clay minerals and is a quartz arenite, as previously classified (Bell, 1978). A 3.8 cm × 7 cm cylindrical plug was used to identify the porosity and permeability of the sandstone. The porosity was determined using a helium gas expansion porosimeter. The permeability was determined using a nitrogen gas permeameter. Initial laboratory testing of the sandstone provided a porosity of 20.3% and a permeability of 221.33 mD.

Understanding the pore network of the Fell sandstone is advantageous when evaluating the mechanisms involved in the transport of noble gases, SF_6 and CO_2 tracers. Hence, extensive mapping of the pore network connectivity of the sample from images was conducted. The pore network, distribution and interactions within the rock were investigated using image analysis (via ImageJ at https://imagej.nih.gov/ij/index.html).

Data from four thin sections are presented using this image

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