Characterizing Natural Hydrogen Occurrences in the Paris Basin Using OCR-Enhanced Well Database Studies

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Abstract

This study investigates natural hydrogen (H2) occurrences in the Paris Basin, using Optical Character Recognition (OCR) technology to analyze an extensive, yet historically underexploited, well database that contains older drilling records. With the growing demand for carbon-free energy, natural hydrogen, produced through processes like serpentinization and water radiolysis, offers a promising alternative to fossil fuels. However, its potential has been largely unexplored in conventional oil and gas wells. Utilizing the BEPH (Office of Exploration and Production of Hydrocarbons) French database, which includes well logs, mudlogs, and End Drilling Reports (EDRs) in PDF image format, we applied the Tesseract-OCR Engine to convert these documents into searchable formats for efficient data analysis. Our analysis revealed several H2-bearing wells across the French sedimentary basins. The hydrogen occurrences in the Aquitaine Basin correlate with the geological context, but those in the Paris Basin present an anomaly, as their H2 occurrences do not align with the expected geological factors. In the Paris Basin, H2 has been detected in four main formations: the Lusitanian aquifer, Dogger aquifer, Triassic aquifer, and the basement. The highest hydrogen concentration (52 vol%) was found in the Dogger formation. These wells are primarily located along the Bray fault and thrust, indicating a geological influence on H2 distribution. This research demonstrates the effectiveness of OCR in reprocessing historical drilling data for natural hydrogen exploration, highlighting the need for comprehensive exploration methodologies in this emerging field.

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9	Key Points:
10	• Natural hydrogen exploration in former oil & gas province.
11	• Use of OCR algorithm to optimize processing of a large drilling report database.
12	• Indices of a potential new H ₂ system (source, migration, trap) in the Paris Basin.
12	

13

14 Abstract

This study investigates natural hydrogen (H₂) occurrences in the Paris Basin, using Optical 15 Character Recognition (OCR) technology to analyze an extensive, yet historically 16 17 underexploited, well database that contains older drilling records. With the growing demand for carbon-free energy, natural hydrogen, produced through processes like serpentinization and 18 water radiolysis, offers a promising alternative to fossil fuels. However, its potential has been 19 20 largely unexplored in conventional oil and gas wells. Utilizing the BEPH (Office of Exploration 21 and Production of Hydrocarbons) French database, which includes well logs, mudlogs, and End 22 Drilling Reports (EDRs) in PDF image format, we applied the Tesseract-OCR Engine to convert these documents into searchable formats for efficient data analysis. Our analysis revealed several 23 H₂-bearing wells across the French sedimentary basins. The hydrogen occurrences in the 24 Aquitaine Basin correlate with the geological context, but those in the Paris Basin present an 25 26 anomaly, as their H₂ occurrences do not align with the expected geological factors. In the Paris Basin, H₂ has been detected in four main formations: the Lusitanian aquifer, Dogger aquifer, 27 28 Triassic aquifer, and the basement. The highest hydrogen concentration (52 vol%) was found in the Dogger formation. These wells are primarily located along the Bray fault and thrust, 29 30 indicating a geological influence on H_2 distribution. This research demonstrates the effectiveness 31 of OCR in reprocessing historical drilling data for natural hydrogen exploration, highlighting the 32 need for comprehensive exploration methodologies in this emerging field.

33 Plain Language Summary

This study explores the presence of natural hydrogen (H₂) in the Paris Basin, employing Optical Character Recognition (OCR) technology to sift through an extensive database of older drilling records that have not been fully utilized in the past. As the world increasingly seeks carbonneutral energy sources, natural hydrogen, produced through interactions between water and rocks, emerges as a promising alternative to fossil fuels.

Our research focuses on the BEPH (Office of Exploration and Production of Hydrocarbons)
French database, which contains detailed information on drilling activities, but in a nonsearchable PDF image format. OCR is a tool that turns images containing text, like scanned
documents, into text files that we can easily search and analyze.

Our findings indicate the presence of H_2 in several wells across the French sedimentary basins. Particularly intriguing are the results from the Aquitaine Basin, where hydrogen occurrences align with the geological context, and the Paris Basin, which exhibits unexpected H_2 occurrences not directly linked to anticipated geological factors known for H_2 exploration. In the Paris Basin, the highest hydrogen concentration (52 vol%) was discovered in the Dogger formation. These wells are predominantly situated along the Bray fault and thrust, suggesting a geological influence on the distribution of hydrogen.

50 This research underscores the utility of OCR technology in re-evaluating historical drilling data 51 for natural hydrogen exploration. It highlights the necessity for thorough exploration strategies in 52 this nascent yet promising field.

53 **1 Introduction**

54 Geochemical interactions between water and rock on Earth are known to generate molecular hydrogen 55 (H_2) . This process, extensively documented in the literature (e.g., Klein et al., 2020), includes the 56 serpentinization of ultramafic rocks which is a reaction characterized by the hydration of olivine and 57 pyroxene minerals to form serpentine, brucite, and magnetite, accompanied by H_2 production (Malvoisin 58 et al., 2012; Marcaillou et al., 2011; Mayhew et al., 2013; McCollom and Donaldson, 2016). Additionally, water radiolysis, which involves the dissociation of water molecules into H₂ and O₂ due to radiation, also 59 60 contributes to H₂ generation (Lin et al., 2005; Sauvage et al., 2021; Warr et al., 2019). These geochemical 61 processes are not only crucial for understanding Earth's deep microbial ecosystems but also play a significant role in the abiotic synthesis of organic molecules, as evidenced by numerous studies (Etiope et 62 al., 2015; Fiebig et al., 2007; Johnson et al., 2015; Lin et al., 2005; Sherwood Lollar et al., 2006). 63

In light of the global shift towards sustainable energy sources, naturally occurring H_2 has garnered 64 significant attention as a viable, carbon-neutral energy alternative to traditional fossil fuels. This interest 65 is reflected in recent research exploring the potential of H_2 in various geological contexts (Donzé et al., 66 67 2020; Moretti et al., 2021; Prinzhofer et al., 2018; Smith et al., 2005; Truche et al., 2018). Current 68 exploration methodologies for targeting H_2 in potential geological reservoirs predominantly rely on soil 69 gas analysis at a depth of approximately 1 meter. However, this approach is somewhat limited in scope 70 and does not encompass a comprehensive and efficient methodology. This gap in methodology 71 underscores the need for more integrated and systematic exploration strategies, as highlighted in recent 72 studies (Lefeuvre et al., 2022, 2021).

73 In the domain of petroleum geology, the presence of molecular hydrogen in natural reservoirs has

- historically been underappreciated. This oversight can be attributed to the fact that H₂ was rarely detected
- 75 in the multitude of wells drilled globally for oil and natural gas exploration, as noted by Gaucher (2020).
- 76 The standard geochemical sensors employed in these wells were primarily calibrated for detecting fossil
- hydrocarbons, such as methane, with less sensitivity or focus on H_2 .
- 78 However, emerging research and field data have begun to challenge this long-standing viewpoint. Recent 79 studies have identified significant concentrations of H_2 in various geological settings across the world. 80 Notable examples include the "Bougou-1" well, which was drilled in 1987 in Mali, the concentration of H₂ was found to be 98 vol% (Maiga et al., 2023). The "Tisovita well", which was drilled before 1978 in 81 Romania, the concentration of H₂ was found to be 28.7 vol% (Mitrofan et al., 2021). The "#1 Wilson 82 83 Well" drilled in 2006 in the Kensas (USA). the H₂ concentration measured was 10.0 vol% (Newcombe, 84 1935). The well EVDD008 drilled in the Yilgarn Craton, in Australia, the H₂ concentration measured was 85 42,7 mol% (Boreham et al., 2021). At the "Copper Cliff well", in Canada, the H₂ concentration ranged 86 from 9.9 to 57.8 vol% (Sherwood et al., 1988)
- These findings underscore the potential for natural hydrogen reservoirs and highlight the necessity for refined geochemical analysis techniques in hydrocarbon exploration, particularly for the detection and quantification of H_2 . The implications of these discoveries are significant, not only for understanding subsurface geochemical processes but also for evaluating the potential of H_2 as an energy resource in the context of a transitioning global energy landscape.
- 92 In France, the exploration and production of oil and gas have led to the drilling of over 5,000 wells. 93 However, none of these wells have been designed for natural hydrogen exploration. The BEPH (Office of 94 Exploration and Production of Hydrocarbons) database used in this study is composed of well logs, 95 mudlogs and End Drilling Reports (EDRs) which are in PDF image format. Manual examination of each 96 well would take a considerable amount of time, so an Optical Character Recognition (OCR) algorithm 97 (Tesseract-OCR google Engine; Smith, 2007) was used to transform these scanned image PDFs into 98 searchable PDFs.
- 99 Through the analysis of an extensive dataset, we have identified several H₂-bearing wells distributed 100 across the French sedimentary basin (Fig. 1). These wells can be categorized into two distinct groups: 101 those situated in the eastern region of Paris, and those in the southern part of the Aquitaine Basin.

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Figure 1 : Geological map of France highlighting the locations of wells where H_2 occurrences were detected using OCR.

105

In the Aquitain Basin, the observed concentrations of H_2 can be linked to the geological context. Indeed, these wells are at the vicinity of a mantle body (peridotite) present at relatively shallow depths and are also near major drain facilitating fluid migration (Chevrot et al., 2022, 2018; Jammes et al., 2010; Lehujeur et al., 2021; Tugend et al., 2014).

In the Paris Basin the H_2 -bearing wells are an enigma, as the H_2 occurrences cannot be readily explained by the local geology. Consequently, our study will focus on investigating the underlying geological factors influencing H_2 presence in the Paris Basin. Our findings reveal promising indications of H_2 potential in the Paris Basin. This intracratonic basin is characterized by a geologically diverse basement, comprising peridotite rocks at relatively shallow depths (less than 4 kilometers; Averbuch and Piromallo, 115 2012). Notably, the basin is intersected by major faults that extend through both the basement and the 116 overlying sedimentary cover, which could be conduits for H_2 migration and accumulation.

117 The primary objective of this study is to demonstrate the efficacy of applying OCR technology to old but 118 extensive drilling datasets. This approach enables the rapid identification of unexpected but promising 119 areas for H2 exploration. The Paris Basin, with its unique geological features, serves as a focal point for 120 this investigation, potentially positioning it as a H₂-rich geological province. Our research underscores the 121 value of innovative data processing techniques in enhancing the efficiency and scope of geological 122 exploration, particularly in the context of emerging energy resources like natural hydrogen.

123 **2 Geological context**

124 The Paris Basin, a prominent intracratonic basin, encompasses approximately 3 km of sedimentary 125 deposits spanning from the Trias to present. These deposits overlie the Variscan suture zones, a 126 significant geological feature from the Upper Paleozoic era, as documented in several studies (Fig. 2a; 127 Curnelle and Dubois, 1986; Delmas et al., 2002; Mégnien et al., 1980; Pomerol, 1978). The basin is 128 geographically bounded by four Paleozoic massifs: the Armorican Massif to the south, the Bresse Graben 129 to the southeast, the Vosges Massif to the east, and the Ardennes Massif to the north. The Paris Basin 130 exhibits a distinct geological structure, characterized by concentric outcrops of Meso-Cenozoic rocks. 131 This structure is the result of a series of depositions and erosional processes, as detailed in various studies 132 (Beccaletto et al., 2011; Delmas et al., 2002; Guillocheau et al., 2000). Additionally, the basin extends to 133 connect with the London and Belgium Basins to the northwest and north, respectively (Dercourt et al., 134 2000).

Two major fault systems are prominent in the Paris Basin: the Sennely fault and the Bray fault system
(Fig.2). The latter is a N130° dextral strike-slip fault that impacts the sedimentary cover (Matte and Hirn,
1988, Raoult and Meilliez, 1987).

138 The basement of the Paris Basin is a lithological and structural inheritance of the Variscan orogeny, 139 which occurred during the Carboniferous period following to the north-south convergence of the 140 Avalonia and Gondwana plates, culminating in the closure of the Rheic Ocean (Averbuch and Piromallo, 141 2012; Ballèvre et al., 2009; Matte, 1986). Seismic tomography beneath the Paris Basin has revealed 142 anomalies in the upper mantle, with Vp velocities oriented along NW-SE axes. These anomalies, situated 143 along the Bray fault, are associated with the Variscan suture zone, indicative of a Variscan paleoslab (Fig. 144 2b; Averbuch and Piromallo, 2012; Cazes et al., 1986). Recent P-wave seismic tomography studies 145 suggest the presence of a subducted paleo-slab beneath a segment of the Bray Fault (Autran et al., 1994; Averbuch and Piromallo, 2012; Matte and Hirn, 1988). Complementary gravimetric and magnetic data 146 147 along this major fault structure have identified anomalies correlating with granite intrusions (Baptiste,

148 2016; Thébault et al., 2006). The Lizard-Rhenohercynian suture zone, as illustrated by the Lizard
149 ophiolitic complex in southern part of Great Britain, comprises both ultramafic (peridotite, serpentinite)



and crustal rocks (amphibolite, gneiss, etc.), further elucidated in various studies (Cook et al., 2002, 1998;

151 Leake and Styles, 1984; Roberts et al., 1993).

152 Figure 2: a) The structural map of the Paris Basin displays the main units and the surrounding

153 crystalline massifs (modified from Baptiste, 2016). In this map, the red stars represent the wells showing

154 evidences for the presence of H_2 , as identified using the OCR algorithm developed in the study 1 -

- 155 Longeuil; 2 Betz; 3 Montreuil Aux Lions; 4 Cramaille; 5 Connantre 1 and 2; 6 Grandville; 7 -
- 156 Saint Martin de Bossenay; 8 Hericy; 9 Le Luteau; 10 Coubert. b) Crustal-scale cross-section
- 157 through the Variscan orogenic system and the Paris Basin based on the Northern France crustal cross-
- 158 section (modified from Averbuch and Piromallo, 2012 and Matte and Hirn, 1988).

159

160 The Parisian Basin, primarily recognized for its hydrocarbon reserves, is characterized by over 3,000 161 boreholes that have been instrumental in delineating its structural framework. This basin encompasses several key aquifers and reservoir (Fig. 3). These include: (i) The Triassic Sandstone aquifer, capped by 162 163 mudstone and an evaporitic formation, particularly in the central part of the basin with a temperature of 120°C (e.g., Montmirail well; Torelli et al., 2020); (ii) The Middle Jurassic limestone aquifer, containing 164 165 geothermal waters with temperatures ranging between 50 to 80°C and (iii) the Lower Cretaceous green sandstone aquifers. A recent study calculated the geothermal gradient at T = 10 + 34.9*z, (where T is the 166 temperature in °C and z is the depth in km), derived from an analysis of existing well data and aquifer 167 168 temperatures (Bonté et al., 2010; Pinti and Marty, 1998).

Furthermore, Pinti and Marty (1998) identified a helium anomaly in the Dogger Formation, characterized by an unusual excess of mantle-derived ³He in the Dogger aquifer, particularly near the Bray fault (Fig. 3). This fault is hypothesized to serve as a conduit for helium-rich fluids, facilitating their migration through the low-permeability shales that separate the Triassic and Dogger aquifers (Worden and Matray, 1995).





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178 Integrating geodynamic, geophysical, and geochemical data, it is assumed that a paleo-slab, 179 composed of mantellic and metamorphic rocks, is connected to the basin via the Bray fault (Bril et al., 180 1994). The detection of mantle-derived ³He in the Dogger aquifer suggests a deep structural connection to
these formations (Fig. 3).

The Paris Basin's geology is particularly promising for H_2 exploration. This potential is attributed to the presence of ultramafic rocks undergoing serpentinization, water radiolysis in granite bodies, the existence of preferential pathways for fluid migration and the excess of ³He. Additionally, numerous reservoirs within the basin exhibit favorable porosity and permeability characteristics, making them suitable candidates for hydrogen storage. Collectively, these factors underscore the basin's potential as a complete H₂ system from sources to traps.

188

189 **3 Methodology**

190 3.1 Data origin

191 The database used in our research, whose management is delegated to the French Bureau of Hydrocarbon Exploration and Production, encompasses a comprehensive collection of well data. This repository, 192 known as the BEPH database, contains 5,139 records dating back to 1927, covering hydrocarbon 193 194 exploration activities in both metropolitan France and its overseas territories. However, a significant 195 portion of the database, approximately 36.8 % (equating to 1,893 files), is missing, primarily consisting of 196 End Drilling Reports (EDRs) in PDF format. The database incorporates a diverse array of data, including 197 scanned EDRs, well logs (such as Gamma-ray, etc.), mudlogs, seismic coring, and other relevant 198 documents (site logs, tests, etc.). For the purposes of our study, we concentrated exclusively on the 199 analysis of the EDRs, which comprise 3,246 scanned documents in PDF format, with individual files 200 ranging from 10 to 300 pages.

201 3.2 Pytesseract Screening RFS

202 Given the image-based nature of these documents, conventional keyword search techniques are infeasible, 203 and manual examination would be prohibitively time-consuming. To address this challenge, we employed 204 Optical Character Recognition (OCR) technology. Specifically, we utilized Pytesseract, a Python-based 205 OCR tool that integrates Google's **Tesseract-OCR** Engine (available at 206 https://github.com/madmaze/pytesseract), to convert the EDR PDFs into searchable formats (Fig. S1). 207 The process requires preliminary process of each EDR. As Pytesseract is incapable of directly processing

208 PDF files, we first converted each page of the EDRs into image files (PNG, JPG, etc.) using an initial 209 conversion tool. This tool transforms the scanned PDF images into Python Imaging Library (PIL) format, with each PIL image representing a page from the EDR. Subsequently, Pytesseract OCR processes each page, converting the text into a searchable PDF format, which we labeled as "EDR reference".

To further analyze the newly created searchable database, we employed PyMuPDF, a Python library designed for extracting, analyzing, converting, and manipulating PDF file data. This library facilitates keyword filtering within each searchable PDF, allowing us to extract the file name and the specific page number where the keyword appears, and store this information in a .txt file. This approach significantly streamlines the process of identifying relevant files and their locations, thereby enhancing the efficiency of quality checks and validation. In our case study, we focused on keywords such as "H₂", "Hydrogène".

218 **4 Results**

Applying the OCR algorithm to the Paris Basin well database, 141 occurrences were found in the French EDR database containing "Hydrogen" ("hydrogène" in French) and/or "H₂". Initially, each time the word "hydrogen" is detected, *Pytesseract* records it in the results file. However, in the boreholes of the Paris Basin, there are many occurrences of Hydrogen Sulfide (H₂S) which biases the results. The inability to distinguish between "H₂" and "H₂S" can lead to problems, therefore rapid human verification is essential. The main results have been reported in the supplementary materials.

After the validation process, only 11 wells of the Paris basin, the presence of the " H_2 " or "Hydrogen" keyword is mentioned in their EDR (Tab. S1). In EDR, when hydrogen is detected, geologists propose various hypotheses to explain its presence, such as: i) hydrogen arising from tool deterioration, ii) hydrogen of unknown origin, iii) hydrogen detected but not quantified, iv) trace concentrations of hydrogen in the background gas, and v) hydrogen identified using neutron porosity techniques.

The Longueil 1 Well (drilled in 1972), has detected H₂ and N₂ trace (no concentrations reported in the EDR) during the mud gas logging at 3 differents depths i) In Lusitanian formation at 988 to 1241m and composed of limestone (porosity of 19.2 %); ii) in the Middle Jurassic at 1241 to 1347m and composed of Massigny Marles (porosity ranging from 9 to 19 %) and iii) In Triassic formation at 1916 to 1981 m and composed of Saint Maur Red Clay. The geologist in charge doesn't explain the origin of H₂.

The Betz 101 drilling well (drilled in 1963) revealed a concentration of H₂ gas ranging to 3 to 6 vol% in Lustianian formation at a depth of 1325 to 1335 m, however the report does not describe the methodology used for this gas analysis. The Lusitanian formation is mainly composed of limestone with a porosity ranging from 7 to 11 %. The geologist in charge revealed that detecting high H₂ concentrations in the Lusitanian formation in this area is common, but its origin remains unknown.

The Montreuil Aux Lions 1 drilling well (drilled in 1988) detected H₂ gas in a bottle sample of 1 liter taken at a depth of 2165-2175 m corresponding to Marles to Calcareous Clay formation in the Dogger. The pressure inside the bottle was 25 bar. The gas measurement revealed a gas composition of H₂ 52 vol%, CH₄ 42 vol%, C2 4.3 vol%, C3 0.9 vol% and nC4 0.19 vol%. The EDR did not provide any hypothesis about the origin of this H₂ concentration.

- The Cramaille 101 drilling well (drilled in 1961) detected H_2 during the mud gas logging while crossing the Lusitanian formation, which is composed of limestones with a porosity of 16.7 % and a permeability ranging from 0.1 to 12 mDy. However, the EDR did not indicate the exact concentration of H_2 gas, and there was no explanation provided for this gas detection.
- The Connantre 2 drilling well (drilled in 1981), have detected the presence of H₂ and nitrogen from 1508 to 1533 m, the Dogger formation was composed of limestones with porosity ranging from 7 to 11 %. The useful height of the formation was 9.5 m. The methodology for gas analysis is not described in the EDR.
- The Grandville 109 drilling well (drilled in 1982) has identified during the mud gas logging
 the presence of H₂ at two different depths i) The Aalenian formation is made up of clayey
 limestones and ranges from 1159 to 1725 m. The background mud gases have a H₂
 concentration that ranges from 0.25 to 0.65 vol%. ii) The Triassic formation, spanning from
 2053 to 2555 m in depth, consists of dolomite, clay, and evaporite. Within the background
 gases, a concentration of 0.2 vol% of H₂ was detected.
- The Coubert 1 drilling well (drilled in 1986), has detected H₂ during the mud gas logging in the bottom hole at 2547 m depth but no concentrations are reported in the EDR. The Hercynian formation is mainly composed of Gneiss-type rocks. The gas composition recorded showed unusual fluctuations, with a disappearance of C4 hydrocarbons and a reduction in C3 and C2 concentrations, but an increase in total gas, along with the release of H₂. The main hypothesis proposed by geologists is gas production linked to tool alteration.
- The Luteau 1 drilling well (drilled in 1986) detected H₂ and CH₄ during the mud gas logging
 while crossing Keuper from 2569 to 2578 m, which is mainly composed of clay but locally
 contains anhydrite. Unfortunately, the gas chromatograph (GC) used was unable to

distinguish between H_2 and CH_4 . The H_2 - CH_4 compounds were detected during core sampling in the background gas ranging from 0.6 to 1.1 vol%.

- The Hericy 1 drilling well (drilled in 1986) detected H₂ and CH₄ in the gas background during the mud gas logging. However, the GC cannot distinguish between them, similar to Luteau 1. This potential H₂ detection occurs during the crossing of Liassic formation composed of calcareous clay. The gas Background is ranging from 1 to 3.5 vol%.
- 278 The Saint Martin de Bossenay 17 drilling well (drilled in 1976) detected H₂ traces in two 279 different formations. I) The upper Triassic (Rethien) Clayay formation was tested at a depth 280 of 2084 m. They detected H₂ gas but did not report its concentration. The well was open for 32 minutes, during which they recovered 20 liters of gas and 0.5 liters of mud. ii) The Upper 281 282 Triassic formation, made up of sandstones, clay, and anhydrite rocks, was tested at a depth of 283 2295 m. During the test, the well was open for 37 minutes and recovered 410 liters of mud, a 284 small quantity of gas and 120 liters of gasified mud. They also reported that the gas was detected in the same formation as the Saint Martin de Bossenay 201 well. The pressure of 285 196 kg/m^2 was unstabilized at 2321m. 286
- The Jeumont 1 drilling well (drilled in 1963), has detected H₂ in mud gas at two differents depths i) At a depth of 4443 m in the Upper Devonian, there is a composition of quartzite and argillite. The detected H₂ is present in background gas at a concentration of 0.5 vol%. ii) At a depth of 4807 m, the Middle Devonian is mainly composed of quartzite and shale. The H₂ concentration is also in background gas at a concentration of 1.8 vol%. The H₂ concentration for both depths was unexplained.

All the data originate from wells drilled at various times during the 20th century and have not been previously correlated. To comprehend the natural hydrogen potential of this region, it is essential to integrate these data with the knowledge acquired in recent decades and with contemporary geological studies of the Paris Basin. Consequently, a comprehensive compilation of both geochemical and geological knowledge will also be carried out.

298 **5 Discussion**

299 300 5.1 Hydrogen Detection and Origin Hypotheses in Various Geological Formations of the Paris Basin

According to the OCR algorithm, the drilling wells' location is concentrated within an area of 8600 km^2 in the middle east of the Paris Basin (Fig. 2a). The H₂ concentration in the Paris Basin is not randomly distributed throughout the formations. It is mainly present in three formations: Lusitanian, Dogger, and Keuper.

305

5.1.1 The Lusitanian reservoir

Three sites north of the Bray fault—Longueil, Betz, and Cramaille—reported H_2 content while intersecting the Lusitanian formation. According to the Initial interpretations, this would indicate a potential tool degradation. However, H_2 was also detected in the same formation in Longueil and Betz wells. The well geologist revealed in the Betz 101 EDR that detecting H_2 in the Lusitanian formation is common. Therefore, the hypothesis of H_2 production by steel corrosion in Cramaille could be discarded.

An alternative hypothesis involves the production of H_2 through mechano-radical reactions occurring during drilling. This suggests that the mechanical crushing of rocks results in the production of fresh Si surfaces that are highly reactive with water, generating H_2 (Hirose et al., 2011; Kita et al., 1982). Nevertheless, the Lusitanian is mainly composed of limestone and contains little to no silica. Lefeuvre (2022) conducted a limestone grinding experiment in a confined atmosphere and demonstrated that this rock did not produce H_2 . This leads to questioning the origin of the H_2 .

317

5.1.2 The Dogger reservoir

318 Investigations into the Dogger formation, specifically at the Montreuil Aux Lions, Connantre, and 319 Grandville wells north of the Bray Fault, have identified H₂ concentrations. At Montreuil Aux Lions, a 320 sampling approach was employed, utilizing a specialized bottle to collect a sample from within the 321 Dogger formation. The analysis of this sample revealed 52 vol% H_2 concentration within a 1-liter vessel 322 under 25 bar pressure. Notably, the hypothesis of H₂ production by mechano-radical processes was 323 discarded due composition of Dogger formation, corresponding to Oolitic limestones. Moreover, this 324 conclusion is reinforced by the fact that the sampling procedure did not take place at the same time as the 325 drilling activities.

Gases have been sampled in Dogger formation from geothermal production wellheads during both artesian flow and production under pumping (Fig. 3). These wellheads exhibited pressure ranges from 4 to 12 bar, indicative of a monophasic state at depth for such fluids (Marty et al., 1988). Marty et al. (1988) 329 conducted analyses on 34 dissolved gas samples, which were isolated using a vacuum flask vessel half-330 filled with the separated liquid phase containing the dissolved gases. They recorded H_2 concentrations reaching up to 12 x 10⁻⁵ mol/l (at 71 °C and 9.81 bar). The prevalence of steel corrosion as possible origin 331 332 for elevated H₂ levels remains a consideration. However, only 9 out of the 34 wells displayed 333 concentrations exceeding 1 mol/l, suggesting a different origin for the elevated H_2 concentrations. These 334 observations support the existence of an H₂-rich aquifer in the Dogger.

335 The Dogger aquifer is also known for the presence of sulfate-reducing, methane-producing bacteria, which are mainly thermophilic (Fouillac et al., 1990). Isolated methane-producing bacteria from 336 337 Mellaray's well have shown the capability to thrive utilizing H_2 and CO_2 as their sole carbon and energy 338 sources (Daumas et al., 1986; Marty et al., 1993). This observation suggests that the H_2 measured in the 339 Dogger is a source of energy for bacterial communities and it raises questions about the deeper origin of 340 H₂.

341

5.1.3 The Triassic reservoir

342 In the Upper Triassic formation situated south of the Bray Fault, H_2 presence was documented in the 343 drilling wells at Le Luteau and Saint Martin de Bossenay. This formation is characterized by a composite 344 of clay, anhydrite, and sandstones.

345 At Le Luteau, the detected background H_2 concentrations during the drilling might be associated with 346 mechano-radical mechanisms, a hypothesis supported by the silica-rich composition of the rocks.

347 At Saint Martin de Bossenay, H_2 detection occurred during post-drilling production testing, rendering the 348 mechano-radical hypothesis less plausible for this site. It is important to note that no acidification of the 349 well was performed prior to conducting this test. In this formation, the anhydrite may act as a promising 350 sealing rock for trapping H₂.

351

5.1.3 H_2 detection through the basement

352 Lastly, two other wells reached the basement at Coubert and Jeumont, and they revealed the concentration 353 of H₂ at depth. The Coubert well, situated to the south of the Bray fault, traverses a geological formation 354 composed predominantly of Gneiss and quartzite with galena intrusions. At this site, geologists cannot 355 differentiate between methane and H_2 gases due to instrumental reasons. However, a notable diminution 356 in C4 to C2 hydrocarbon concentrations suggests a possible H_2 degassing, potentially originating from 357 drilling tools or the surrounding rock matrix as suggested in the EDR. The Jeumont well only revealed H_2 358 concentration during the core sampling of quartzite formation. This observation supports the hypothesis 359 of H₂ generation through mechano-radical processes.

360

361 Due to limited EDR data, deciphering the source of H₂ remains challenging. Geological and geochemical
 362 contextualization may offer a starting point for further analysis.

363

5.2 Geological Trends and Structural Analysis of Drilling Wells in the Paris Basin

Initial observations indicate that the wells Longueil 1, Betz 101, and Montreuil Aux Lions 1, located to 364 365 the north of the Bray fault delineate a trend of N130° orientation. This aligns with the orientation of the 366 Bray fault, albeit situated approximately 20 km northward. This trend is highlighted and well correlated with the map of the vertical gradient of the pole-reduced magnetic anomaly extended to 600 m (Fig. 4; 367 368 Baptiste et al., 2016). The vertical gradient technique can be used to identify lithological features, 369 delineate contacts and discern structural variations between distinct geological formations (Baranov, 370 1953). This pattern could correspond the Bray Thrust located by Averbuch and Piromallo (2012) but 371 regarding the seismic cross section this discontinuity doesn't affect the sedimentary cover (Fig. 2b).

Contrastingly, the wells Cramaille 101, Connantre 1, 2, and Grandville 109 do not align with this specific trend but are situated within a large regional anomaly. This anomaly has been linked with magmatic rocks, with the axis of this anomaly appears to be correlated with the closure of the Rhenohercynian ocean during the Carboniferous period (Thébault et al., 2006).

Regarding the wells located south of the Bray fault (Fig. 4), their distribution is more dispersed and their association with structural features is more complex. These wells are part of the Bloc Every-Tonnerre, an area where the basement has been mapped through magnetic and gravimetric analysis (Baptiste, 2016).

379 The drilling well Hericy 1 is located on the Evry Fault, while Saint Martin de Bossenay 17 is located on

the Lalaye-L Fault (Baptiste, 2016). The map reveals the presence of basic rocks along the Evry fault, and a combination of gneissic or granitic rocks interspersed with Néoproterozoic/Paleozoic formations along the Lalaye-L Fault. The two other wells, Le Luteau 1 and Coubert 1, while not associated with any

383 specific faults, are located above granodiorite bedrock.

384 While the majority of the wells' locations appear to be correlated with fault lines, further investigation is 385 needed to determine if these faults could act as conduits for fluid migration.



Figure 4 : Map of the vertical gradient of the magnetic anomaly reduced to the pole and extended to
600 meters. The black circle corresponds to the H2-bearing well (Modified from Baptiste, 2016).

5.3 Characteristics and Deep Fluid Helium Studies of the Dogger Aquifer in the ParisBasin

The Paris Basin is characterized by several key aquifers, including Triassic sandstones, Dogger limestones, Lusitanian limestones, and Albian sandstones (Fig. 3). Among these, the Dogger aquifer has been the primary focus of numerous studies, primarily due to its notable helium concentration (Bril et al.,

393 1994; Castro et al., 1998a, 1998b; Marty et al., 1993, 1988; Pinti and Marty, 1998)

The Dogger aquifer exhibits a relatively consistent helium concentration and isotope ratio, though it presents minor variations in water chemistry (Marty et al., 1993). According to studies by Marty et al. (1988), the reported free gas concentration of helium in the aquifer ranging from 1.02 to 4.65 x 10^{-5} mol/l (at 70 ± 10 °C, the pressure is not reported). Additionally, isotopic analyses have revealed a distinct excess of ³He, indicative of a mantle-derived origin. Pinti and Marty, (1993) suggest that the enrichment in mantle-derived ³He, alongside high ⁴He concentrations, may be attributed to the influx of basement fluids into the Dogger and potentially to in-situ production within the Middle Jurassic formation. To explain the high helium concentration in the aquifer, one plausible explanation is the presence of a preferential migration pathway, notably the Bray-Vittel Fault (Fig. 3). This fault affects both the sedimentary cover and the basement, and can serve as intermittent vertical drains for helium-rich fluids across the 700m of low-permeability rocks that separate these aquifers (Bril et al., 1994; Pinti and Marty, 1998; Worden and Matray, 1995).

406 The analysis of the Dogger aquifer has revealed fluid migration along the major faults, while our OCR 407 analysis has pinpointed wells exhibiting H_2 anomalies correlated with these faults. These findings suggest 408 the existence of a potentially fertile H_2 system.

409 5.4 A putative H_2 system ?

410 The potential for H₂ exploration in the Paris Basin is closely linked to its geodynamic characteristics and 411 structural features, particularly those associated with the closure of the Lizard-Rhenohercynian ocean. 412 This suture zone is composed of ultramafic rocks, including peridotites and amphibolites, which are able 413 to produce H_2 through hydrothermal reactions such as serpentinization. The current temperature near the 414 basin/basin discontinuity is approximately 120°C (Pinti and Marty, 1998), significantly lower than the 415 250 to 300°C typically required for optimal H_2 production kinetics (Malvoisin et al., 2012; McCollom et 416 al., 2016). However, a significant aspect of the Lizard complex is its composition, consisting of dunite, 417 which is largely serpentinized and contains magnetite (Leake and Styles, 1984). Recent findings by 418 Geymont et al. (2023) have revealed the potential of magnetite to facilitate H₂ production at relatively 419 lower temperatures through hydrothermal alteration processes. Therefore, in this geological context, 420 serpentinized dunite, particularly rich in magnetite, may represent a viable source rock for H₂ generation. 421 This raises the question of the actual production of H_2 .

422 To the north of this closure zone, magnetic and gravimetric anomalies suggest the presence of granitic 423 formations, which can facilitate H_2 generation via radiolysis of water, a process independent of rock 424 temperature (Lin et al., 2005b; Sherwood Lollar et al., 2006).

A key factor in these H_2 -producing processes is the presence of a water source, originating from a recharge zone. The basin's sedimentary cover and basement are influenced by regional tectonic lineaments, notably the Bray-Vittel and Rouen-Couy faults, which may act as conduits for vertical fluid migration. Studies on noble gases suggest a vertical flow of fluids through the Rouen-Couy fault, allowing water infiltration into the sedimentary layer through this discontinuity, eventually reaching the basement (Pinti and Marty, 1998; Rouchet, 1981). Additionally, the Paris Basin hosts multiple aquifers,

431 potentially serving as sources for water essential to these reactions.

Helium analysis within the basin has provided insights into fluid migration patterns, with mantle-derived helium detected in the Dogger aquifer, as evidenced by the excess of 3 He. The H₂ produced in the 434 basement may migrate through the Bray-Vittel fault and major detachments, and reach the aquifer. The

- 435 Triassic and Jurassic formation, are marked by evaporitic rocks and clayey rock, offering effective sealing
- 436 properties for H₂ entrapment. Furthermore, the temperature in the Triassic formation seems suitable for
- 437 H₂ trapping over geological time, being too hot for microbial activity and too cold for efficient abiotic
- 438 reactions involving H_2 (Lefeuvre et al., 2022).

439 The Paris Basin stand for a good case study for natural H_2 exploration. All the essential components 440 necessary for the establishment of a H_2 system are gathered: i) a water source facilitating H_2 production,

441 ii) iron-rich and granitic rocks, iii) preferential migration pathways along faults impacting both the

- 442 basement and sedimentary layers, and iv) efficient reservoirs characterized by the presence of clay,
- 443 evaporites, and aquifers, which may act as effective seals.

444 **5** Conclusions

445 After processing the BEPH database with an OCR algorithm, we discovered multiple wells that detected 446 H_2 gas. First, across France, several drilling wells have been identified where H_2 concentrations are 447 documented in their EDR. The distribution of these wells is not random in terms of the geological 448 context, they are frequently situated near mantle bodies at shallow depths. This discovery should lead to 449 further research.

450 In the Paris Basin case, the OCR algorithm revealed four main formations in which H_2 has been detected: Lustianien formation, Dogger aquifer, Triassic aquifer and in basement. A maximum of 52 vol% of H₂ 451 452 was obtained in the dogger, whereas up to 6 vol% has been measured in the lusitanian. In the others 453 formations, the concentration of H_2 is not measured, but its presence is still reported. These wells are 454 situated in a small area in the central-east of the Paris Basin, and their distribution does not seem random. 455 All of them are situated along the Bray fault and the Bray thrust, which are N°130 faults that affect both the basement and the sedimentary cover. The basement comprises rocks that have the potential to be 456 sources of H₂, such as peridotite or granitic rocks. An excess of ³He is reported in the Dogger Formation, 457 458 suggesting a contribution from the mantle and deep fluid migration. Finally, the evaporite and clay 459 formation reported in the basin represent a promising trap for H₂.

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