Natural hydrogen: a new source of carbon-free and renewable energy that can compete with hydrocarbons

Christophe Rigollet[1*] and Alain Prinzhofer[2,3] provide an inventory of knowledge of the hydrogen produced naturally by the Earth and present exploratory guidelines.

Abstract

Emanations of natural hydrogen are observed on the surface of the Earth at multiple points, on the five continents and on the mid-ocean ridges. When the geological conditions are favourable, this gas can accumulate at shallow depths and thus be of economic interest in contributing to the decarbonation of the energy mix.

The first deposit of natural hydrogen was accidentally discovered in 1987 in Mali and is currently in the industrial development phase. In the last two years, a dazzling multiplication of exploration projects dedicated to natural hydrogen have been launched and the first successes have been announced. At the same time, few countries have adapted their mining codes to facilitate permit submission.

The year has also been marked by the second H-Nat international congress, which brought together scientists and industrialists on the issue of exploring and producing natural hydrogen in the short term. At the same time the European EARTH2 club was founded on the initiative of 45-8 Energy, CVA and the Avenia cluster, to overcome competitive tensions and jointly promote underground solutions to the ‘hydrogen revolution’.

This article provides an inventory of knowledge of the hydrogen produced naturally by the Earth and presents exploratory guidelines.

The hydrogen currently on the market is of manufactured origin, but natural hydrogen can also be exploited from the subsoil

70 Mt of hydrogen are consumed each year worldwide, mainly for industrial purposes. This hydrogen, called ‘grey hydrogen’, is manufactured by steam reforming of hydrocarbons (78%) and coal (18%). ‘Green hydrogen’, produced by electrolysis of water, represents only 4% of this mix. However, hydrogen also exists in the subsoil, in its natural state (Prinzhofer and Deville, 2015), it is called ‘white hydrogen’ or ‘native hydrogen’.

- Steam reforming is a developed technology but emits a lot of CO_2 (more than 10 kg of CO_2 per kg of H_2). Including CO_2 capture and storage, the production cost is around $ 2-4/kg.
- Water electrolysis uses available but energy-intensive production processes. The cost of production from renewable electricity remains high, between 5 and 8 $/kg.
- Natural hydrogen is a resource in constant renewal. Its exploitation requires little energy, no fresh water and does not emit CO_2. Production costs are estimated at less than 1 $/kg and decrease in a coproduction business model (geothermal energy, helium, high-value brines).

Natural hydrogen is therefore cheaper than manufactured hydrogen and does not emit CO_2. It would therefore be an ideal complement to hydrogen produced by electrolysis in a carbon-free energy mix. Its lower cost than other renewable and carbon-free energy sources places it, in terms of competitiveness, in a favourable position to challenge fossil hydrocarbons. The small investments needed today to develop it, its possibly local and decentralized use, make it a paradigm changer for our energy future.

Even if the presence of natural hydrogen was highlighted in water or hydrocarbon drilling more than a century ago in France and Australia (Ward et al., 1933), the first drilling devoted to this exploration is much more recent. The first large-scale exploratory projects were carried out by the Hydroma company in Mali from 2008 in the Bourakebougou region, 20 years after the accidental discovery of the deposit (Prinzhofer et al., 2018). Today, small companies are focused on hydrogen exploration such as in the USA where NH2E carried out deep drilling in Nebraska in 2019 or Desert Mountain Energy which announced in February 2022 the discovery of a natural hydrogen field in Arizona. In Australia, Santos, after several exploration wells, announced in 2021 the completion of a first natural hydrogen producing well in the Amadeus basin. In Europe companies are also developing this type of activity, such as Hynat in Switzerland, 45-8 Energy and Engie in France or Helios in Spain.

To reduce costs, natural hydrogen can also be considered as a co-product of geothermal energy. But hydrogen can also be associated with other gases of economic interest such as methane, CO_2 and more particularly helium. A coupled H_2-He production for example, would make it possible to optimize this type of operation. This covalorization approach, which has been developed by

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45-8 Energy since its creation, has now been widely disseminated in the scientific and industrial community.

In most countries, the mining code is not yet adapted to the regulation of hydrogen exploration and production, but updates are in progress

The mining codes were drafted and adopted when natural hydrogen was still unknown as a natural resource. It is therefore necessary to adapt it so that natural hydrogen can be classified in one of the categories explicitly mentioned by the mining code.

Several countries modified (or are modifying) their mining code to provide industrial initiatives with the necessary regulatory framework, such as Australia, Mali, Morocco, Congo, Ukraine, France and Germany.

Natural hydrogen finds its source in the subsoil, at depth, before migrating to the surface and finally dispersing in the atmosphere. However, this source-migration-accumulation-leakage system has elements that distinguish it from the oil system.

Natural hydrogen can be produced in the Earth's crust from different processes. Some even propose a deeper origin in the mantle or the core of the earth which would have preserved primordial hydrogen (Larin et al., 1993).

The natural hydrogen produced in the Earth's crust can be generated by the radiolysis of water due to natural radioactivity, or by the oxidation of 'ferrous' iron to 'ferric' iron reducing water into hydrogen. In the natural context, this last reaction, such as the serpentinization of mafic and ultramafic rocks, is particularly effective around 300°C in the presence of water, but it can also take place more slowly at lower temperatures, then at a shallower depth, as has been shown in the laboratory.

Natural hydrogen can also result from other processes such as pyritization (Arrouvel and Prinzhofer, 2021) and ammonium decomposition (Jacquemet, 2022), mechanical friction of silicates at faults, dark fermentation of matter organic matter, the bio-photolysis of water or the cracking of organic matter.

If we define the hydrogen system as the dynamic association source-migration-accumulation-loss, the comparison between the petroleum system is tempting. However, the differences are numerous. First of all, the depths that are at stake. The genesis of hydrogen may be deeper than that of hydrocarbons and the accumulations of hydrogen may, on the contrary, be shallower, as is the case in Mali. The main source of hydrocarbons is organic matter, while hydrogen is formed by mineral chemistry reactions, in rocks which may be sedimentary or plutonic. While for hydrocarbons it is necessary to have traps to capture the fluids, the accumulations of hydrogens can be perceived as more dynamic. Any change in rock properties that would help in slowing the gas on its migration path can promote transient accumulation on human timescales. Consequently, while the resource of a hydrocarbon deposit is measured in volume, the resource of a hydrogen deposit must integrate the notion of feeding flow.

From a temporal point of view, the petroleum system is a system that operates on the scale of geological time. Hydrocarbons are therefore considered non-renewable on a human scale. In comparison, the natural hydrogen accumulations are continuously fed by large flows and the hydrogen that reaches the surface oxidizes in the form of water, which makes this new renewable carbon-free energy resource part of the water cycle. Hydrogen fluxes are much larger, both in terms of their genesis and in terms of surface exudations.

The inventory of natural hydrogen emissions at the surface shows that the resource is widely distributed on all continents, in various geological contexts.

Natural hydrogen is present in the atmosphere but in very low concentrations, around 0.5 ppm. However, it is found in higher concentrations at point sources such as submarine or continental fumaroles, hot springs, 'fairy circles' or along fractures and faults. Many boreholes have also found hydrogen at varying depths, from a few metres to more than 1000 m (Guélard, 2016, Prinzhofer et al., 2019, Boreham et al., 2021 and Pélissier et al., 2021).

Surface emissions have been mapped globally and show a wide distribution (Prinzhofer and Deville, 2015, Zgonnik, 2020, see Figure 1). They appear along oceanic ridges, on obducted oceanic plates (ophiolites from Oman, New Caledonia, the Philippines, Turkey, etc.) or in mountain ranges (Pyrenees). They are also observed on the edges of graben (Rhine Graben and Rhine Ditch) and in Proterozoic cratons (Russia, USA, Brazil, Australia, Africa, etc.).

Bourakébougou field in Mali

In 1987, a water borehole in the village of Bourakébougou was abandoned at a depth of just over 110 m, after a gas eruption

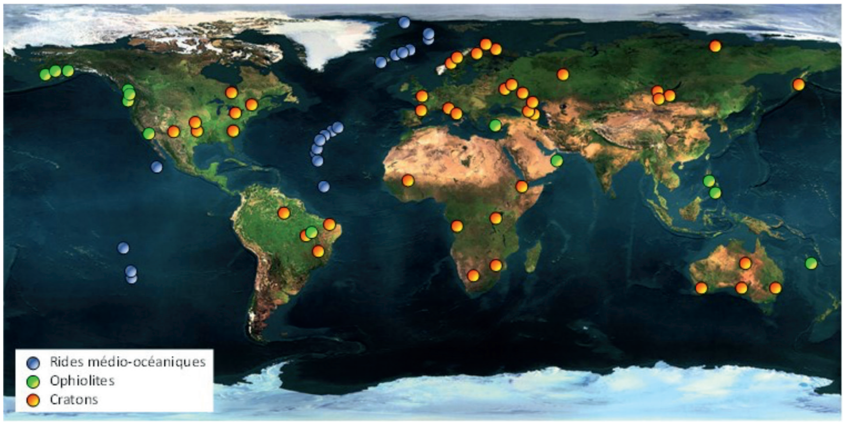

Figure 1 Map of natural hydrogen emissions – updated from Prinzhofer and Deville, 2015.

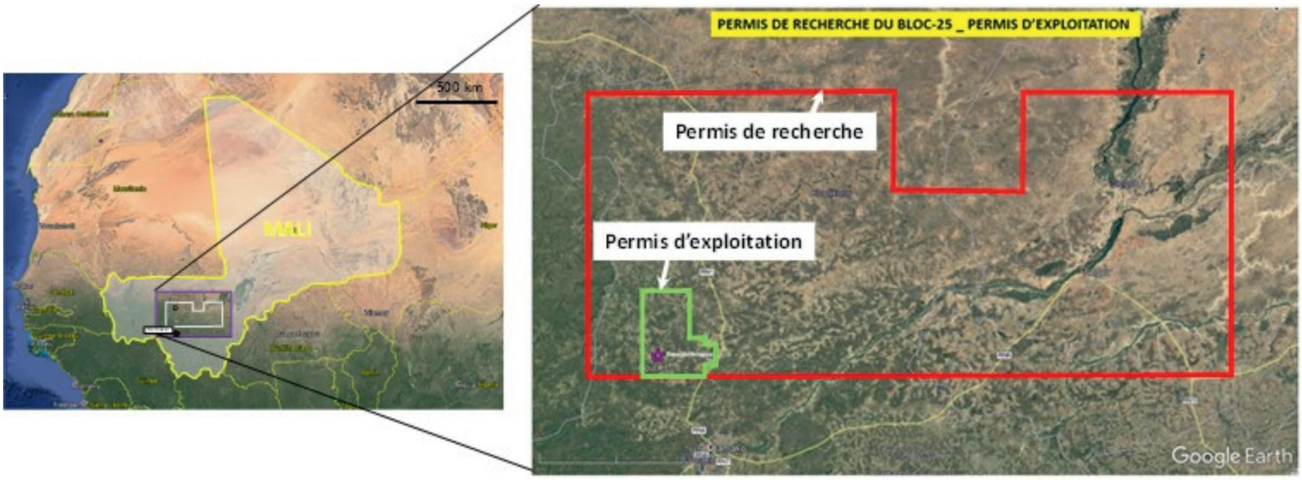

Figure 2 location of the Bourakébougou fields and the exploration.production block of the Hydroma company in Mali.

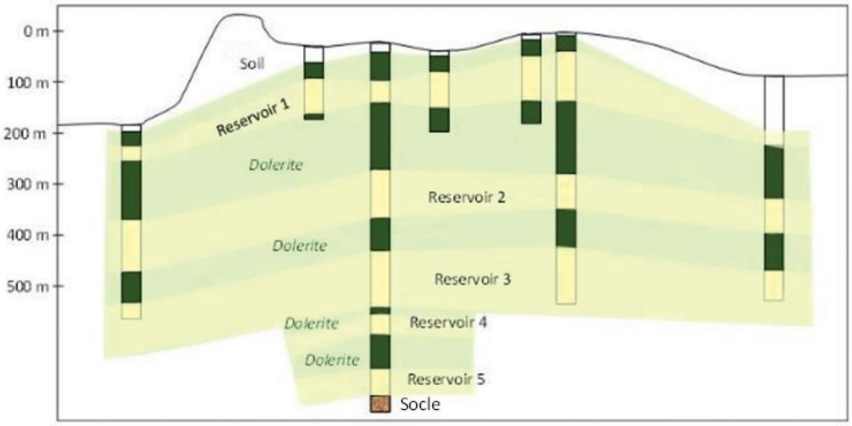

Figure 3 Synthetic section of the Bourakébougou field, showing the five superimposed reservoirs, made up of Neoproterozoic sedimentary rocks, intercalated by doleritic sills of Triassic age. The basement was reached by one of the wells.

which ignited, burning a borehole operator. The gas pocket is at a depth of 110 m, so much more superficial than all the hydrocarbon deposits exploited today. In 2008, the company Petroma (now Hydroma) reopened the well, and analysed the gas which contained 98% hydrogen, the rest being mainly nitrogen and methane. After having acquired an exploration block the size of Switzerland and an exploitation block centered on the discovery of Bourakébougou (Figure 2), geophysical and geochemical campaigns were undertaken, and the pioneer well is used for pilot production and supplies electricity to the village of Bourakébougou. Even if this production remains modest, it is the first time that electricity has been produced from natural hydrogen. Since then, Hydroma has drilled 24 other wells, some to 1800 m deep, crossing part of the plutonic basement under the Neoproterozoic sedimentary formations. It turns out that five reservoirs filled with hydrogen were discovered, from the most superficial at about 100 m deep, to the deepest at more than 1000 m (Figure 3). Natural hydrogen is also found in the underlying bedrock, showing its deep origin, although its lightness and small size allow it to migrate to more superficial formations and eventually into the atmosphere. The deposit is not associated with surface emanations; we do not observe 'fairy circles' in the immediate vicinity. This may be due to the fact that the reservoirs are locally well sealed, or that the hydrogen emitted is consumed by the lateritic soil.

The beginning of the exploitation of the Bourakébougou field makes it possible to obtain scientific information that greatly increases our knowledge of hydrogen systems. For example, the gas pressure at the wellhead either remained constant during production or increased slightly, indicating natural replenishment at the same time as it was industrially drained. This renewable and renewed character of this resource is confirmed, while many indirect arguments went in the same direction: the hydrogen fumaroles on the oceanic ridges and in the ophiolites cannot be fossil and are therefore produced at the same time as they disperse in the atmosphere; the monitoring of hydrogen emanations on a fairy circle in Brazil (Prinzhofer et al., 2019; Moretti et al., 2021b) shows the arrival of deep hydrogen pulses throughout the monitoring, with their modulations by phenomena superficial (variation in atmospheric pressure, desorption of hydrogen from soil clays, bacterial activity, etc.). Finally, the gas concentration profiles observed during drilling in the Bourakébougou field show heterogeneity between the hydrogen peaks and the methane peaks (Figure 4), showing that within a reservoir itself the gaseous species do not rebalance each other because their migration and accumulation is faster than the time required for diffusive rebalancing in a reservoir rock.

In conclusion, the Bourakébougou field represents the first natural hydrogen deposit studied both scientifically and industrially. It gives us information on its renewability, on the natural flows involved and therefore on its sustainable exploitation. It is possible to estimate that the cost of operating hydrogen would be

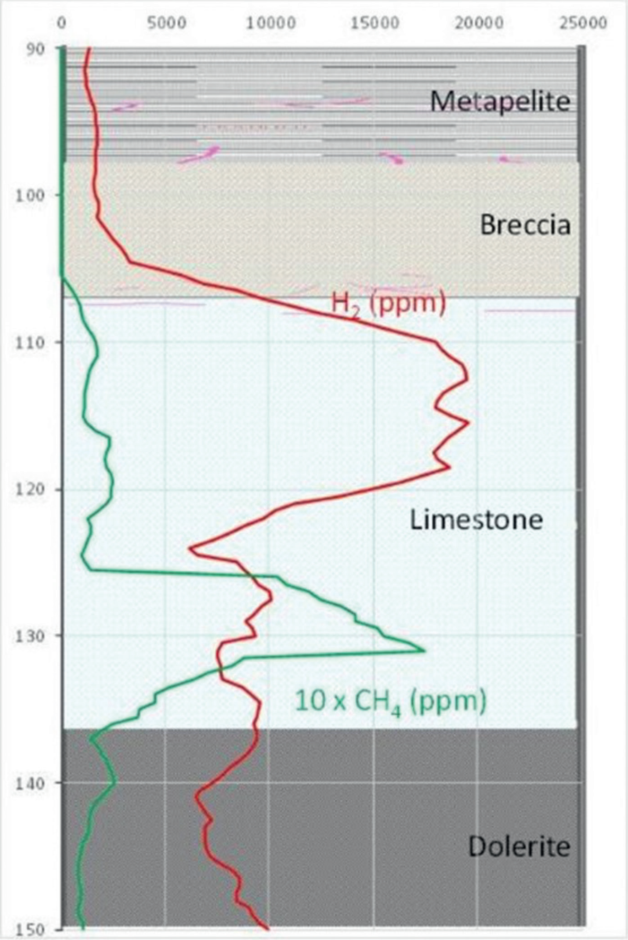

Figure 4 Detail of a 'gas-logging' profile during drilling in a reservoir in the Bourakébougou field. It can be seen that the accumulations of hydrogen and methane are not in diffusive equilibrium, the hydrogen peak being located above that of methane.

less than $1/kg, which is significantly cheaper than any manufactured hydrogen, whether green, grey or blue. Equivalent work is in progress in other continents, in order to be able to compare our knowledge of this Malian field with other fields in the world, which will make it possible to better ensure the industrial and societal interest of R&D for this new field.

Other geological contexts favourable to natural hydrogen

Fumaroles, known for half a century along ocean ridges, are also observed in a terrestrial context, such as on the Icelandic hotspot (Sano et al., 1985) or the active Djiboutian rifts (Pasquet et al., 2022). In Djibouti, the highest natural hydrogen contents were observed in the fumaroles of Fiale in the axis of the Lake Assal rift (>1000ppm). A little further south, in the Lake Abbhé rift, natural hydrogen appears at the level of hot springs where it is associated with helium of mantle origin (Rigollet et al., 2021, Figure 5). The evaluation of the resource in this region, and the possibility of co-producing it with geothermal energy and helium, are the subject of the DjiboutHy project led by the Office Djiboutien de Développement de l'Energie Géothermique (ODDEG), involving French private and public partners (CVA, 45-8 Energy, GEO4U and UPPA) with IFPEN.

Figure 5 Gas sampling in a hot spring by geologists from ODDEG, CVA, GEO4U, UPPA and IFPEN, Lake Abbhé, Djibouti. Chromatographic and isotopic analysis shows the presence of hydrogen associated with helium of mantle origin (Rigollet et al., 2021).

Figure 6 'Fairy circle' in the Sao Francisco Basin (Minas Gerais, Brazil) where geochemical monitoring was carried out to characterize the fluxes of surface hydrogen systems.

Natural hydrogen seeps may also be associated with discrete circular topographic depressions where vegetation struggles to develop. These structures are visible in aerial or satellite photos in the absence of infrastructure; we speak of 'fairy circles' or 'witch circles'.

However, we must be careful not to confuse these structures with similar structures such as dolines, for example, due to other processes (Moretti et al., 2021a). Among the many fairy circles observed in the world, those of the Sao Fransisco basin in Brazil have been the subject of long-term geochemical monitoring which confirms the association between these circular structures and the presence of hydrogen fluxes. natural and variable over time (Prinzhofer et al. 2019, Moretti et al., 2021b). On the ground, in the case of the Sao Fransisco basin, they appear as circular depressions of a few hundred metres in diameter, with a negative topography of a few metres and lean vegetation (Figure 6). The genetic link between these hydrogen emanations and the fairy circles is not yet established.

Natural hydrogen has also been measured near large outcropping faults, such as the seeps observed in France along the northern Pyrenean thrust in the Sauveterre-de-Béarn region (Lefeuvre et al., 2021, Figure 7). Equivalent observations have been made in the Cotentin along the Granville fault. In another geological context, natural hydrogen was measured in the axial fractures of the Lake Assal rift in Djibouti.

The hydrogen flux can be measured at the surface, but the evaluation of the resource requires multi-physical exploration campaigns and drilling to be carried out.

In the imposssibility of being able for the moment to evaluate the quantitative potential of natural hydrogen worldwide, it is, however, possible to estimate the volume of hydrogen which can theoretically be produced per year from the available 'stock of source rock'. It is also possible to measure fluxes at the level of superficial emanations. The 'fairy circles' that have been monitored, such as those in the Sao Francisco basin in Brazil, present significant flows, for example of the order of 7000 m^3/day for a structure of 0.4 km^2 (Moretti et al. ., 2021b). These results combined with others make it possible to estimate, depending on the structures, flows between 50 and 1900 kg/km^2/day (Moretti, 2020).

Figure 7 Soil gas measures with GA5000 in the North Pyrenean Frontal Thrust context (France) — H_2NA project. Perforator is used in percussive mode to avoid artificial increase of H_2 content by drill bit rotation.

To assess the natural hydrogen resource, it is necessary to carry out regional and multidisciplinary exploratory campaigns. These campaigns make it possible to identify targets that must then be drilled to characterize them and measure concentrations and flows in order to model production profiles. Today, drilling data are far too rare and incomplete to respond to such quantification on a global scale.

To carry out an investigation campaign and evaluate the reserves, the basic rules of exploration developed for mining or for hydrocarbons are valid:

- We start from the regional scale to have a 'big picture' understanding of the processes and we gradually refocus the investigations on targets by increasing the spacial resolution
- Multidisciplinary methods are applied, integrating at least geology, geochemistry, geophysics, hydrogeology and microbiology associated with hydrogen.

It is then necessary to adapt the exploratory strategies to the specificities of natural hydrogen. For example, if we are in the case of a hydrogen system fed by the serpentinization of a mafic rock, it will be necessary to consider that at the level of the 'hydrogen kitchen', this alteration is accompanied by an increase in volume, a loss of density and seismic velocity (Mevel, 2003) and a modification of the electrical and magnetic properties by transformation of ferrous iron into ferric iron. If we wish to image 'hydrogen cooking', we therefore understand that we must favour a multi-physical approach, integrating at least magnetic, gravimetric and tomographic data (Figure 8, Rigollet, 2022).

The main H_2 migration pathways are faults corridors, discontinuities and sedimentary drains, that can be highlighted by standard seismic imaging. Moreover, new developments in passive seismic acquisition would enable the recording of seismicity induced by fluid movements. It would be useful to track H_2 migration from the 'kitchen' to accumulations zones and seeps and thus reconstruct its pathway.

The trapping of H_2 is a dynamic accumulation due to bottleneck effects, possibly combined with more classical trapping in anticline structures. The residence and renewal times are quick, contrary to the duration of oil and gas trapping, at the geological scale. Consequently, the standard seismic imaging of the reservoir shape is not enough and must be completed by an estimation of the H_2 flow, based on a dynamic understanding of the accumulation.

The geological risk assessment will require an evaluation of the geological factors that are critical to the discovery of

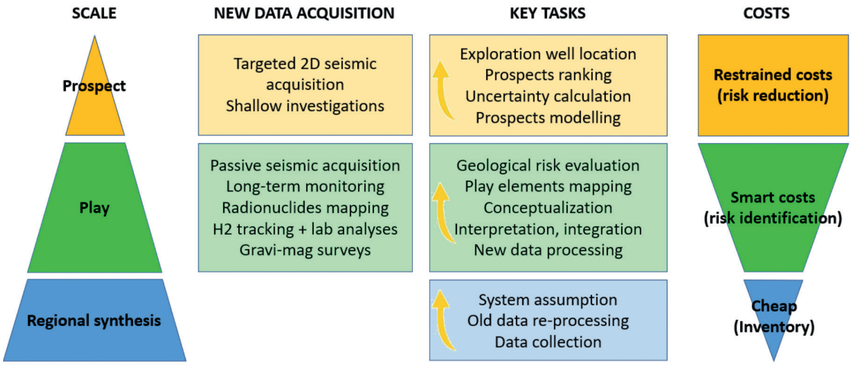

Figure 8 methodological guidelines for natural hydrogen exploration (from Rigollet, 2022).

recoverable quantities of hydrogen in the modelised prospect (H_2 generation, current flow, accumulation, preservation…). The probability of discovery could be defined as the product of the following:

- Probability of hydrogen generation
- Probability of hydrogen current flow
- Probability of hydrogen accumulation
- Probability of hydrogen preservation

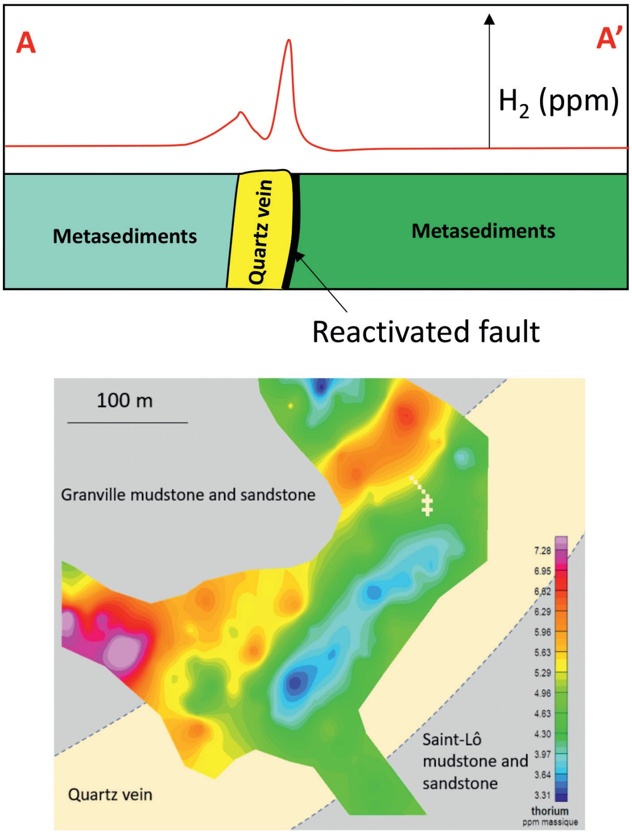

Figure 9 Demonstration of a radiometric anomaly associated with hydrogen emanations greater than 1000ppm, along the Granville fault in the Cotentin, France (work in progress).

The main deliverables of the exploration phase are :

- Conceptual hydrogen system description with a static and dynamic approach
- Mapping of play elements to define prospective areas
- Evaluation of the chance of presence and effectiveness of each element (play chance).

To explore natural hydrogen, we must also innovate by developing new tools or new methods adapted to this type of exploratory programme. For example, Engie and 45-8 Energy are developing new sensors for long-term geochemical monitoring. A French company, Geolinks, implements geophysical sensors for passive seismic monitoring of 'hydrogen cooking' and the rise of fluids. GEO4U and CVA propose to integrate into the exploratory guide the mapping of radionuclides of the zones to be explored, having highlighted several intracratonic sites overlapping between the zones of natural hydrogen emanation and radiometric anomalies (Figure 9 et 10, Prinzhofer et al., 2022).

The same association between hydrogen seeps and positive thorium anomalies have been evidenced in the area where a geochemical monitoring of hydrogen exudation has already been done in Brazil (Figure 10). Every fairy circle, as well as a macrostructure involving these fairy circles, show clearly that hydrogen is correlated with thorium superficial concentrations, without any relation with the topographic variations.

> *Natural hydrogen is a subject that today mobilises many researchers and industries, but there are still many scientific obstacles to understanding the components and the dynamics of the system.*

There are many research teams working on natural hydrogen in particular in France, the USA, Australia and Russia. The preferred subjects are those that provide a better understanding of the origin of natural hydrogen, the characterization and quantification of flows, the transfer of hydrogen and its accumulation. Industrial research teams are focusing more on geophysical observation means, on geochemical sensors and on the implementation of exploratory guides, in order to better assess the resource.

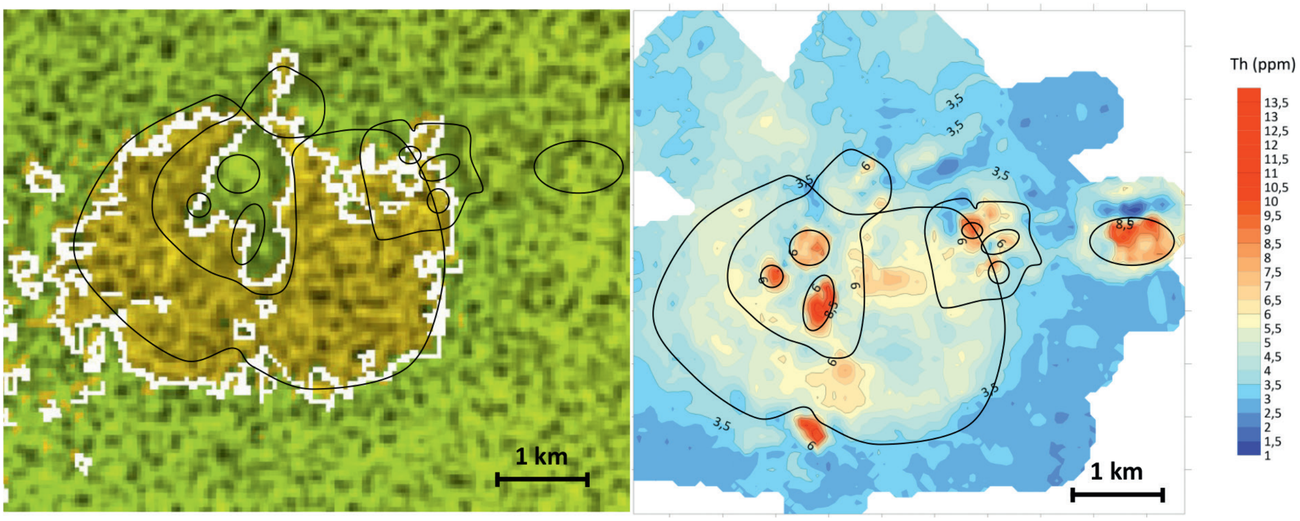

Figure 10 a) topography of fairy circles structures in the Sao Francisco Basin (Brazil) with a macrostructure around 4 km in diameter presenting a slight high in the topography, whereas the fairy circles are presenting depressions. B) Thorium anomalies measured by gamma spectrometry in the same location. Positive thorium anomalies are associated with fairy circles, as well as with the macrostructure (from Prinzhofer et al., 2022).

French researchers are recognised as among the leaders in this field. They met at a congress at the initiative of the CNRS/INSU in 2019 in Paris and then organised the first world congress on natural hydrogen H-NAT in 2021 and 2022. They are involved in numerous European and international research projects. In France, public and private entities have also come together to offer assessments of the natural hydrogen resource at the territorial level. For instance, the Regional Council of New Aquitaine decided in 2021 to launch the H2NA project, intended to characterize the hydrogen systems of the region, assess concentrations and flows, and locate possible accumulations (Blanchard, 2022). The partners in this project (CVA, 45-8 Energy, Engie, Storenengy, UPPA and BRGM) are committed to a three-year schedule. Other equivalent projects are being set up.

Conclusion

Natural hydrogen is a geological reality and a resource of the future. It is produced naturally by the Earth, can migrate and accumulate or escape to the surface and provide explorers with valuable clues. The Malian example of Bourakebougou proves that it can be exploited, with concentrations that exceed 98% and costs much less than those of manufactured hydrogen, which puts this gas in direct competition with hydrocarbons.

The identification of H_2 plays and prospects needs, first of all, a knowledge of the system. What is the source of H_2, which are the migration pathways and the type of accumulation? This terminology echoes that used for the petroleum system, but the oil and gas exploration approach cannot be simply transferred to this new domain of application: the method must be adapted to H_2 and complemented by new developments.

The numerous ongoing exploration projects worldwide are opening a new era, that of underground exploration for a new, carbon-free energy resource.

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