Natural hydrogen exploration in Australia – state of knowledge and presentation of a case study

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\textbf{ABSTRACT}

Hydrogen will play a major role in Australia’s transition to a net zero emissions energy future. The hydrogen industry and technology are scaling up with hydrogen produced via two pathways, thermochemical and electrochemical, that involve the use of fossil fuel feedstock or the use of an electrical current to split water into hydrogen and oxygen. Exploration for and production of natural hydrogen is one of the most promising ways to get large quantities of green hydrogen cheaper than the ‘blue’ hydrogen produced from methane. Some predictions from this growing industry even estimate that the production of natural hydrogen can quickly become economically viable. We propose to review the state of knowledge of natural hydrogen exploration and production in the world and focus on the exploration of the Australian natural seeps in the frame of the incredible exploration rush we are currently experiencing. Surface emanations often referred to as ‘fairy circles’ are often associated with high hydrogen soil-gas measurement and have been described in numerous countries. In the frame of our research, we recently showed that similar hydrogen-emitting structures are present in Australia. New regional scale soil-gas measurements reveal persistent hydrogen concentration along the Darling Fault, in the Perth Basin and on the Yilgarn Craton. Those geological settings promote processes such as deep serpentinisation of ultramafic rocks as potential hydrogen sources that are of massive potential economic value. We review the results of different techniques to explore and quantify the presence of natural hydrogen leakage.

\textbf{Keywords:} fluid and gas leakage, hydrogen seeps, natural hydrogen exploration, Perth Basin, renewable, structural geology, Yilgarn Craton.

\textbf{Introduction}

Hydrogen will play a major role in Australia’s transition to a net zero emissions energy future. The hydrogen industry and technology are scaling up with hydrogen produced via two pathways, thermochemical and electrochemical, that involve the use of fossil fuel feedstock or the use of an electrical current to split water into hydrogen and oxygen. Exploration for and production of native hydrogen is one of the most promising ways to get large quantities of green hydrogen cheaper than the ‘blue’ one produced from methane (Deville and Prinzhofer 2016; Gaucher 2020). Some analysis estimates that the production of natural hydrogen close by (< 300 km) a hydrogen exportation hub or a major city can quickly be economically viable, with a targeted production cost below $2 \text{ kg}^{-1}$ (Levin and Hart 2021).

The discovery of a large hydrogen accumulation in Mali and the steady production rate since the start of production 7 years ago (Prinzhofer et al. 2018; Moretti 2021) tends to demonstrate the potential of this carbon-free and potentially renewable energy. However, the elements of the natural hydrogen system are not yet well understood, and researchers are currently investigating the potential sources and migration of hydrogen in continental settings that could be explored. Examples include a potential active serpentinisation kitchen in the Pyrenees orogenic belt separating Spain from France (Lefèvre et al. 2021), or the migration pathways and their surface expression with a
focus on natural seeps forming circular depressions referred to as ‘fairy circles’ (Larin et al. 2015; Zgonnik et al. 2015; Deville and Prinzhofer 2016; Prinzhofer et al. 2019; Myagkiy et al. 2020). Large uncertainties remain on the very nature of natural hydrogen plays and a larger variety of natural cases need to be studied to assess if we are dealing with conventional types of accumulation with a charged reservoir overlaid by a top seal or if there could be continuous flux of hydrogen charging poorly defined and sealed reservoirs where the recharge rate overcomes the diffusion and/or leakage rate.

In this paper we review natural hydrogen occurrences in Australia and document examples of potential hydrogen seeps in three different geological settings: a structurally complex passive margin, a cratonic environment and an intracratonic saliferous basin. These data aim at increasing the knowledge of natural hydrogen seep systems in Australia and bring calibration data for exploration activities.

**Natural hydrogen exploration in Australia**

In Australia, natural hydrogen occurrences started to be widely investigated in the last couple of years with the publication of desktop studies and petroleum-well compilations revealing a high potential for the cratonic rocks to generate high quantities of hydrogen (Rezaee 2020; Boreham et al. 2021a; Moretti 2021a) (Fig. 1a). Moretti et al. (2021a) highlighted similarities between the Australian intra-cratonic geological settings with those of the Bourakebougou hydrogen accumulation in Mali, and the similar geometry of high-flux hydrogen seeps reported worldwide (Myagkiy et al. 2020), with a particularly high cross-correlation with monitored seeps in the iron-rich San Francisco Basin in Brazil (Prinzhofer et al. 2018; Moretti et al. 2021b). The Australian cratons are composed of unique iron-rich, radiogenic plutonic and sedimentary rocks, such as banded iron formations and ultramafic and mafic rock, which have been identified as prolific sources of natural hydrogen by deep hydration (e.g. radiolysis or serpentinisation) and oxidation processes (Gaucher 2020; Klein et al. 2020; Geymond et al. 2021; Lefevre et al. 2021). Isotopic analyses of well samples so far tend towards a high contribution from abiotic processes, particularly radiolysis and serpentinisation, for hydrogen generation in the currently published case studies, such as the sub-salt wells drilled by Santos in the Amadeus Basin (McInnes et al. 2017; Mendes 2021), the analysis of mineral wells near Kalgoolie on the Yilgarn Craton and petroleum wells (Boreham et al. 2021a; 2021b). The Mt Kitty 1 well test, in the Amadeus Basin, returned high levels of hydrogen (>10%) and helium, (9%); helium gas isotopic analysis (i.e. He/4He and He/20Ne ratios) was done to constrain the source of the free gas released by a fractured basement rock located below a thin salt interval. Hydrogen gas stable isotope analyses of historic well samples with more than 10% mol of hydrogen located above Precambrian basement rocks (South Australia, Tasmania, Western Australia and Northern Territory) mostly show abiotic hydrogen sources (Boreham et al. 2021a). Based on serpentinisation and radiolysis processes, Boreham et al. (2021a) estimated an onshore Australian potential to generate hydrogen between 1.6 and 58 MMm³ per year down to a depth of 1 km.

The presence of hydrogen seeps similar to the ‘fairy circles’ identified worldwide have been confirmed by measurements in the Moora area, in the vicinity of a major crustal boundary separating the North Perth Basin from the plutonic, mafic and ultramafic rocks of the Yilgarn Craton (Frery et al. 2021). In late 2021, Buru Energy installed a specialised hydrogen mudgas detection unit on their rig and announced the discovery of hydrogen in the Rafael-1 and Currajong-1 petroleum wells drilled in the Canning Basin. Those wells were drilled in different geological formations and both indicated elevated hydrogen concentration in reservoir intervals, from 4.9% in Rafael-1 to 6% in Currajong-1 (Fig. 1a).

In February 2021, the South Australia Government amended the Petroleum and Geothermal Energy Regulations 2013 to add hydrogen, hydrogen compounds and by-products from hydrogen production regulated substances in the Petroleum and Geothermal Energy Act 2000. This opened the doors for companies to explore for natural hydrogen and allowed the transmission of hydrogen or compounds of hydrogen via pipeline. The exploration block release (Fig. 1b) created a vacuum for natural hydrogen exploration and all the exploration licences were applied for by September 2021. The regulation is still evolving with the current review of the Petroleum and Geothermal Energy Act 2000 proposed to become the ‘one stop shop’ for hydrogen under a new name – the Energy Resources Act.

**Geological settings and method**

This study details three field examples from (A) the Beermullah Trough in the North Perth Basin, (B) the Officer Basin overlying the Yilgarn Craton in the Far East Yilgarn province and (C) the central Yilgarn Craton (Fig. 2).

The North Perth Basin is separated from the Yilgarn Craton by the Darling Fault, a major crustal boundary still seismically active (Fig. 2). This passive-margin basin initiated during the Permian above Archean to Proterozoic formations of the Pinjarra Orogen (Bodorkos et al. 2016). The main hydrocarbon source rocks are the Permo-Triassic Irwin River Coal Measures and Kockatea Shale (Fig. 3a). Faults have an important role for hydrocarbon migration from those sources to Jurassic structural plays (Langhi et al. 2012; Grosjean et al. 2013; Frery et al. 2017). The study area (Fig. 2a) is located east of the Gingin Scarp in the Beermullah Trough that is interpreted as a depocentre
cross-cut by a complex system of north-west to north-east and north-trending faults (Crostella and Backhouse 2000).

The second study area (Fig. 2b) is in the Officer Basin that is overlying the ultramafic, mafic and plutonic rocks of the Far West Yilgarn province. This Neoproterozoic basin is partly covered by the Canning Basin (Fig. 2). It is composed of four super-sequences including a basal evaporite layer (~800 Ma) (Fig. 3b). This thick salt layer is located above the Townsend Quartzite reservoir and is punctually doming and intruding the sedimentary series up to the surface (Haines 2020).

The third study area (Fig. 2c) is located on the Yilgarn Plateau, from the Eastern Goldfield Superterrane to the Wheatbelt region (Fig. 2). Archean komatolites, basalts and volcanoclastic rocks are overlaid by a thin layer of regolith and narrow north trending basins hosting Neo-Archean to Paleoproterozoic sediments (Fig. 3c).

Soil-gas in the Beermullah and Yilgarn Craton sites was analysed with a GA5000 gas analyser coupled with an 80-cm inox tube as presented by Prinzhofer et al. (2018). The inox tube is introduced in the soil from a few 10s of centimetres to a metre depth to measure methane, carbon dioxide, oxygen and hydrogen concentrations. Additional methane and ethane concentrations are recorded with another gas analyser at ppm level. This technique is useful for a primary exploration, to determine the presence and concentration of hydrogen in the near surface. However, it does not provide any insight on the hydrogen flux, evolution with time and potential underlying reservoir.

Fig. 1. Hydrogen discovery in Australia. (a) Map showing a compilation of hydrogen occurrences in wells (Boreham et al. 2021a) with potential seeps of high concentrations, the first seeps measured in the North Perth Basin and new drilling hydrogen discoveries in the Canning Basin (BuruEnergy 2021; Frery et al. 2021; Moretti 2021a; Oil and Gas News 2021a, 2021b). (b) Map showing the hydrogen exploration licences released in 2021 in South Australia, in purple (Government of South Australia website 2021).
Results

North Perth Basin-Beermullah Trough

The swamp sampled in the Beermullah Trough is located west of the Gingin Scarp and is part of a series of swamps and lakes aligned along north-west trends subparallel to the scarp and laying on the Pleistocene Bassendean sands. Those swamps are separated by the Gingin Scarp from the Gingin petroleum field and outcropping Tertiary laterites and Cretaceous green sands (Fig. 4a). The Gingin Scarp correlates with a deep north-west trending fault well imaged on the magnetic grid (Fig. 4b) along which a low magnitude (<2 orders of magnitude) has been recorded in the last 10 years (Geoscience Australia 2021, https://earthquakes.ga.gov.au/). The gravity reprocessed grids (Fig. 4c–e) show low gravity values increasing towards the north of the Beermullah Trough. The swamp is located on the western flank of an anticline characterised by a high magnetic signal (Fig. 4d).

The hydrogen soil-gas values are highly variable and reach values above 200 ppm. The values rapidly decrease outside of the swamps to a baseline value of 0 ppm (Fig. 4f).

By comparing historical satellite images over the zone of interest, four fairy-circle like features were identified between April 2006 and January 2010 (Fig. 5). Those structures, B1–B4, are located on Fig. 4b–e and are well aligned along...
the deep north-west fault and could potentially be linked to
gas escape along the fault plane triggered by a seismic event.
They are small with a diameter <100 m (Fig. 5) and no
human infrastructure, such as a track, can be observed linked
with those occurrences. The occurrence B2 even crosscuts an
existing track.

**Far East Yilgarn Province**

High densities of fairy-circle like features (a–c, Fig. 6) are
observed on satellite images along basal fault cross-cutting
the sedimentary basins, as indicated by the joint presence
of SEEBASE faults, in red, and more recent faults interpreted
on the 1:500 000 geological map, in purple. Note that those
potential gas seeps are located close by localised outcrops
ping salt diapirs and in highly magnetic zones, as indicated
by the high magnetic intensity of the magnetic worms. The
size of those potential seeps ranges from 400 m of diameter
to few tens of metres with a majority or those circular to
ellipsoidal features presenting an external ring of green
vegetation.

**Yilgarn Craton**

The Yilgarn Craton hydrogen soil-gas survey returned a
large range of values, from zero to more than 400 ppm
(Fig. 7) (detailed measurements in the Supplementary dataset). The highest concentrations were systematically measured along deep fault zones and, at a large scale, correlate with known gold deposits. Baseline measurements away from the identified fairy circles return hydrogen concentrations up to 20 ppm on the Kalgoorlie Terrane. These baseline values decrease towards the west. West of Kalgoorlie, on the Yilgarn Plateau, towards the Wheatbelt region, a transect of baseline measurement shows hydrogen values generally between 0 and 50 ppm; anomalous values superior to 100 ppm were recorded and clearly correlate with deep faults (close by fairy circle) and known gold deposits (Fig. 7).
Local-scale results show some degree of complexity in the correlation of the high hydrogen measurements with the deep fault network as well as with a wide range of fairy circle characteristics (Fig. 8).

For instance, hydrogen measurements south of Lake Lefroy (Fig. 8a, b) show that the hydrogen concentrations measured along an east–west profile vary greatly while crossing three different main fault segments, with significant values (i.e. >100 ppm) only recorded along the westernmost fault segment (Fig. 8a). Detailed interpretation and field assessment show that in this area, the hydrogen concentration is below 200 ppm, with a large local variations that seem more correlated to the soil compaction and type than the location of the measurement on the fairy circle (Fig. 8b). Note that no hydrogen concentration in soil-gas has been returned by our measurements on the salt layer of Lake Lefroy (Fig. 8a).

The south Lake Lefroy, Yellowdine and north Kalgoorlie hydrogen seeps (Fig. 8b, c) return hydrogen values in the same range, with measurements in the internal and external rings of the structures mainly ranging between 50 and 100–200 ppm. However, those seeps have different surface characteristics. The Yellowdine seeps and the larger north Kalgoorlie seeps present an external ring of green vegetation, geometries varying from circular to a mouse face shape (Fig. 8c, d). The south Lake Lefroy seeps are elongated, ranging from more than 500 m long to only few tens of metres, and only parts of those seeps are surrounded by external rings of green vegetation (Fig. 8a, b). In the north Kalgoorlie area (Fig. 8d), numerous small circular seeps (diameter < 10 m) are observed towards the north of the large mouse face seep. Those seeps are only identifiable from the satellite images by a darker colouration of the soil which correlates, in the field, with a local higher water content and a small depression. Comparisons of satellite images over a 10-year period and different seasons clearly show that these features are permanent (Fig. 9) and field measurements returned hydrogen concentrations between 50 and 100 ppm (Fig. 8d).

Discussion

This study shows soil-gas hydrogen measurements in different Western Australian geological settings, ranging from 0 to 207 ppm along the Darling Fault system near Beermullah in the North Perth Basin (Fig. 4 and detailed measurements in the Supplementary dataset), from 0 to 447 ppm on the Yilgarn Craton (Figs. 7, 8 and detailed measurements in the Supplementary dataset), as well as potential hydrogen seeps in the Far East Yilgarn province, nearby Neoproterozoic salt intrusions from the Officer Basin supersequences (Fig. 6). In a previous study along the Darling Fault, that represents a major crustal boundary separating the North Perth Basin from the Yilgarn Craton, Frey et al. (2021) already demonstrated the presence of hydrogen in soil-gas measurement with values up to 96 ppm. The new results presented here confirm the presence of a high density of natural hydrogen seeps in Western Australia inferred by recent desktop studies (Rezaee 2020; Moretti 2021a). However, a recent review of historical well-data in Western Australia (Haines 2021) showed that natural hydrogen has already been measured in more than 10 wells in the Perth Basin and the Canning Basin (Fig. 2). These results expand the known areas with a potential active hydrogen system in Western Australia by documenting active seeps on the Gold Fields province of the Yilgarn Craton where high hydrogen levels have recently been measured in open mine wells (Boreham et al. 2021a) and within the North Perth Basin, where hydrogen values up to 50 000 ppm have been measured at 6 m depth close by the Gingin gas and condensate field (Gole and Butt 1985).

This study measured seeps over a large range of surface features (Figs 4–9), from circular lakes to elongated swamps. The soil-gas sampling did not show any direct link with high salinity (i.e. Lefroy Lake measurements, Fig. 8) or different degrees of green vegetation in the external ring of the seeps. Some small seeps without any green vegetation rings have been identified in the North Kalgoorlie area (Fig. 9) and have been demonstrated to be permanent for at least the last 11 years, showing that the seeps do not always evolve from a small to a larger depression with an external ring of green
vegetation. This type of observation, at the continental scale, is critical to complete the observations already available in other countries such as Brazil, Russia and the United States (Larin et al. 2015; Myagkiy et al. 2020; Zgonnik 2020) and is necessary to calibrate exploration activities, from remote sensing analysis to desktop studies.

The measured hydrogen seeps are classically located along deeply rooted fault zones that connect the surface to the basement in different geological settings. Those fault zones are not only located along major crustal boundaries (e.g. Darling Fault in the North Perth Basin) but can also be located within sedimentary basins. The Beermullah seep is located at the limit between two structural elements of the complex Perth Basin (Figs. 2, 4) and the surface seeps location fits with the flank of a basement anticline well imaged on the gravity data (Fig. 4). Currently, additional isotopic measurements are required to document the source of the hydrogen; however, one can infer three main hypotheses:

- The hydrogen can be locally generated by deep hydration processes of a basal source indicated by observed magnetic and gravity anomalies,
- The hydrogen can be regionally generated at the base of the sediments and transported in a dissolved phase in one or several deep aquifers, thanks to high heat flow anomalies and low-salinity of the groundwater as discussed in Frery et al. (2021),
- The hydrogen can be linked to surface and biological processes; this case would imply that the seeps are not connected with the high hydrogen levels measured 6 m deep along the same structure near the Gingin gas and condensate field (Gole and Butt 1985).

Fig. 8. Yilgarn Craton sampling results: (a, b) Lake Lefroy; (c) Yellowdine; and (d) North Kalgoorlie. Locations on Fig. 7.
The potential seeps in the Far East Yilgarn province are located above a thick sedimentary cover and along a major subsurface fault that correlates with a basement fault interpreted on the SEEBASE top basement model (Fig. 6). The density of these seeps can be high and correlates with the density of magnetic anomalies as indicated by the distribution of the magnetic worms (Fig. 6). Interestingly, these seeps are located in zones of deep salt intrusions from the Neoproterozoic Officer Basin up to the surface. It is currently not clear if this salt shares the same age and origin as the Gilles salt under which high hydrogen (>10%) and helium (>4%) anomalies have been recorded in the South Amadeus Basin (Mendes 2021). The seeps appear to be located above ultramafic and mafic rocks of the Yilgarn Craton, interpreted under the regolith. However, those occurrences need to be precisely mapped in that area to ascertain a direct link to potential surface seeps.

On the Yilgarn Craton, random baseline measurement along the Great Eastern Highway (Fig. 7), every 30–60 km, show higher values of hydrogen in soil-gas along major north–south fault trends. These fault trends are also correlated with numerous gold mines or documented gold deposits. Boreham et al. (2021b) showed an abiotic origin for hydrogen seepage in open gold mine wells located in northwest Kalgoorlie.

This study showed that the hydrogen seeps are also associated with ethane and methane gas emissions above the baseline limit, suggesting that the hydrogen can be transformed in abiotic methane near the surface, as already proposed (Moretti 2021). However, as in the Perth Basin example, the origin of the hydrogen recorded on the Yilgarn Craton is still uncertain (Fig. 10).

The soil-gas signature against hydrogen content was measured with a GA5000 analyser coupled with an 80-cm inox tube (Prinzhofer et al. 2018; Dugamin et al. 2019; Moretti et al. 2021b). This technique was useful for a primary exploration of the presence or absence of hydrogen

Fig. 9. Yilgarn Craton sampling results, North Kalgoorlie site for the period 2009–2021. Location on Fig. 7.

Fig. 10. Conceptual models for the North Perth Basin, Yilgarn Craton and Far East Yilgarn province.
concentration in near-surface seep settings and required a hand-drilling phase. It does not provide any insight on the hydrogen flux, evolution with time and potential underlying reservoir (Frery et al. 2021).

Some authors recommend the use of a portable drill operating in percussion mode only (no rotation of the drill bit) (Halas et al. 2021; Lefevre et al. 2021), to prevent any production of hydrogen through heating or cracking of the organic matter (Lewan 1997; Lorant et al. 2002; Li et al. 2017) and through ‘mechano-radical’ processes linked to the dissociation of silicates in water-saturated rocks (Kameda et al. 2003; Hirose et al. 2011). This process is supposed to be at the origin of high concentrations of hydrogen in soil-gas associated with active tectonic faults (Kita et al. 1982; Sato et al. 1984). Halas et al. (2021) showed that all drilling with a rotary drill bit produced hydrogen concentration of at least 1000 ppm in a case study in Gironde (France). This observation highlights the importance of baseline measurements and assessing the type of soil drilled. For instance, in the case of the soil-gas sampling along the Darling Fault, Frery et al. (2021) used a rotary drill and observed a fluctuation of hydrogen values of almost 100 ppm in the outside rim of the fairy circles; these measurements were repeated, at the same locations, over three field campaigns with similar results. Additionally, drilling in similar soils outside of the fairy circles returned null hydrogen concentrations, suggesting that the drilling process is unlikely to produce artificial hydrogen. This experiment shows the importance of recording baseline measurements and testing the effect of drilling-induced heating and water saturation for each new soil type or to proceed to systematic soil X-ray diffraction analysis to assess the level of silicate content. It also shows the limitation of the rotary drilling technique for soft soil poor in silicate. Some authors (Prinzhofer and Deville 2015; Prinzhofer et al. 2019; Frery et al. 2021) defend that soil-gas surveys aiming at measuring the concentration of near-surface hydrogen are likely to target remaining hydrogen pockets that are opened by the drilling process; the free hydrogen gas is immediately released and mixed with the atmospheric gas. Only a monitoring phase including a real-time recording of the hydrogen flux and gas isotopic analysis could better define those systems, such as shown by the use of the ENGIE PARYS system in Brazil (Moretti et al. 2021b).

Conclusion

The present results confirm the high density of hydrogen seeps with recorded hydrogen values up to 447 ppm in Western Australia and demonstrate that this type of feature can be find along major fault segments in various geological settings, from a passive margin basin to a cratonic plateau and to an intra-cratonic saliferous basin.

Different surface expressions of the hydrogen seeps have been documented from few tens of metres circular depressions to hundred metres elongated features and highly variable degree of green vegetation. Interestingly, some small circular depressions without green vegetation do not tend to evolve spatially with time and could indicate a lack of recent leakage, at least over an 11-year period.

The variety of hydrogen systems is high, and the main challenge is to constrain the type of system the industry needs to focus on, e.g. a system similar to a conventional petroleum system that includes a charged reservoir overlaid by an efficient seal over geological time or a dynamic system more like a geothermal flux indicative of an active source continuously generating hydrogen.

Future research must focus on the monitoring of identified seeps to calibrate the hydrogen flux and the origin of the hydrogen. The start of natural hydrogen exploration over more than 30 licences in South Australia will certainly greatly improve our understanding of this system in the coming years.

Supplementary material

Supplementary material is available online.

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Boreham CJ, Edwards DS, Czado K, Rollet N, Wang L, van der Wielen S, Champion D, Blewett R, Feitz A, Henson PA (2021a) Hydrogen in near-surface seep settings and required a concentration of at least 1000 ppm in a case study in Gironde (France). This observation highlights the importance of recording baseline measurements and testing the effect of drilling-induced heating and water saturation for each new soil type or to proceed to systematic soil X-ray diffraction analysis to assess the level of silicate content. It also shows the limitation of the rotary drilling technique for soft soil poor in silicate. Some authors (Prinzhofer and Deville 2015; Prinzhofer et al. 2019; Frery et al. 2021) defend that soil-gas surveys aiming at measuring the concentration of near-surface hydrogen are likely to target remaining hydrogen pockets that are opened by the drilling process; the free hydrogen gas is immediately released and mixed with the atmospheric gas. Only a monitoring phase including a real-time recording of the hydrogen flux and gas isotopic analysis could better define those systems, such as shown by the use of the ENGIE PARYS system in Brazil (Moretti et al. 2021b).


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**Data availability.** Hydrogen measurements are compiled in an Excel spreadsheet that is available as supplementary material to this paper.

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