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Hydrogen storage in porous geological formations – Onshore play opportunities in the Midland Valley (Scotland, UK)

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Abstract

Hydrogen usage and storage may contribute to reducing greenhouse gas emissions by decarbonising heating and transport and by offering significant energy storage to balance variable renewable energy supply. Underground storage of hydrogen is established in underground salt caverns, but these have restricted geographical locations within the UK and cannot deliver the required capacity. Hydrogen storage in porous geological formations has significant potential to deliver both the capacity and local positioning. This study investigates the potential for storage of hydrogen in porous subsurface media in Scotland. We introduce for the first time the concept of the hydrogen storage play. A geological combination including reservoir, seal and trap that provides the optimum hydrogen storage reservoir conditions that will be potential targets for future pilot, and commercial, hydrogen storage projects. We investigate three conceptual hydrogen storage plays in the Midland Valley of Scotland, an area chosen primarily because it contains the most extensive onshore sedimentary deposits in Scotland, with the added benefit of being close to potential consumers in the cities of Glasgow and Edinburgh. The formations assessed are of Devonian and Carboniferous age. The Devonian storage play offers vast storage capacity but its validity is uncertain due to due to a lack of geological data. The two Carboniferous plays have less capacity but the abundant data produced by the hydrocarbon industry makes our suitability assessment of these plays relatively certain. We conclude that the Carboniferous age sedimentary deposits of the D'Arcy-Cousland Anticline and the Balgonie Anticline close to Edinburgh will make suitable hydrogen storage sites and are ideal for an early hydrogen storage research project.

Introduction

The use of hydrogen can make significant contribution to reducing greenhouse gas emissions in line with the international climate protection agreements to decarbonising the economy and the society [1]. For the UK, the use of hydrogen as a fossil fuel substitute opens a range of opportunities to achieve these goals. Together with other energy decarbonised and emission reducing technologies such as CCS and renewable energy generation, hydrogen can become an essential vector in a clean future energy system [2–5]. Hydrogen can contribute to reduce the emissions from fossil fuel-based energy sources in three energy sectors; transport, heating and energy storage. This study focusses on the storage of hydrogen in subsurface formations for medium to large scale inter-seasonal energy storage. We have allocated a relative value to assess the scale of the potential storage sites. A saline aquifer with storage greater than a mega tonne of hydrogen is a "very large" storage play, capable of supplying the energy needs of a large town. A medium storage play is one comparable with a small oil and gas field or a large salt cavern with a hydrogen working gas capacity of 2-3Mkg, which would supply a small town annually, with a population less than 10,000. As a reference Glasgow has an annual energy use of 10,892GWh [6].

Energy storage is increasingly vital for energy security becoming even more important as the distribution of carbon-based energy moves towards renewable electricity, such as wind and solar. Hydrogen from renewable energy offers a unique opportunity for the UK's clean energy future [2,4,7]. One of the most significant challenges for renewable energy is the imbalance between supply and demand. One way to tackle this imbalance is to store energy generated during periods of renewable energy oversupply and release it in times when demand exceeds supply. Large-scale energy storage is currently undertaken through rechargeable options such as pumped hydroelectric plants and batteries. Pumped hydroelectric plants provide the greatest rechargeable storage capacity in the UK to date for centralised energy storage [8]. However, their installation cost, dependence on specific and limited geography, and remoteness preclude then from becoming a major contributor to large-scale future energy storage. Batteries will certainly have the potential to store large amount of energy in the near future. Batteries contain expensive and often rare metals and their lifetime is limited [9] and the technology development required for large capacities of energy storage batteries at interseasonal level is not yet available. Additionally, the current worldwide race to increase the number of electric vehicles on the streets makes gigantic battery farms for seasonal energy storage unlikely.

Hydrogen is commercially generated using two methods; thermochemical processes releasing hydrogen from hydrocarbons and electrolysis releasing hydrogen from water. Hydrogen production from steam methane reformation, is a well-established industry process with an energy efficiency of 65-85 % [10]. Cost of hydrogen from steam methane reformation is cheap, currently about 15-20% of the cost of hydrogen from electrolysis. However, significant CO_2 is generated during this process. If the emitted CO_2 was released into the atmosphere, the process would make little sense from an environmental and economical point of view and hence an additional CO_2 (carbon) capture and storage (CCS) program is required to decarbonise this energy system. The main benefit of this process can be demonstrated if the hydrogen is used as a substitute for natural gas in industrial, or especially in domestic heating. Domestic heating based on burning natural gas emits CO_2 into the atmosphere and it is currently not possible to capture this CO_2 because of too many very small emitters. If the natural gas is replaced by hydrogen and the hydrogen is generated by steam methane reforming combined with CCS, the heating system would no longer emit CO_2 into the atmosphere. From a CO_2 emission reduction perspective, the creation of hydrogen from the electrolysis of water using renewable energy is preferred and regarded as the future of hydrogen usage (EPSRC). Hydrogen generation using electrolysis is already finding applications at small-scale projects and the commercial production achieving an energy efficiency of 55-75 % [4,10]. Wind power is one of the fastest growing renewable energy sectors with 539,581 MW of installed wind turbines worldwide and 18,872 MW in the UK [11]. However, wind energy is variable and does not inherently correlate with demand. Hydrogen production via water electrolysis using surplus wind energy with its high-energy yield ratio of up to 70 [12] can enable a wider utilisation of wind energy, offset costs and balance supply variability. Significant experience has been gained through a number of small-scale renewable-hydrogen systems [13-16]. The use of hydrogen offers a feasible clean energy storage option. Excess renewable energy can be transformed to hydrogen through the electrolysis of water during periods of oversupply and stored in the subsurface, until it subsequently can be back-transformed to energy during periods of energy shortage. Considering a Spanish wind farm estimated a yield of 40% of excess energy generation from wind capable of regeneration into the grid using hydrogen [17]. This could provide energy storage of a scale comparable to the Rough Field, once the UK's biggest natural gas storage facility, reduce the greenhouse gas emissions of the energy sector, and therefore present a future way of preventing energy shortages, net instabilities and shutdowns.

The UK is already conducting and planning hydrogen projects at a number of different scales. These include the Leeds City Gate H21 Project [18], which aims to implement the conversion of natural gas fuelled heating to hydrogen heating for the city of Leeds, with a population of 650,000 one of the largest cities of Northern England. In Scotland, the Hydrogen100 project is even more advanced and will commence test conversion of domestic properties during 2018, and will pilot onshore electrolytic hydrogen generation and storage from wind power before 2020 [19]. And the Aberdeen Hydrogen Bus Project, which is designed to contribute to Scotland's green transport targets by introducing hydrogen fuelled busses [20]. The smaller projects tend to generate hydrogen using electrolysis whereas larger scale projects, such as H21 in Leeds, plan to use steam methane reformation.

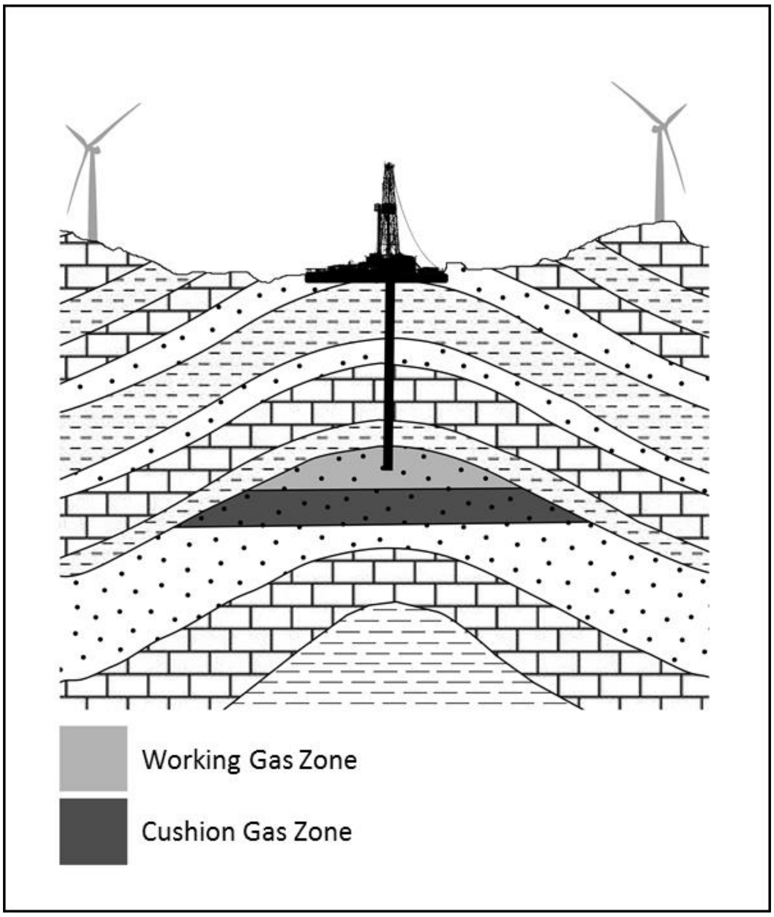

Figure 1: Simplified hydrogen storage scenario in a brine filled reservoir formation (dotted pattern) underneath an impermeable shale-rich sealing formation (dashed pattern). For simplicity, the injection well also acts as the production well. The anticlinal shape of the structure prevents the hydrogen form migrating away from the well. The reproducible gas ('working gas') as well as the cushion gas volume are highlighted.

If large scale hydrogen usage projects are to become truly sustainable using hydrogen generated through hydrolysis from excess renewable energy, the storage of hydrogen is likely to become a limiting factor. Small volumes of hydrogen can be stored in surface tanks; their size is limited due to cost and safety. Medium scale volumes of hydrogen can be stored in salt caverns. Currently, three hydrogen storage projects in salt caverns are operational; two in the US ('Clemens', working gas capacity: 2.56 Mkg; 'Moss Bluff', 3.72 Mkg) and one plant in the UK ('Teesside', 0.83 Mkg) [21]. In addition to their limited capacity and their high cost, suitable onshore salt formations are not present to the north of Teesside. Since hydrogen storage in salt caverns offshore is very expensive, a different storage approach must be considered for other regions of the UK, including Scotland. Hydrogen storage in geological porous media is the most suitable alternative in which hydrogen is stored in deep geological formations in a similar manner to natural gas or CO_2.

The literature of hydrogen storage in subsurface porous media is relatively sparse compared to hydrocarbon or CO_2 storage in the subsurface. Lewandowska-Smierzchalska et al. [22] ranked potential subsurface reservoirs and salt caverns based on an Analytic Hierarchy Process for hydrogen storage in onshore Poland. According to their analysis, the most important criteria for suitable hydrogen porous media storage sites are tectonic activity and the overburden lithology and we agree with them that a detailed analysis of the regional geology to understand these to aspects is crucial. Other work on hydrogen storage in porous media has been presented by Tarkowski [23], who introduced potential hydrogen storage sites in Poland based on their geological setting, and Amid et al. [24], who investigated potential hydrogen storage in depleted gas fields. In this paper, we offer a more practical approach to assess potential hydrogen storage sites in subsurface porous media. We introduce for the first time the concept of the 'hydrogen storage play', a geological assembly consisting of suitable reservoir, seal and trap structures. We then apply this to the Midland Valley of Scotland, identified as the most promising location for hydrogen storage in Scotland, which includes Edinburgh and Glasgow, the two most populated cities in Scotland. This study considers three potential hydrogen storage plays because of their similarity to established storage reservoirs for natural gas.

Hydrogen storage in subsurface porous media

The concept of hydrogen storage in subsurface porous media consists of several steps essentially comprising injection, storage, and withdrawal, of hydrogen (Figure 1). For this study we do not consider the source of the hydrogen or its ultimate use and we assume the hydrogen will be transported via pipelines or in tanks on lorries and ships to the storage site. The hydrogen will be injected into a porous and permeable reservoir formation via injection wells. In the reservoir, the injected hydrogen will displace the in-situ pore fluid, usually brine, and spread out underneath an impermeable seal due to its lower density. A trap structure, such as an anticline, will contain the hydrogen and prevent it from migrating away. When the energy demand requires it, the hydrogen will be produced via production wells directly from the reservoir. A share of the hydrogen, often referred to as cushion gas, will remain in the reservoir as a precaution to maintain an operational pressure and to minimise subsurface water encroachment into the working reservoir during withdrawal periods. Additionally, hydrogen will be lost due to a process defined as residual trapping in CO_2 storage research [25-27]. Small isolated hydrogen bubbles will remain in the pores and cannot be removed; these are residually trapped. Hence a share of the injected hydrogen initially injected will be lost, likely to be in the range of 55%, based on estimates for natural gas storage saline aquifers according to Le Fevre [28]. It should be mentioned that the majority of the cushion gas hydrogen and residually trapped hydrogen loss happens during the first injection and withdrawal cycle. Once the pore space is saturated with hydrogen, all hydrogen injected during later cycles will be recoverable, with minimal loss to cushion gas. There is very limited research on cushion gas for hydrogen storage but future research will show how much is required, how much will be lost. In particular, it may be that other fluids, such as nitrogen or CO_2, are more suitable as cushion gas, at much less cost and similar functioning performance [3].

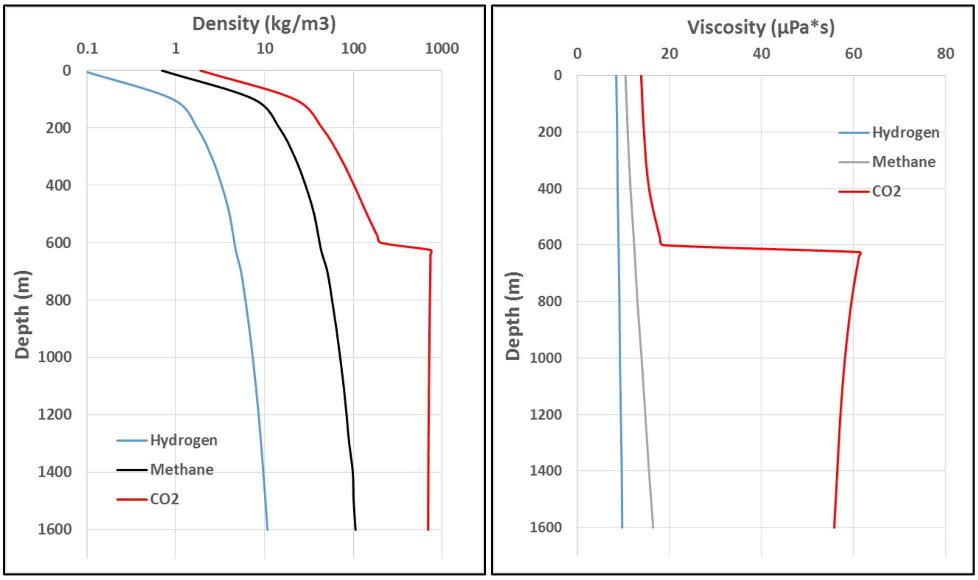

Figure 2: Density and viscosity changes of methane, CO_2 and hydrogen with depth calculated using the NIST database [29]. The geothermal gradient is based on data mainly taken from the Midland Valley region [30] and a pressure gradient based on a freshwater pore fluid density of 1g/cm^3.

Gas storage is not a new concept, and natural gas and CO_2 storage are important analogues for hydrogen storage in subsurface porous media. However, there are important operational differences, unique to hydrogen, which have to be taken into account:

- Natural gas storage is usually conducted in depleted hydrocarbon fields. This leads to two important advantages: Firstly, the remaining in-situ hydrocarbons can be used as cushion gas which decreases the initial costs; secondly, depleted hydrocarbon fields have proven their ability to retain buoyant fluids over geological timescales. Hydrogen is more diffusive, has a lower viscosity and a lower density than natural gas and is therefore more likely to leak. Hence a seal which managed to retain hydrocarbons over geological timescales does not automatically retain hydrogen. However, the presence of buoyant hydrocarbon fluids can act as a first confirmation for appropriate seal quality.
- When hydrogen is stored in the presence of hydrocarbons, there is the possibility of reactions between hydrogen and micro-organisms, and between hydrogen and the host rock or other fluids [31,32]. Potential biological reactions could consume hydrogen or could lead to precipitation reactions, reducing injectivity close to the wells.
- The minimum depth for hydrogen storage depends on the hydrogen storage play. If a suitable reservoir, trap and seal assemblage can be found a shallow depths and to access it is technically feasible, the storage operation is theoretically possible. For CO_2 storage, a minimum depth of 800m is required due to its phase behaviour [33-35]. This phase change, although it occurs at 200m burial, does not lead to significant property changes of hydrogen (Figure 2). However, as the confining pressure increases with depth, the fracture and rock permeability is generally lower in deeper formations and hence seals are less likely to allow leakage at greater depth.
- Hydrocarbon fields have proven their ability to retain buoyant fluids over geological timescales and are therefore targets for CO_2 storage operations, where CO_2 must remain in the subsurface for at least several thousands of years. The timescales for hydrogen storage are shorter, with approximately one full injection and production cycle per year.

Although hydrogen is much more capable of leakage, it actually has much less time to do so. In the case of hydrogen leakage out of the storage site, the hydrogen still in place could quickly be back-produced.

- The energy content of hydrogen is 2.5-3 times higher by weight than natural gas, giving it the highest energy content per unit mass of any fuels, but has a lower density than natural gas7 [6]. At reservoir conditions however, the density of hydrogen is roughly ten times lower than that or natural gas, meaning that approximately 3-4 times the storage space is required to store the same amount of energy with hydrogen, compared to natural gas in the subsurface (Figure 2).

There is no operational pure hydrogen storage project in operation in saline aquifers or depleted hydrocarbon fields [21]. The three hydrogen storage projects operational today store pure hydrogen in salt caverns [21]. Nevertheless, the transferable lessons learned from CO_2 storage and natural gas storage are of vital importance and provide hydrogen storage pilot projects in porous media with a crucial advantage.

Hydrogen storage plays

The term 'storage play' is derived from the hydrocarbon industry term 'petroleum play'. According to Allen & Allen [36] a petroleum play is a "model of how a producible reservoir, petroleum charge system, regional topseal, and traps may combine to produce petroleum accumulations at a specific stratigraphic level". Bennion et al. [37] suggests that to be a candidate for gas storage there must be sufficient volume to allow storage without exceeding containment pressure, or requiring compression; satisfactory containment by sealing formations, suitable permeability to allow injection and withdrawal at required rate and limited sensitivity to permeability reduction. Any hydrogen storage play will require a reservoir with sufficient volume, porosity and permeability, a regional seal and a trap to allow the hydrogen to accumulate and allow safe storage for a specific duration at a specific stratigraphic level. The 'reservoirs' are predominantly composed of sedimentary rocks, such as sandstone. For buoyant fluids to be trapped within a reservoir, it is required to be overlain by an impermeable layer, or 'seal', such as a mudstone. These sedimentary reservoir and seal successions are very common, and can accumulate in many different depositional settings over geological timescales. Lastly, a trap is needed to stop the fluids from leaking laterally within the succession. These are often either trap structures (e.g. a fold in the strata), which are most commonly formed after deposition of the sediments during tectonic events, or sedimentary traps of lateral thickness change of the porous reservoir (e.g. sedimentary pinchout) which usually derive from facies changes during the deposition process. Storage plays are simpler than hydrocarbon plays, which require the additional complexity of an organic rich source rock capable of generating gas or oil within the correct geological time window so that the fluid migrates into a trap, in addition to the reservoir, seal and trap. Thus, hydrogen storage sites should be more common in the subsurface. Similar to petroleum plays, storage plays for hydrogen require geological models, which need to be improved and tested by research and experimentation, and ultimately confirmed by actual industrial campaigns.

Hydrogen storage in the Midland Valley

Most of onshore Scotland consists of thick successions of metamorphic rocks, such as the Neoproterozoic Moine Schist to the north of the Great Glen Fault and the Dalradian Supergroup to its south [38]. These rocks originated as sedimentary deposits, but they have been extensively altered by successive tectonic events and are considered to have no matrix permeability and therefore uneconomic for hydrogen storage. Scotland also contains of a large quantity of igneous intrusions and volcanic successions that are also considered uneconomic for hydrogen storage because of the complexity of their formation and shape within the subsurface, poor to absent matrix permeability, and extreme potential for diagenetic alteration. Additionally, there is relatively little data available about the internal structures, such as potential traps and zones of higher porosity, in part because they do not normally contain hydrocarbons and have therefore not previously been investigated for these attributes. However, the discovery of the Lancaster oil field within fractured basement offshore in the West of Shetlands, and other test cases in the UK, may eventually provide sufficient data to allow gas storage prospects to be considered within metamorphosed or igneous rocks in the UK [39].

The most obvious region for hydrogen storage in Scotland is within the extensive sedimentary deposits of the Midland Valley in the south of Scotland. Scotland's two main cities, Glasgow, and Edinburgh, are situated within the Midland Valley, so potential storage sites are close to numerous consumers. Furthermore, the Midland Valley is an area with a long tradition of coal exploration, and minor oil and gas exploration, and consequently there is a wealth of readily available data relating to the sediments and the structure of the area. The Midland Valley is rich in shale oil, shale gas and coal bed methane, which are produced from organic-rich sedimentary rocks, such as coal, mudstones, marine band shales, oil shale and fine siltstone and has abundant sandstone reservoirs providing the necessary reservoir and seal couplets for hydrogen storage prospects.

Three potential hydrogen storage plays have been identified in the Midland Valley of Scotland within: (1) the Devonian Stratheden and Inverclyde Groups; (2) the upper part of the Carboniferous Strathclyde Group; and (3) the lower part of the Carboniferous Clackmannan Group (Figure 3). Historically many of the named formations comprising these groups have been known by various lithostratigraphic names (e.g. Cameron and Stephenson [40]). This is generally unhelpful as the same laterally extensive geological unit can have different names in geographically adjacent localities leading to a confusing and complicated regional appraisal. Here, we use the most up-to-date, regionally appropriate stratigraphic nomenclature in line with the British Geological Survey [41-43].

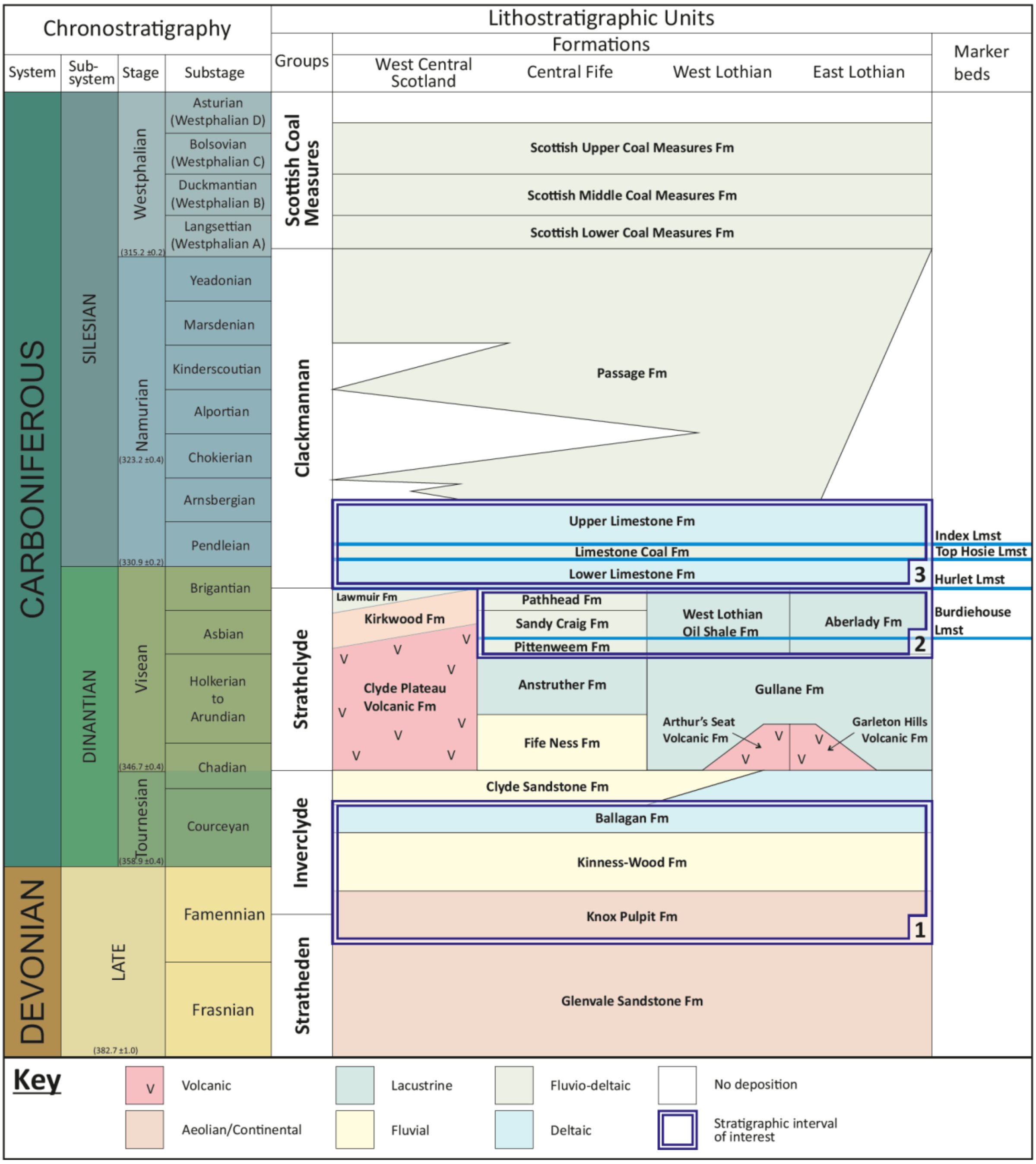

Figure 3: Lithostratigraphic nomenclature and stratigraphic succession. Also shown is the level of the three selected storage plays (blue boxes) discussed in the text from oldest to youngest. Modified from [41-44].

Geological history of the Midland Valley and the generation of structural traps for hydrogen storage

The evolution of the Palaeozoic Midland Valley is bookended by two tectonic events: the Caledonian Orogeny and the Variscan Orogeny. Geologically, the Midland Valley is an elongate, fault-bounded, WSW-ENE trending, Late Palaeozoic sedimentary basin formed after the Caledonian orogeny due to the closure and subduction of the Iapetus Ocean in the UK [45-47]. The basin is bounded to the north by the Highland Boundary fault (and the Southern Highlands metamorphosed Dalradian rocks) and in the south by the Southern Upland fault (and the metamorphosed Early Palaeozoic rocks of the Southern Uplands; Figure 4). The juxtaposition of weathering resistant metamorphosed strata to the north and south of the Midland valley with the weaker, unmetamorphosed sediments within the

Midland Valley have created this geographically distinct central lowland belt. The Carboniferous clastic sediments (and probably the Devonian sediments) which infilled the Midland Valley and the North Sea Basin were derived from the mountains of East Greenland, Norway and NE Scotland, that were uplifted during the Caledonian Orogeny [48-51]. The Late Palaeozoic Midland Valley basin was filled by a variety of sediments deposited in differing environmental conditions [52]. During the Devonian the UK was landlocked within a semi-arid continental landmass that lead to the deposition of aeolian desert and fluvial sandstones collectively known as the Old Red Sandstone (ORS) facies [53]. The Upper Devonian part of the ORS facies in the Midland Valley comprises the Stratheden Group which comprises multiple formations across the Midland Valley, here simplified to the Glenvale and Knox Pulpit Formations. These formations are dominated by conglomerates and cross-bedded sandstones that were deposited from braided fluvial river systems. However, aeolian influence becomes more recognisable in younger units, such as the upper part of the Knox Pulpit Formation [54]. The overlying Kinnesswood Sandstone Formation of the Inverclyde Group defines a change back to fluvial deposition [55,56]. It is overlain by the Lower Carboniferous mudstone-dominated Ballagan Formation (Figure 3) of the Inverclyde Group where traces of marine incursions from the east, mainly carbonates, gypsum and anhydrite, have been identified [41,57]. These successions form the basis for storage play 1, discussed below.

Northwards drift of the tectonic plates throughout the Carboniferous resulted in a change from an arid to a sub-tropical climate and instigated the deposition of south-westwards prograding clastic deltas in a dominantly fluvio-lacustrine depositional environment interspersed with periods of marine incursions and volcanism [38]. The Lower Carboniferous sediments of the Strathclyde Group were predominantly deposited in a freshwater lacustrine environment. This palaeo-lake is referred to as Lake Cadell [51], which may have covered an area of between 2000 and 3000 km^2 [50]. The lake was land-locked to the north, south and west; and may have periodically drained towards the south-east where minor marine incursions may also have originated. The sediments that deposited into the lake are dominated by repeated (cyclic) thick, coarsening and cleaning upwards siltstone and sandstone successions that represent prograding non-marine clastic deltas which alternate with subordinate black shale (sometimes oil shale) and thin limestone horizons [50]. These successions form storage play 2, discussed below.

During the early part of the Upper Carboniferous (Namurian) in the Midland Valley, sedimentary rocks belonging to the Lower Limestone Formation, Limestone Coal Formation and the Upper Limestone Formation of the Clackmannan Group were deposited (Figure 3, 4 & 5). Frequent fluctuations in eustatic sea level and localised extensional tectonics led to relative sea level rises which triggered marine incursions that deposited mudstones and limestones which can be laterally extensive, sometimes being correlated across many tens of kilometres [40]. For example, the Hurlet Limestone (Figure 3), which marks the boundary between the Strathclyde Group and Clackmannan Group in the Midland Valley, can be correlated with the Oxford Limestone of the Tweed and Northumberland Basins to the south [42,58,59]. Fluvio-deltaic clastic sediments interrupt the marine rocks resulting in cyclic successions of reservoir and seal couplets. These successions form the basis for storage Play 3, discussed below. The number of marine incursions decreased throughout the Upper Carboniferous Namurian and Westphalian [42,60]. As the UK continued to move northwards into a tropical environmental belt, deposition was dominated by fluvial-deltaic rocks and coal, leading to the development of the Scottish Coal Measures Group that eventually drove the industrial revolution [61].

Carboniferous and later volcanism has affected the Midland Valley deposits with numerous extrusive igneous and volcaniclastic deposits together with significant intrusive igneous activity in the form of dykes and sills. The effect of the igneous activity on the reservoir quality is unknown. However, in other regions intrusive igneous activity can lead to a loss of reservoir quality due to the circulation of hot, mineral laden hydrothermal waters which choke pore space with precipitated minerals [62]. Additionally, it is not well understood if the igneous dykes and associated steep margin-parallel fractures act as vertical conduits for fluid flow (or, in this case hydrogen flow) or significant horizontal baffles to flow.

The culmination of the Variscan Orogeny, which led to the amalgamation of Pangaea at the end of the Carboniferous, ended Palaeozoic sedimentation in the Midland Valley basin. A phase of compressional deformation associated with the Variscan orogeny led to folding and minor reverse faulting of the sediments in the Midland Valley [44,54]. The structures generated during the Variscan compression are the main source of traps for potential hydrogen storage plays and pre-existing hydrocarbon fields. No significant phases of deformation have affected the Midland Valley during the Mesozoic-Paleogene, despite numerous events recorded in the North Sea Basin. Neogene tilting of the UK south-eastwards towards the present North Sea caused significant uplift and erosion over Scotland where up to 2 km of strata may have been eroded [63]. Geologically recent glaciation events in the past 2Ma have overprinted Scotland, including the Midland Valley [64].

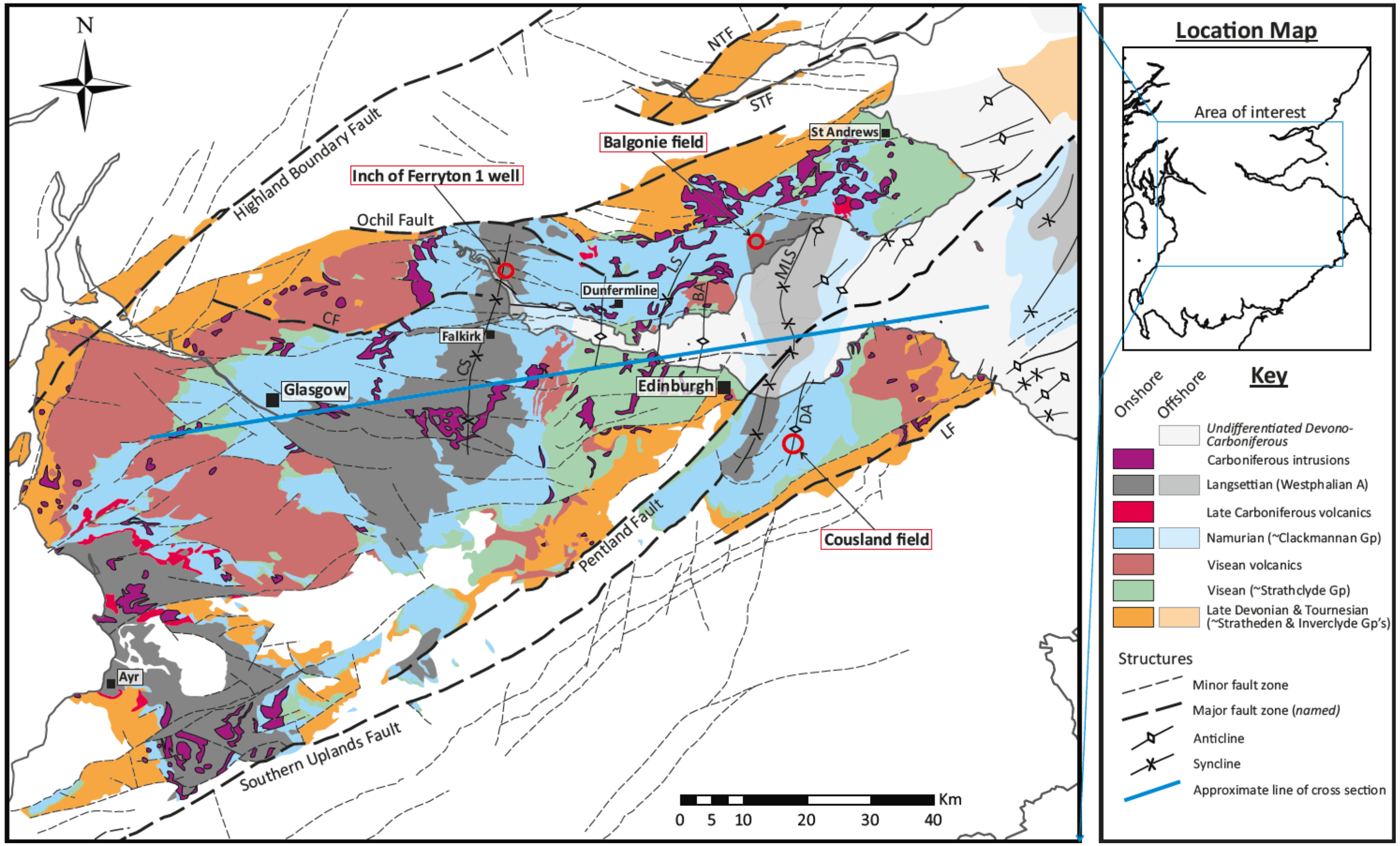

Figure 4: Location map and simplified geological map of the Midland Valley of Scotland. The basin is bound to the north by the Highland Boundary Fault and to the south by the Southern Uplands Faults. Other major and minor faults together with major anticlines and synclines are indicated (including: CF, Campsie Fault; NTF, North Tay Fault; STF, South Tay Fault; LF, Lammermuir Fault; CS, Clackmannan Syncline; LS, Lochore Syncline; BA, Burntisland Anticline; MLS, Midlothian-Leven Syncline; DA, D'Arcy-Cousland Anticline). The location of the cross section in Figure 5, the Inch of Ferryton 1 well (Figure 6) and the location of the Balgonie Oil Field (Figure 7) are highlighted. Compiled from BGS data and inspired by Monaghan [43] and Underhill et al. [44].

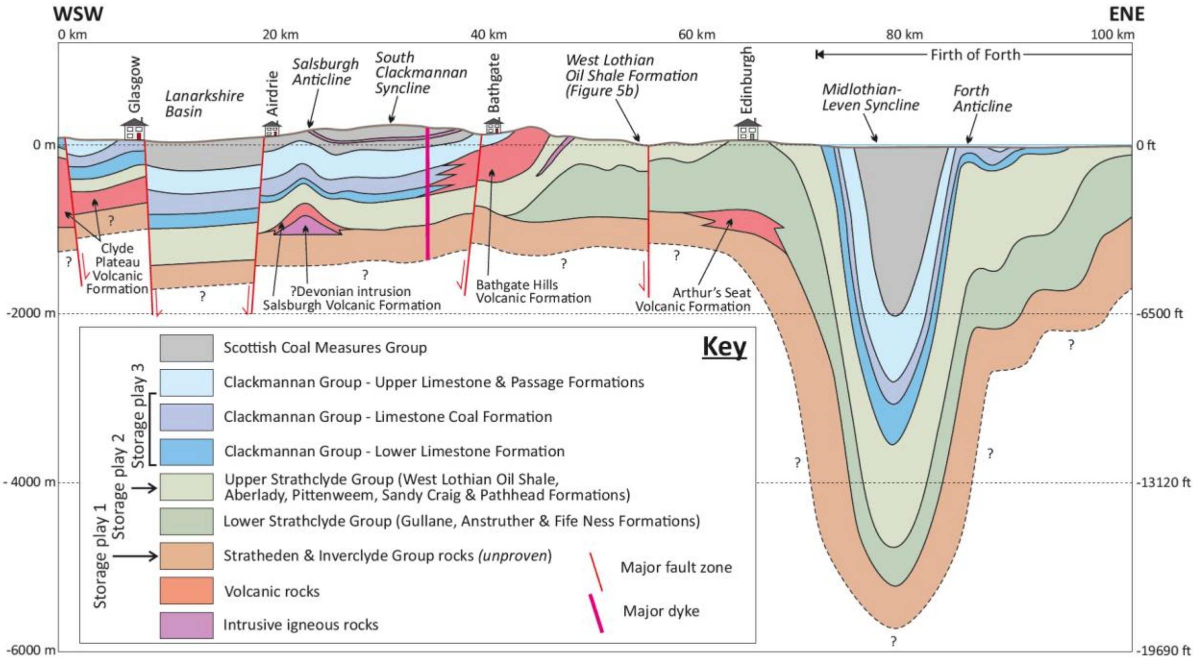

Figure 5: Simplified cross section through the Midland Valley showing some of the Groups and Formations that constituent the Palaeozoic fill of the basin. Adapted from Managhan [43].

Midland Valley hydrogen storage plays

None of the storage plays outlined below have been tested for hydrogen storage, but are recommended to be considered for investigation because of their similarity to established hydrocarbon plays.

Storage play 1: The Devonian Stratheden and Inverclyde Group

The reservoirs are good quality aeolian sandstones, overlain by a seal of thick mudstone and evaporites, with traps of thrusted and inverted late Carboniferous anticlines.

The most suitable Upper Devonian storage reservoirs are sandstones from the Knox Pulpit and Kinnesswood Formations (Stratheden and Inverclyde Groups, respectively), which are well developed in the east of the Midland Valley, although with significant lateral depositional variability. The total thickness of the Upper Devonian sandstones varies; northern outcrops show more than 1000 m thickness in the west [65], 600 m in Stirlingshire [66] and between 350 m and 440 m in Fife and Kinross [67]. In the south of the Midland Valley, thicknesses vary from 100-425 m in Ayrshire [68] and 640 m in the Edinburgh area [69]. The Knox Pulpit Formation reaches 130-180 m thick in Fife but is absent under the Edinburgh area [57]. The Kinnesswood Formation ranges from 100-200 m but up to 640 m in the Edinburgh area [41]. The sandstones outcrop to the north and the south of the Midland Valley. Additionally, boreholes, such as the Inch of Ferryton 1 well (Figure 6), and seismic survey analysis [70], have confirmed significant thicknesses of the Stratheden and Inverclyde Groups in the subsurface (Figure 5), hence the Upper Devonian sandstone reservoir rocks are expected to extend in the subsurface between the north and the south of the Midland Valley.

The Knox Pulpit Formation comprises white to buff coloured, fine- to coarse-grained feldspathic sandstones (dominantly quartz with minor feldspar and other detrital grains) that were deposited in an arid aeolian environment [41,70]. Although very different in age, the formation is comparable in reservoir type to the aeolian reservoirs of the prolific gas-bearing sandstone reservoirs of the Permian Rotliegend Group in the Southern North Sea. The Kinnesswood Formation is mineralogically similar to the Knox Pulpit Formation. However, it was primarily deposited from fluvial rivers and becomes finer grained with caliche-bearing desert soils stratigraphically higher, potentially causing lateral connectivity and vertical permeability challenges [41]. The mean horizontal permeability of the Knox Pulpit Formation, calculated assuming the data is normally distributed, ranges from 60-80 mD [66,71].

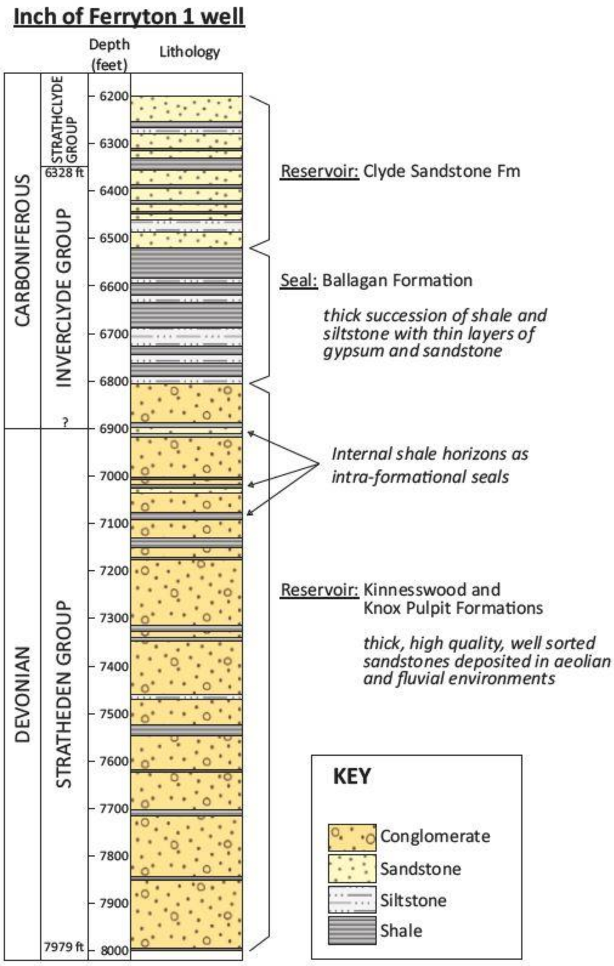

Figure 6: Reinterpreted stratigraphy of the "Inch of Ferryton 1" well. See Figure 4 for location. The Devonian Sandstone consist of sandy conglomerates which could act as reservoirs for stored hydrogen. The overlying Ballagan Formation of the Inverclyde Group could act as regional seal. Additional, thinner Devonian mud layers could act as regional seals.

The most promising regional-scale seal is the Lower Carboniferous (Courceyan) Ballagan Formation of the Inverclyde Group, which overlies the fluvial sandstone deposits of the Kinnesswood Formation except in the Lomond Hills in Central Eastern Fife where it has been eroded [70]. The Ballagan Formation dominantly comprises grey mudstones and siltstones with minor, thin sandstones deposited in lacustrine and lagoonal environments [52]. Minor marine gypsum and anhydrite layers occur that were deposited by short-lived marine incursions from the east, and may also form laterally extensive seals. Porosity in the Ballagan Formation has been reported as very low (mean <2%) and mainly represented by microporosity [70] and may therefore represent a potentially good seal rock. The Ballagan Formation is laterally extensive and its thickness varies from 1000 m in Berwickshire down to 300 m near Fintry [52]. The Ballagan Formation is overlain by the Clyde Sandstone Formation, the youngest sediments of the Inverclyde Group, both formations have been eroded over much of the southern part of the Midland Valley [52].

The geological uncertainties for this storage play are significant. The main source of information about the Upper Devonian Sandstone comes from outcrop; only a few wells have been drilled into the Devonian stratigraphy and hence the lateral extent and connectivity of potential sandstone reservoirs remains uncertain. The sandstones are reported to have undergone a high degree of mechanical compaction and patchy dolomitic diagenesis (reducing primary porosity), although secondary porosity may have been generated from the dissolution of feldspar grains [70]. No hydrocarbons have been found in the Old Red Sandstone facies in the Midland Valley and hence the seal quality of the Ballagan Formation (and potential internal shale layers within the Stratheden Group) is not confirmed. Where the Ballagan Formation is absent alternative seals need investigating. The overall structure and the existence of geological traps within the Upper Devonian Knox Pulpit and Kinnesswood Formations is unknown. However, given the suitable thickness and potential lateral extent of the Kinnesswood and Knox Pulpit group reservoir facies, with permeabilities in the region of 60-80mD and the extensive Ballagan formation seal, the Upper Devonian Stratheden and Inverclyde Group represent a significant hydrogen storage opportunity.

Storage play 2: The Lower Carboniferous Upper Strathclyde Group

This play comprises coarse arkosic fluvial and delta front sandstones as reservoirs, sealed above, below and laterally by enclosing shales and oil shales of very low permeability, forming stratigraphic traps, and traps on syn-depositional and post-depositional reverse faulted anticlines.

The Strathclyde Group, specifically the West Lothian Oil Shale Formation and lateral equivalents (Figure 3), in the Eastern Midland Valley region contains proven hydrocarbon reservoir and seal assemblages. Repeated, or cyclic, fluvio-deltaic clastic packages prograded into Lake Cadell resulting in the deposition of coarsening and cleaning upwards sandstone bodies that are encased within vertically and laterally impermeable lacustrine mudstones. The sandstone bodies are potential hydrogen storage reservoirs and typically range in thickness from 10-20 m, although amalgamated sandstone bodies can have a significant thickness of more than 100 m, as seen at the Craigleith quarry in Edinburgh. The Fife Ness, Gullane and Anstruther Sandstone Formations have good reservoir properties with sufficient permeability to allow for hydrogen storage, as gas production tests in the Cousland Field show. The organic-rich oil shales of the Upper Strathclyde Group are proven and effective hydrocarbon seals and could potentially retain hydrogen too. The Lake Cadell oil shale deposits in the Upper Strathclyde Group contain hydrocarbons and therefore display exceptionally high capillary entry pressure that may provide an excellent seal for hydrogen storage. The thickness of individual oil shale beds varies from a few centimetres to approximately 5 m. However, 13 to 20 organic-rich oil shale layers have been recognised within the West Lothian Oil Shale Formation, with a combined thickness of around 35 m in the Eastern part of the Midland Valley Basin [40]. Due to post-depositional compressional and transpressional tectonics related to the Late Carboniferous Variscan orogeny, the Early Carboniferous sediments in the Midland Valley have been deformed, producing numerous anticlines and faulted anticlines, which could act as trap structures for hydrogen storage. Several of these anticlines have been recognised onshore and are located within the depositional area of Lake Cadell, including; the D'Arcy-Cousland Anticline (discussed below), the Balmule Anticline, the Burntisland Anticline and the Earl's Seat Anticline [43] (Figure 4). Additionally, local dome structures, such as the Rosyth dome, could act as smaller scale hydrocarbon storage sites. The actual size of the traps is difficult to determine as, although there are multiple gas shows and small-scale production, Cousland remains the only well-documented hydrocarbon field which has been produced with good records.

There is sufficient data available from hydrocarbon and aquifer wells, mining operations and outcrops to study the oil shale hydrogen storage play. However, there are no large hydrocarbon gas fields available and the reservoir thickness is limited to only several meters at most of the studied localities. The hydrocarbon fields in the D'Arcy-Cousland Anticline provide excellent potential for hydrogen storage close to Edinburgh. The presence of recoverable gas from sandstone reservoirs at relatively shallow depth encountered by the D'Arcy well is particularly promising because it indicates that hydrogen can potentially also be stored at shallow depth in reservoirs of the Upper Strathclyde Group. The overall extent of the play is limited to the extent of Lake Cadell (which equates approximately to the Eastern part of the Midland Valley) and the presence of petroleum-rich oil shales. Another uncertainty is the presence of hydrocarbons and how they will interact with injected hydrogen in reservoirs of the Upper Strathclyde Group. Given the moderate thickness and potential lateral extent of the Pathead, Anstruther, Fife Ness and Gullane formations with moderate permeabilities, the numerous anticlinal structures and the extensive Upper Strathclyde Group seal, the Lower Carboniferous Upper Strathclyde Group represent a hydrogen storage opportunity.

Case study: The hydrocarbon fields of the D'Arcy-Cousland Anticline

Hydrocarbons were found in the early 20th century in the sandstone reservoirs of the Upper Strathclyde Group in the Midland Valley. The Cousland Gas field, one of the major gas accumulations in the Upper Strathclyde Group, contained approximately 9.3 Mm^3 of natural gas, which is relatively minor compared to offshore gas fields in the Southern North Sea, which are significantly larger. For example, the Cygnus field, which started production in 2016, has an estimated gross proven and probable reserves of 18 Gm^3. The D'Arcy-Cousland Anticline has a proven seal, which has retained oil and gas over geological timescales, as well as permeable sandstone reservoirs [41].

Several wells were drilled into the Cousland Gas Field, the main gas field of the D'Arcy-Cousland Anticline. The best gas-bearing sandstone was tested at 5.9 Mft^3 (~167,000 m^3) per day [41]. Production started in 1957 and, within the following ten years, 330 Mft^3 of gas (9.3 Mm^3) was produced. Assuming standard conditions for the calculation (0°C, 1bar) and an average depth of 500 m for the gas-bearing Cousland Reservoir, a flow rate of 5.9 Mft^3 of methane per day is volumetrically equivalent to an approximate hydrogen production of 13.2 t per day. As a capacity estimation for hydrogen storage, the volume of natural gas produced from the Cousland Field during the ten years of production (330 Mft^3/9.3 Mm^3) is equivalent to a total of approximately 700 t of hydrogen.

Storage play 3: The Carboniferous Lower Clackmannan Group

This play comprises fluvial, deltaic and shallow marine arkosic and arenitic sandstone reservoirs, with overlying marine flooding surface top seal and lateral floodplain mudrock seal; the traps are stratigraphic, or faulted inversion anticlines and domes of 4 way dip closure.

The Carboniferous Lower Limestone, Limestone Coal and Upper Limestone Formations in the Midland Valley contain permeable fluvial sandstone deposits, into which hydrogen could be injected and stored (Figure 3). The clastic sequences comprise coarsening and cleaning upwards successions of siltstone and sandstone, often capped by palaeosols and coal horizons. These successions are interpreted as prograding fluvio-deltaic deposits and provide good quality reservoir rocks. Palaeocurrent directions are mainly directed towards the southwest indicating that the sands were likely sourced from the distal Grampians and Scandinavian highlands and potentially also localised adjacent highs. The sandstone layers are typically around 6 m, and up to 30 m in thickness [40]. The prograding clastic successions alternate with low permeability marine limestone and shale accumulations, which could act as local seals for hydrogen stored in sandstone reservoirs. Marine band shales are flooding surfaces which have proven their capability to act as seals in the East Midlands hydrocarbon province of northern England. Together, these facies are arranged into progradational cyclic sequences. A complete cycle is represented by a marine limestone or mudstone horizon, deposited during a relative sea level rise (transgression) followed by siltstones and sandstones during a high (highstand) to falling sea level and palaeosols with coal development deposited during low sea level (lowstand) and early stages of transgression. The cycle is then repeated by another marine incursion and subsequent progradational clastic sequence. However, the cyclicity is often imperfect with out-of-sequence deposition common [40]. This cyclicity may be useful for energy storage purposes as it provides multiple (stacked) hydrogen reservoir storage localities with abundant thin, but potentially laterally extensive seals.

Similar to Play 2, there is sufficient well data, data from mining operations and outcrop data available to study the oil shale hydrogen storage play. For example, the small Balgonie discovery and the wells drilled into it provide important subsurface information that oil shows occur in multiple sands, which could be suitable for exploitation as multiple discrete storage horizons (Figure 7). However, there are no large gas hydrocarbon fields available and the reservoir thickness is limited to only several meters at most outcrop localities. The marine shales are laterally extensive and have been proven to retain significant hydrocarbon columns in the East Midlands (UK). Therefore, the marine bands could potentially act as similar seals, as yet unproven in quality, in the Midland Valley. The main uncertainty is the lateral extent, connectivity and quality of potential reservoir units. Although well data is present, the Carboniferous geology is very complex and heterogeneous, making interpretations based on well logs and seismic data difficult. Additionally, it is unknown whether faults in this area are sealed or are potential fluid migration pathways.

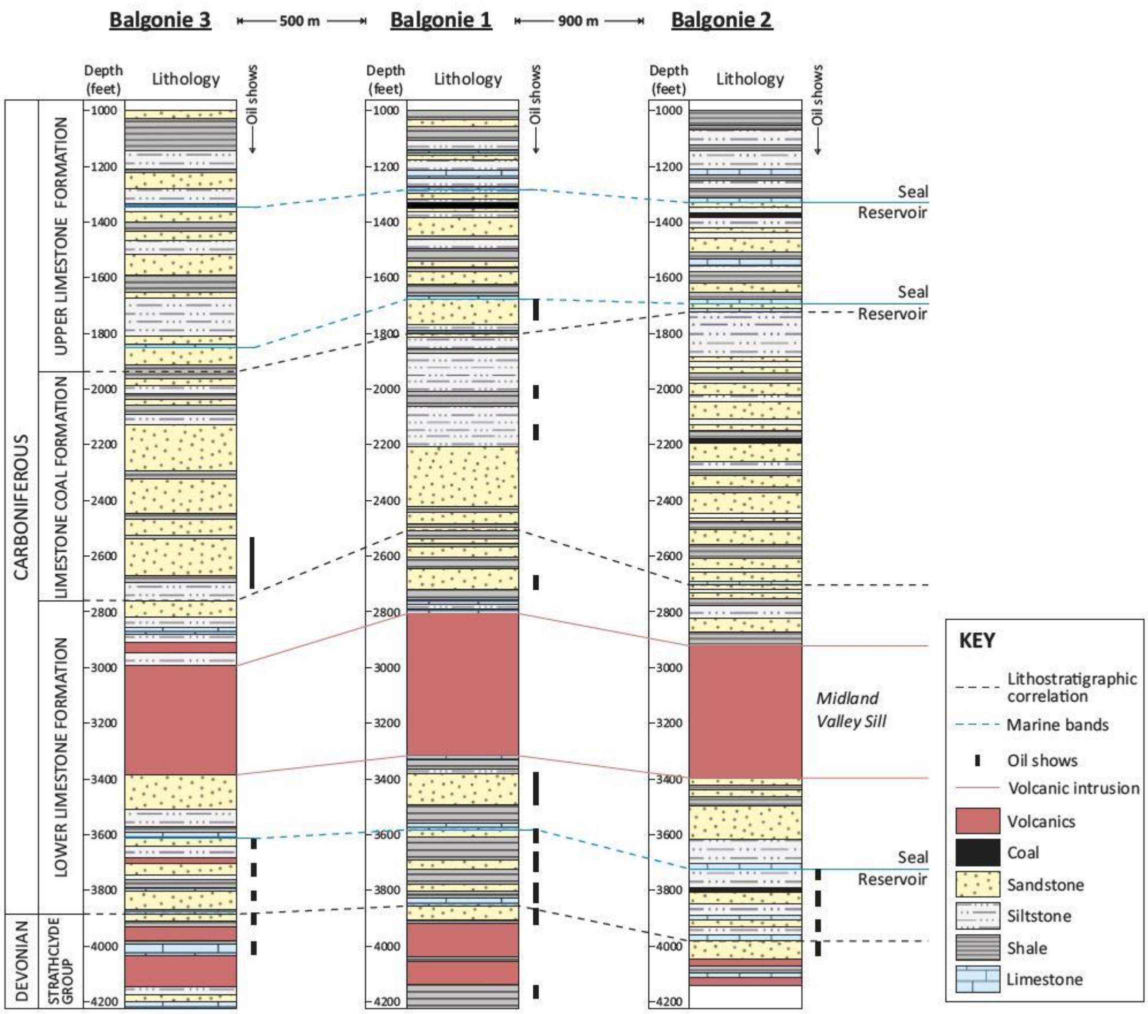

Figure 7: Stratigraphy of three wells of the Balgonie Field (see Figure 4 for location), an oil field with proven but uncommercial oil. The black vertical lines show where oil shows were present. Three marine band shales, potential seal rocks for stored hydrogen, were correlated across the field (brown lines). Formation and group names given in black. As it can be seen, oil might have been retained by marine flooding surface in Balgonie.

Discussion and Conclusions

The usage of hydrogen can reduce reliance upon fossil fuels and support compliance with the Paris climate protection agreements. This paper proposes hydrogen storage in subsurface porous media, in depleted oil and gas fields or saline aquifers, as large-scale storage sites and provide the necessary storage space for a hydrogen based energy sector. The Midland Valley is the most promising area in on-shore Scotland for hydrogen storage and other gas storage operations. It provides potential reservoir rocks, seals and traps; is relatively well understood due to its long hydrocarbon history and potential storage sites are close to consumers.

At a basic science level, there is very little known about quantifying storage capacities for hydrogen storage sites. For this study, we have allocated a value for potential sites within each hydrogen storage play. The range for capacities is exponential and lies between "very high", comparable with large scale saline aquifers, which could store than 100 Mkg of hydrogen, in which an assumed capacities of more than a mega tonnes of hydrogen, and "very low", comparable with surface steel containers. However, it should be kept in mind that these numbers are preliminary estimations and need confirmation by digital modelling applications and real-world storage projects. The storage capacity of storage sites within hydrogen play 2 (fluvial sands encased in shales, Lower Carboniferous Upper Strathclyde Group) and play 3 (fluvial and shallow marine sands sealed by flooding surface marine mudstones, Carboniferous Lower Clackmannan Group) is estimated to be of "medium" size, comparable with small oil and gas fields. Large salt caverns such as the hydrogen storage sites in the US, with a storage volume of up to 580,000 m^3 and a possible hydrogen working gas capacity of 2.56 Mkg, are of comparable size [21]. The capacity of these two Carboniferous plays is mainly controlled by the extent and heterogeneity of the reservoir rocks, reservoir compartmentalisation due to the faults and the size of potential trap structures. By contrast, the Devonian Stratheden and Inverclyde Group sandstones of play 1 offer storage capacity of a much larger scale. The thick and extensive reservoirs could offer enough capacity in the range of megatons. Much larger storage opportunities exist offshore, through re-use of depleted hydrocarbon fields. These will be investigated elsewhere, and although larger will require much greater investments to develop.

The geological uncertainly, a measure on how much is known about a potential storage site, is another important parameter. We qualitatively define the geological uncertainty to lie between "very low", for well understood and recent gas storage sites such as the Sleipner CO_2 storage site offshore Norway, and "very high", for formation with limited geological knowledge and no existing hydrocarbon play. The play 1 (Devonian Sandstones), although potentially providing the largest hydrogen capacity, also have the highest geological uncertainty. The limited geological knowledge is due to the fact that information is mainly based on outcrops, with a lack of information about lateral extent of both cap and reservoir rocks, a poorly investigated stratigraphy and uncertain seal thickness and quality. Additionally, very little is known about trap structures and no known hydrocarbon fields are present. Much more is known about the Carboniferous plays 2 and 3. Although significant data is available, the Carboniferous geology in the Midland Valley is very complex and heterogeneous making interpretation based on well logs and seismic data difficult. Further uncertainties derive from the presence of faults as either pathways or seals for injected hydrogen and little is known about the lateral extent of the reservoir sandstones. Additionally, the extent and quality of the Oil Shale seals to the west of the Midlothian basin is uncertain.

It is difficult to fully evaluate the regional sealing capacity of the oil shales and the marine bands but several small hydrocarbon findings exist in the Carboniferous where oil and gas have been produced to the surface. This indicates that retention can occur on timescales of around 20,000 years and maybe longer. However, more information is present for the marine shale bands as hydrocarbon seals because they have proven to retain significant quantities of hydrocarbons in the East Midlands (UK). Figure 8 summarises the interpretation of the capacity estimation and the geological uncertainty.

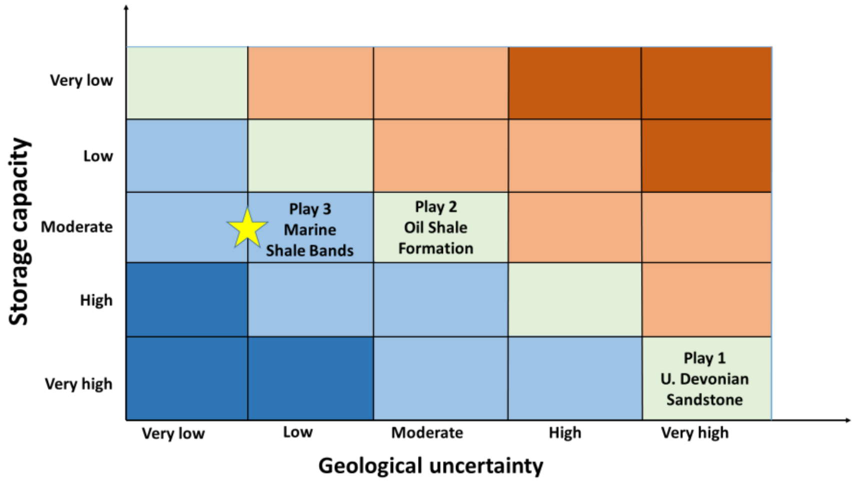

Figure 8: Hydrogen storage capacity vs geological uncertainty plot of the three storage plays. The x-axes represent the geological uncertainty for gas storage operations, scaled from "very low" (e.g. the Sleipner CO_2 storage site with years of operational experience) to "very high" (limited geological knowledge). The y-axes represents the storage capacity. Since there are no official guidelines to assess the storage capacity of porous media for hydrogen, a more intuitive scale is proposed. As a reference size, the storage capacity of large salt caverns used for hydrogen storage in the US were defined as "medium". The greatest storage capacities ("very high") are expected in large scale saline aquifers with 100 or more times the capacity of a large salt cavern to supply a major city such as Glasgow or Edinburgh for one year. The yellow star represents the Cousland Field, a part of play 2 but with lower geological uncertainty compared to less investigated sites of play 2 an 3 due to its long and successful exploration history. Further work is needed to move these summaries towards the lower left corner of the diagram.

According to our analysis, the Carboniferous plays (play 2 and 3) offer medium sized hydrogen storage capacities, which can be investigated at specific sites for regional storage projects. Also added to Figure 4 is the Cousland field which has the lowest geological uncertainty because it is known that the geology works for gas storage: The seals were capable of retaining gas over long timescales and production tests prove an appropriate permeability for production, and also, injection. Future tests are required to show if the geology of the Cousland field also works for hydrogen injection, production and storage but based on current understanding the Cousland field is the best onshore site for a small scale hydrogen storage research and pilot project. The Balgonie site (play 3) could also be investigated to understand hydrogen retention by multiple reservoir layers. If, however, large scale hydrogen storage in porous media is targeted in Scotland, the Devonian Sandstones have to be considered, or more costly opportunities offshore should be targeted.

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