

Vaux-en-Bugey (Ain, France): the first gas field produced in France, providing learning lessons for natural hydrogen in the sub-surface?

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Abstract – The former Vaux-en-Bugey field, first French methane production from early 20th century, is revisited as a case study to address the present generation and accumulation theories for gases like hydrogen and helium. The volume of the initial gas in place is estimated to be 22 million m³. Based on a composition of 5% of hydrogen and 0.096% of helium, the volumes of these gases in the field were respectively around 1.1 million m³ for hydrogen and 24 000 m³ for helium. The different hypotheses of hydrogen sources are reviewed: serpentinization, hydro-oxidation of siderite, water radiolysis, bio-fermentation, mechanical generation, degassing from depth through faults, steel corrosion. For helium generation, the different sources of radioactive minerals and intermediate accumulations are examined. The most probable scenario is the hydrogen production by water radiolysis and helium production by radioactive decay in or near the basement, migrating through deep faults, stored and concentrating in an aquifer with thermogenic methane, then flushed by methane into the gas field, during Jura thrusting. New measurements with portable gas detector, incomplete but including hydrogen, on a former exploration well with accessible flux of gas, give the opportunity to comment gas saturation evolution more than a century after the 1906 discovery. The decreasing of hydrogen content since the discovery of the field is probably due to Sulphate-Reducing Bacteria activity.

Keywords: gas field / gas / natural hydrogen / helium / Jura / Bugey / Vaux-en-Bugey

R esum e – Vaux-en-Bugey (Ain, France): le premier gisement de gaz exploit e en France, source d'enseignement sur l'hydrog ene naturel dans le sous-sol ? L'ancien gisement m ethane de Vaux-en-Bugey, premi ere exploitation fran aise du d ebut du XX^e si ecle, est revisit e comme cas d' etude pour confronter les th eories actuelles de g en eration et accumulation de gaz tels que l'hydrog ene et l'h elium, pr esents dans le gisement. Les roches m eres de ces compos es sont discut ees ainsi que des hypoth eses de mise en place et r etention. Le volume initial du gisement est estim e  a 22 millions de m³. Sur la base d'une composition de 5 % d'hydrog ene et 0,096 % d'h elium, les quantit es accumul ees dans le gisement pour ces deux gaz sont respectivement 1,1 millions de m³ pour l'hydrog ene et 24 000 m³ pour l'h elium. Les diff erentes hypoth eses de sources de l'hydrog ene sont revues : serpentinisation, hydro-oxydation de la sid erite, radiolyse de l'eau, bio-fermentation, g en eration par effet m ecanique, d egazage de source profonde *via* des failles, corrosion de l'acier. Pour la g en eration d'h elium, les diff erentes sources de min eraux radioactifs et les accumulations interm ediaires sont  egalement examin ees. L'hypoth ese la plus probable est la g en eration de l'hydrog ene par radiolyse de l'eau et de l'h elium par radioactivit e dans ou proche du socle, migrant par des failles profondes, s'accumulant et se concentrant dans un aquif ere avec du m ethane thermog enique puis entra in es avec le m ethane dans le gisement lors du chevauchement du Jura. De nouvelles mesures gaz sur d etecteur portable, partielles mais comprenant l'hydrog ene, sur un des anciens puits for es pr esentant un flux gaz accessible, permet de commenter une  volution du compos e gazeux plus de cent ans apr es la date de d ecouverte en 1906. La diminution de la teneur en hydrog ene depuis la d ecouverte du gisement est probablement due  a l'activit e de bact eries sulfato-r eductrices.

Mots cl es : gisement / gaz / hydrog ene naturel / h elium / Jura / Bugey / Vaux-en-Bugey

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1 Introduction

Hydrogen is an attracting fuel, because it does not produce CO₂ when burning. Currently most of the hydrogen used in the world comes from natural gas (by methane reforming) and its interest to de-carbonize is not obvious, except if CO₂ released during the process is captured and sequestered.

Since a couple of years, hydrogen started to be produced by water electrolyze using renewable electricity (when not used in the grid). This type of production, named Power to Gas, can contribute to global warming mitigation and could give the opportunity to store energy.

Storing hydrogen in existing underground gas storage facilities or dedicated storage facilities is possible for salt cavern storage facilities and currently under discussion for porous reservoirs. Research or pilot projects are launched to understand how hydrogen behaves in the subsurface (Marcogaz, 2016).

Occurrence of anomalous concentration of hydrogen gas in the continental subsurface is rare and quite a recent discovery. For example, first reporting in 1984 for Kansas-USA study case, up to 40% (Angino *et al.*, 1990) or in 1987 for Bourakebouyou well in Mali with a content of 97% (Prinzhofer *et al.*, 2018). The fact that H₂ was not measured for routine analysis in the “western” petroleum industry is a lack to be filled when benchmarking V. Sokolov synthesis written in 1974. Measurements of hydrogen used to be more systematic in CIS and they demonstrate that concentration of hydrogen up to 25% is not so rare in natural gases accumulation. Also, set of data shows hydrogen saturation up to 90% for Japanese volcanic activity (Sokolov, 1974).

Some continental natural hydrogen seeps have been discovered (Larin *et al.*, 2015; Prinzhofer and Deville, 2015; Prinzhofer *et al.*, 2019) but we are far from understanding hydrogen sources, hydrogen migration and hydrogen behavior in the subsurface. (Nivin, 2016; Gregory *et al.*, 2019). The interest for the topic was renewed recently and suffers from poor data sets. A source of natural hydrogen, if available in reasonable conditions, would provide de-carbonized and perhaps renewable energy, if the origin is proved to be partially linked with deep crustal or magmatic reservoir source and then would take a great part in the energy transition.

It is the reason why any case study of natural hydrogen deposit is of high interest.

Vaux-en-Bugey gas condensate field, containing around 5% of hydrogen, is one of these case studies. As an upside, helium content was high in Vaux-en-Bugey. This valuable gas generation is also addressed.

The objective of this work is to test the different hypothesis of generation and migration of gases, focussed on hydrogen and helium, for the specific case of Vaux-en-Bugey field consistently with the specific geological setting. The evolution of hydrogen content in the field is addressed based on recent measurements.

The first experience of gas production in France may provide key understandings for natural hydrogen generation and storage.

2 Vaux-en-Bugey gas field exploration-production history and geological context

The geographical location of the field is presented in Figure 1 and Figure S3 for wells location.

2.1 History of the exploration-production

Vaux-en-Bugey gas field was discovered in 1906 when the Pagniez-Bregi well was drilled. The target of this exploration was to find salt or coal. A gas bearing horizon was found at 190.6 and 216 m below ground level and a blow-out happened. The well was roughly plugged and abandoned.

In 1917–1919, the “Syndicat d’Études et de Recherches du Bugey” drilled Torcieu well for coal, 2 km Nord-Est of the discovery well.

After the first World War, a company (“Société Civile de Recherche de Vaux”) drilled a gas well (SCRV) in 1919 but it didn’t provide a sufficient gas delivery rate. A mining licence (called Buisin from the name of the small river crossing the zone) was granted to SREP and the commercial production started in 1924.

A company (“Société de Recherche et d’Exploration Pétrolière”, *i.e.* SREP) launched a drilling campaign of 5 wells (SREP1 to SREP5). All wells (except SREP1) found gas but the main production well of the field was SREP2. A blow out occurred during drilling releasing around 6 million m³ of gas in the atmosphere.

A gas pipeline was built to transport the gas to the nearby city of Ambérieu where it was used mainly for lighting. This discovery raised great expectations, to supply Lyon and a glass factory built in the neighbourhood. The connection to Lyon was never realized and the supply to the glass factory was temporary and not sufficient. (Charpy, 1990).

A process (based on active coal adsorption) to withdraw gasoline (C5+) from the gas provided around 30l/1000 m³ (Locherer, 1927).

A dedicated station was built to pressurize gas into mobile vessels to provide fuel gas for vehicles (see Fig. 1).

The production was very low after 1950 and was totally stopped after 1961.

2.2 Geological context

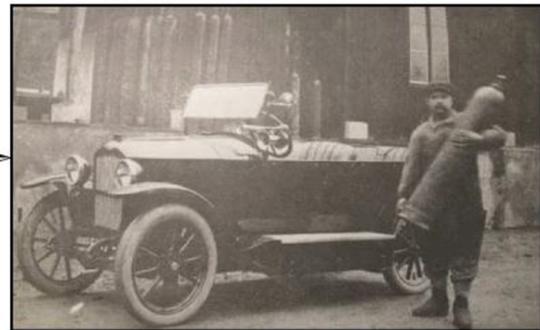
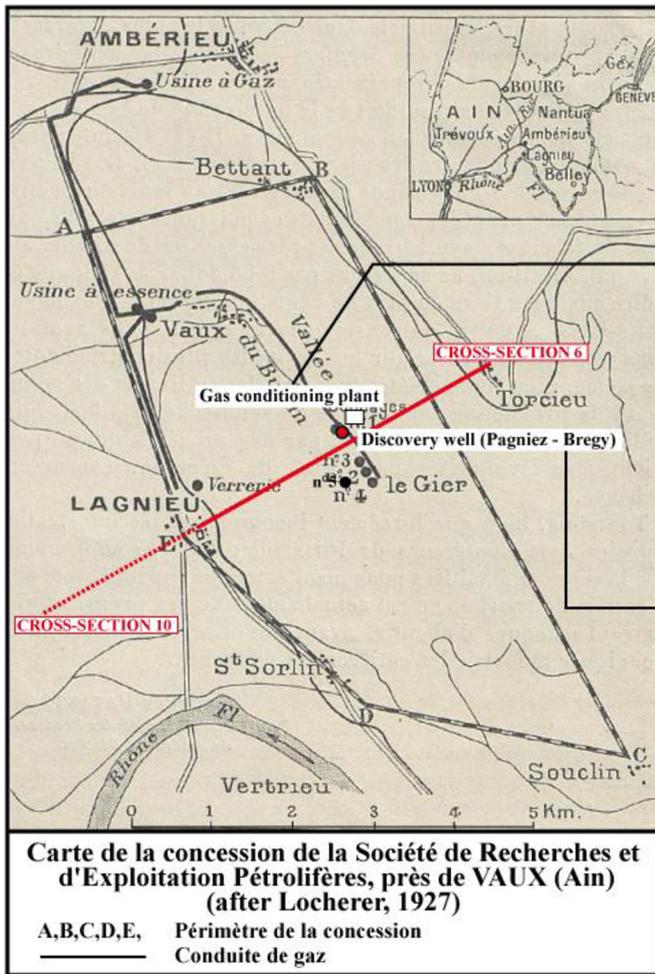
Vaux-en-Bugey gas field is located in the southern part of Jura mountains, outermost margin of western Alps, East of France (see Figs. 2 and 3).

At the end of nappes formation in the Alps (major phase is middle Miocene, around 15 My bp), this banana-shaped mountain was compressed, folded and pushed toward the Northwest. The Mesozoic sediments (above the Triassic evaporitic facies) put under stress were thrust onto the Oligo-Miocene sediments of the Bresse graben (Piffner, 2014). The age of Vaux-en-Bugey thrust is 5 My bp, ante Pliocene to ante Plaisancian, from 5.3 to 3.6 My bp (Vincienne, 1932; Glangeaud, 1953). The gas accumulation is located within the thrust sediments.

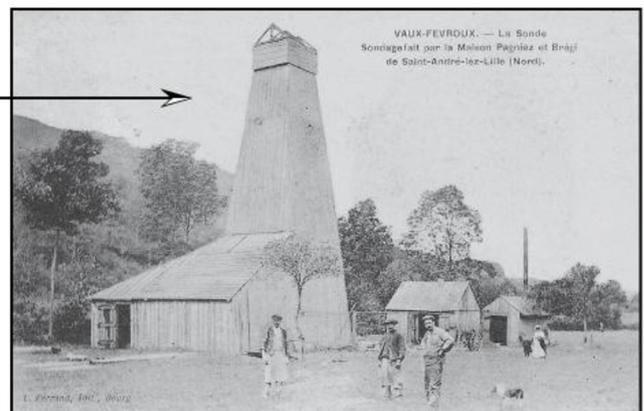
Transverse NW-SE strike sleep faults, as Pont d’Ain-Culoz deep fault (Philippe, 1994) gave the general shape of the chain. Vaux gas field is located along the southernmost NW-SE one, “Rhône fault”. This fault is the border between folded Jura mountains and lower tabular “Ile Crémieu”.

2.2.1 Stratigraphic setting

Synthetic lithostratigraphy for Vaux-en-Bugey and Torcieu area is presented in Figure 4.



Gas conditioning plant. In 1924, the gas contains 5% of H₂.



Discovery well (1904)

Fig. 1. Vaux-en-Bugey gas field location map.

As the field is located within a thrust, lithostratigraphy is uncertain, although 7 wells drilled, because of lacks or series overlaps. No logging tools were available at that times and only two wells get cores (SREP3 and SREP4).

At Vaux-en-Bugey location, the continental crust thickness is around 30 km, so in-between thin one (24 km) for Massif Central-Forez and thick one (up to 58 km) for Alpine domain (Grellet *et al.*, 1993). There is no magmatic evidence recorded in the area. Top of the basement is dipping South-East from the West border of Jura chain toward Alps (Philippe, 1995). The same feature, is known for “Ile Crémieu” basement (Rocher *et al.*, 2004).

The upper part of the basement, outcropping westward in Massif Central (uplifted during Hercynian and Cadomian orogeny) and eastward in Belledonne massif (uplifted during Alpine orogeny) and also South of Ile Crémieu at Chamagnieu, drilled in some few wells is a standard igneo-metamorphic type. The age of the basement is still uncertain: if main part is linked to the Hercynian orogeny (from Devonian to Permian), remains the possibility of neo-Proterozoic pieces, linked to the Cadomian orogeny (Chiron and Kerrien, 1979). Anyway, Vaux-en-Bugey is located 50 km South of internal crystalline nappes and ophiolitic structures of Hercynian orogeny (Grellet *et al.*, 1993).

Paleozoic sediments are present along a SW-NE trend (Debrand-Passard *et al.*, 1984), outcropping South in the exploited Saint-Étienne Basin with Stephanian-Autunian coal measures and bituminous shales but only explored in subsurface within Bas-Dauphiné Basin (Mariton, 1981). These formations get a prolongation North-eastward in Vaux-en-Bugey area, as seen in Torcieu and Chatillon wells. The presence of Permo-Carboniferous is attested in Vaux-en-Bugey SREP4 well, by coring samples. At Torcieu well, the 1187 m drilled section below Triassic got 100 m of Permian shaly sandstones then Carboniferous black schist, psammites and millimetric coal laminations (Fig. S2).

Mesozoic sediments partly outcropping in the area have been detailed in the rich and exhaustive synthesis of the field, by Schoeffler (1941).

Lower Triassic Buntsandstein fluviatile sandstones, the main regional reservoir, is 48 m thick at Torcieu well and absent at Vaux-en-Bugey.

Middle Triassic Muschelkalk is 98 m thick in Torcieu well with marly and dolomitic or calcareous facies.

Upper Triassic Keuper (estimated thickness of 150 m), consists of repetitive sections of iridescent marls, gypsum and dolomitic layers from lacustrine environment.

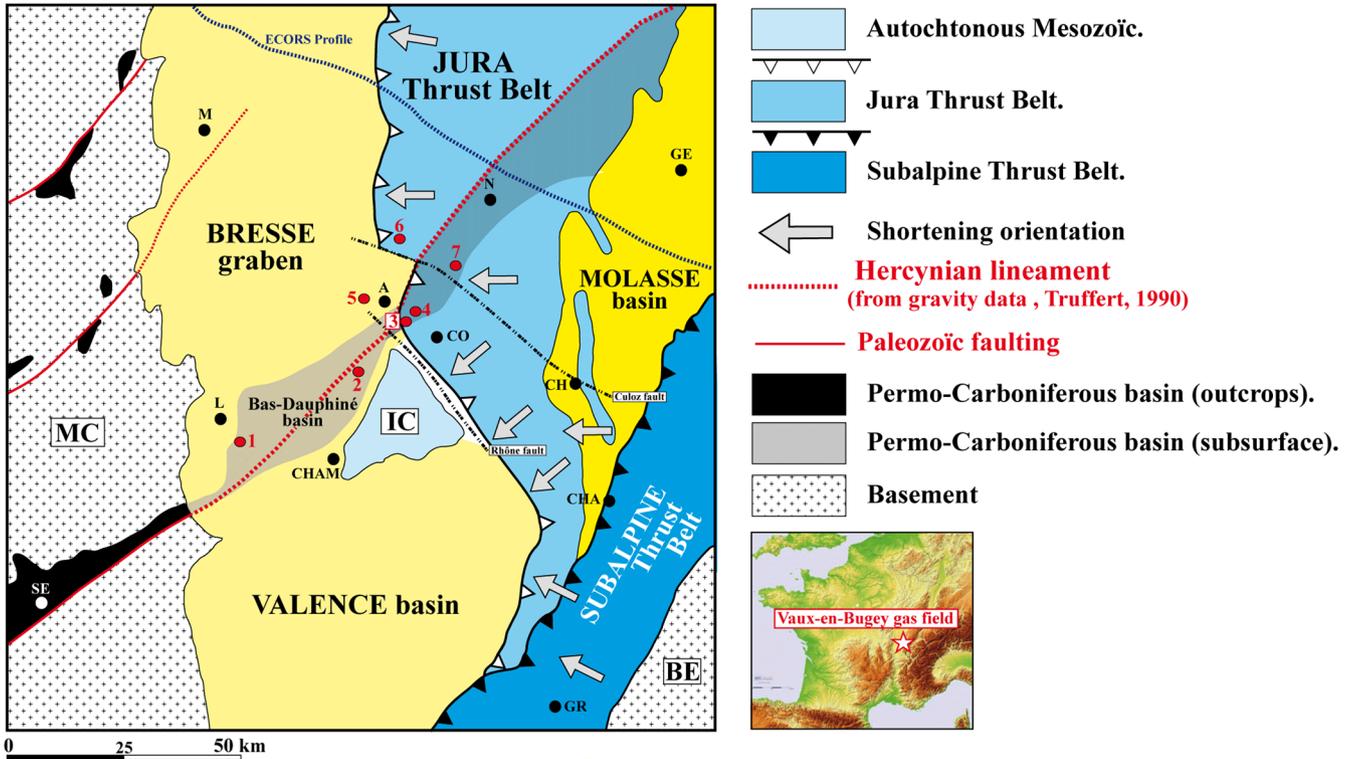


Fig. 2. Regional structural map (from Debrand-Passard *et al.* (1984) and Philippe (1995), modified).

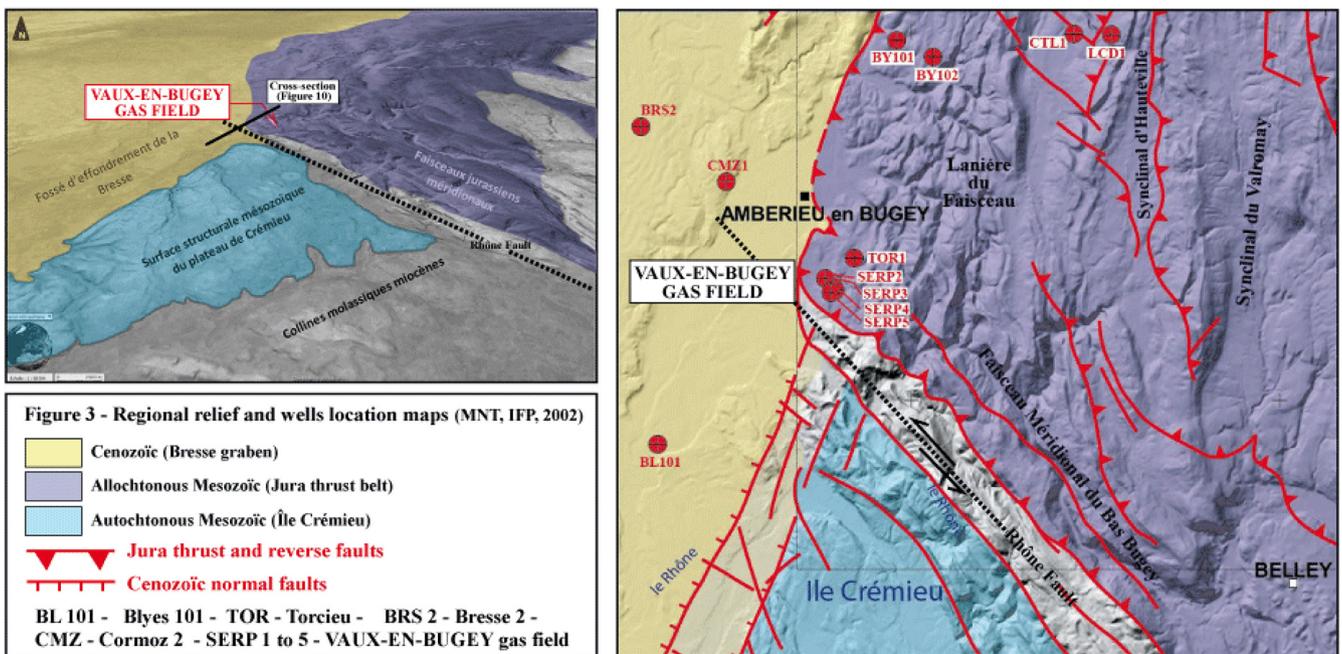


Fig. 3. Regional relief and wells location map (MNT, IFP, 2002).

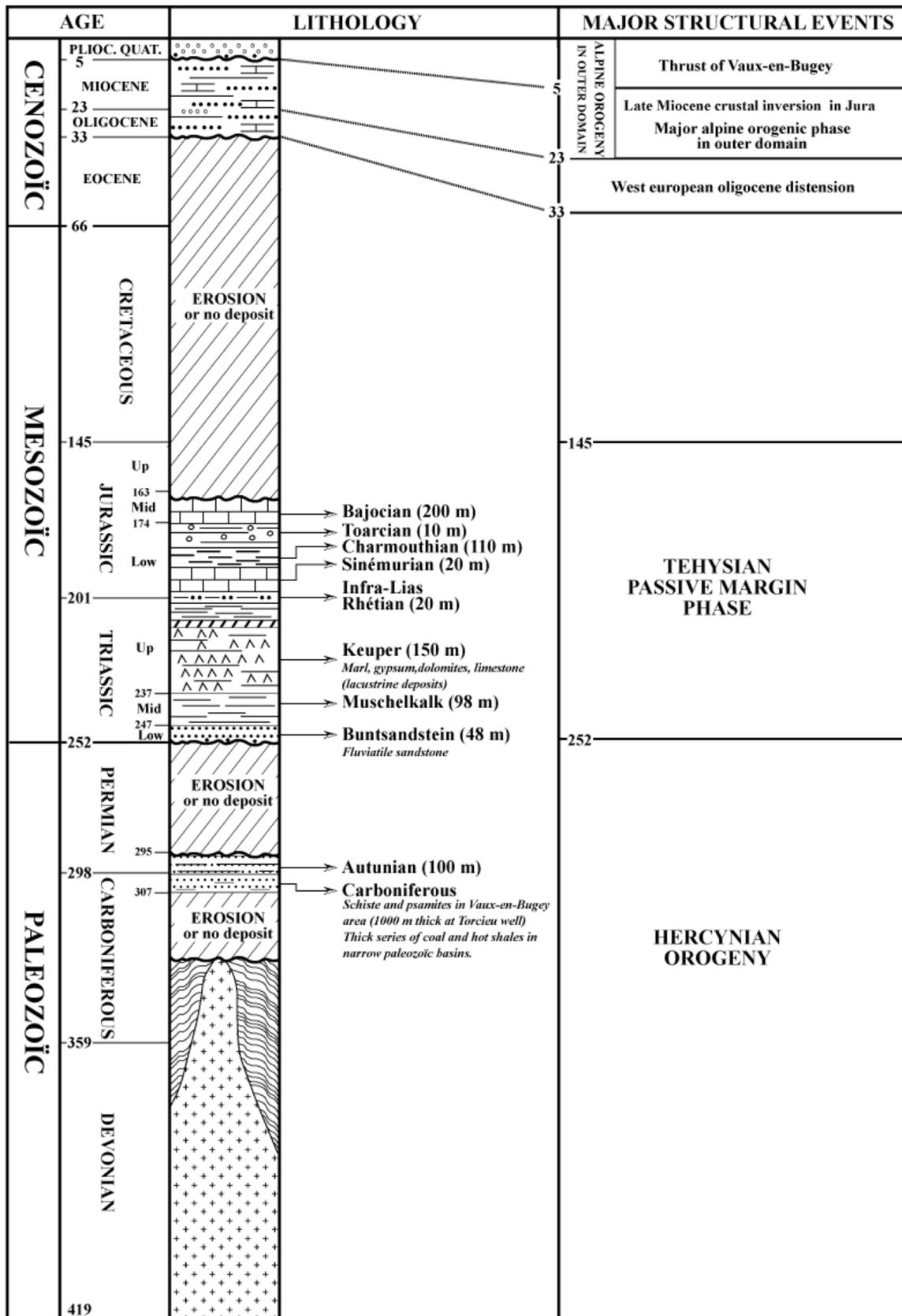


Fig. 4. Synthetic lithostratigraphy for Vaux-en-Bugey and Torcieu area.

Challenging Schoeffler description, the authors suggest that the Keuper dolomitic gas reservoir is an equivalent of Lettenkhole formation and then located at the base of Keuper unit and not 5 m below the top. Because, first, the gas reservoir is always located 100 to 150 m below top Keuper and second because the Lettenkhole formation is regionally observed and not the upper Keuper equivalent “dolomie de Beaumont”. This latter one disappears northward of Vaux as well as salty evaporitic facies (Dromart *et al.*, 1994).

Liassic and Dogger formations (350 m thickness) outcropping partly in the Vaux Valley, develop marine shales and limestones with oolitic ferruginous grey shales (3.5 m) within Toarcian, just below the Bajocian and Bathonian main calcareous reliefs of the site. They have been exploited in Vaux-en-Bugey for iron mining.

Upper Jurassic and Cretaceous formations are eroded in the area.

Discordant Oligocene with green marly sandstones covers the Bajocian limestone in SREP5 well.

Miocene, 200 m drilled at SREP1, is made of molassic sediments including sandstone, marl and limestone.

2.2.2 Trap formation, architecture and gas infilling

Gas trap formation is the result of successive major structural events (Fig. 3):

- Hercynian orogeny: after the Cadomian, the area was deeply impacted and during late Paleozoic, in between mountain ranges was deposited the Stephanian-Autunian coal measures and bituminous shales, which have been affected by late Hercynian pulses with high crustal heat flows (as Saxonian volcanism). Vaux-en-Bugey gas field is located above this Permo-Carboniferous Basin-oriented SW-NE as proved by wells SREP4 (Vaux-en-Bugey), Torcieu and northward Chatillon. According to gravimetric data, an Hercynian lineament follow the Permo-carboniferous basins under the Jura thrust belt (Truffert *et al.*, 1990; Philippe, 1995), see Figure 2;
- Mesozoic passive margin phase with opening of “Liguro-Piemontais” ocean, did not disturb this feature;
- Oligocene extension phase created the deep Bresse Graben with a 2 km uplift of the basement on the East border, front of the Jura, as suggested by Ecors profile (Fig. 5). At a regional scale, there was a possible reactivation of old Hercynian features. As an illustration, the faults bordering the Bresse Graben have the same direction than the faults bordering the Bas-Dauphiné Permo-Carboniferous Basin (Fig. S1 cross-section a);
- Alpine orogeny: this late one got a paroxysmal phase at mid-Miocene, around 15 My bp. As far as Jura is concerned, it is now admitted (Bergerat *et al.*, 1990; Guellec *et al.*, 1990; Truffert *et al.*, 1990; Philippe, 1995; Madritsch *et al.*, 2011; De La Taille, 2015) since the interpretation of the deep seismic Ecors profile, that a basement inversion phase during final Miocene is the origin of the Mesozoic thrusting sedimentary pile occurring post Miocene. This Miocene inversion is illustrated in Figure 5, along the Ecors profile;
- Nowadays, convergence between the Adriatic and European plates is still ongoing and significant seismicity is recorded in the area (De La Taille, 2015). The Ambérieu

(Chautagne) earthquake in 1822 (7.5 MSK intensity), (Fig. 6), or the more recent one in 2006 (3.7 magnitude) at Conand (Bureau Central Sismologique Français, 2006), (Fig. 7), are significative events. Jouanne *et al.* (1995) determine a 4 mm/year horizontal rate for the present-day displacement.

In this context, Vaux-en-Bugey gas field has a very specific structural location and situation.

The field is located at the intersection of two main fault trends affecting the basement (see Figs. 2 and 3). The geothermal gradient, as plotted by BRGM (BRGM, 2008), is high in the zone of junction of these two major structural trends, up to 45 °C/km (Fig. 8).

- the first one is NE-SW direction, bordering Bas-Dauphiné Basin along Ile Crémieu, reactivated during Oligocene extension from old Hercynian features;
- the second main direction of faulting is NW-SE, along Rhône River. It appears to be conjugated to the first one. Seismic line 88 Mex 03 when crossing this direction shows the creation of a narrow trough during Oligo-Miocene (Rocher *et al.*, 2004) (Fig. S1 cross-section b). At present time and possibly since middle Miocene (15 Mybp, main compressive phase in the Alps), a strike-slip faulting is occurring along the Rhône fault in between Jura and “Ile Crémieu” monoclinial unit (Philippe, 1995) (Fig. 3). On the surface, the Villebois fault has a 300 m offset (Kerrien and Monjuvent, 1990).

This location at the intersection of main structural accidents is favourable for deep gas degassing.

The field is also located above a top for lower Triassic fluviatile Buntsandstein reservoir, at a regional scale. This top is exactly located above the old Hercynian SW-NE remaining basin. This geometry is favourable to a good trapping of gases, at least since Oligocene, when the Bresse graben was created and the Jura basement was uplifted.

2.2.2.1 A field in a thrust ramp

The cross-section, presented in Figure 9, N45 orientated, from Torcieu-SREP4 (Vaux-en-Bugey) and SREP5, to Lagnieu city, presents an updated interpretation which try to better illustrate the position and relation of the Mesozoic thrust over the Paleozoic and basement. Previous one is available from Schoeffler (1941) (Fig. 10).

Vaux-en-Bugey field is included within a thrust ramp overlying the old Permo-Carboniferous layers. A mixt of fractured Mesozoic sediments pushed Oligo-Miocene molasse south-westward. This interpretation is consistent with the structural features described above: N130 Rhône fault, which cuts heavily all the Mesozoic series, just South of the field. The scheme is different on the East of Vaux-en-Bugey field (St Sorlin, Villebois), where ramps overlap middle Jurassic series in front of the “Ile Crémieu” (Schoeffler, 1941). To the West, ramps overlap Miocene molasse (Philippe, 1995).

Three different slipping planes are depicted:

- the basal one is dipping SW and so illustrates a possible post Pliocene to present time inversion, as suggested by authors as Glangeaud (1953), Jouanne (1994) or De La Taille (2015). Consistently, Gros Foug relief is currently

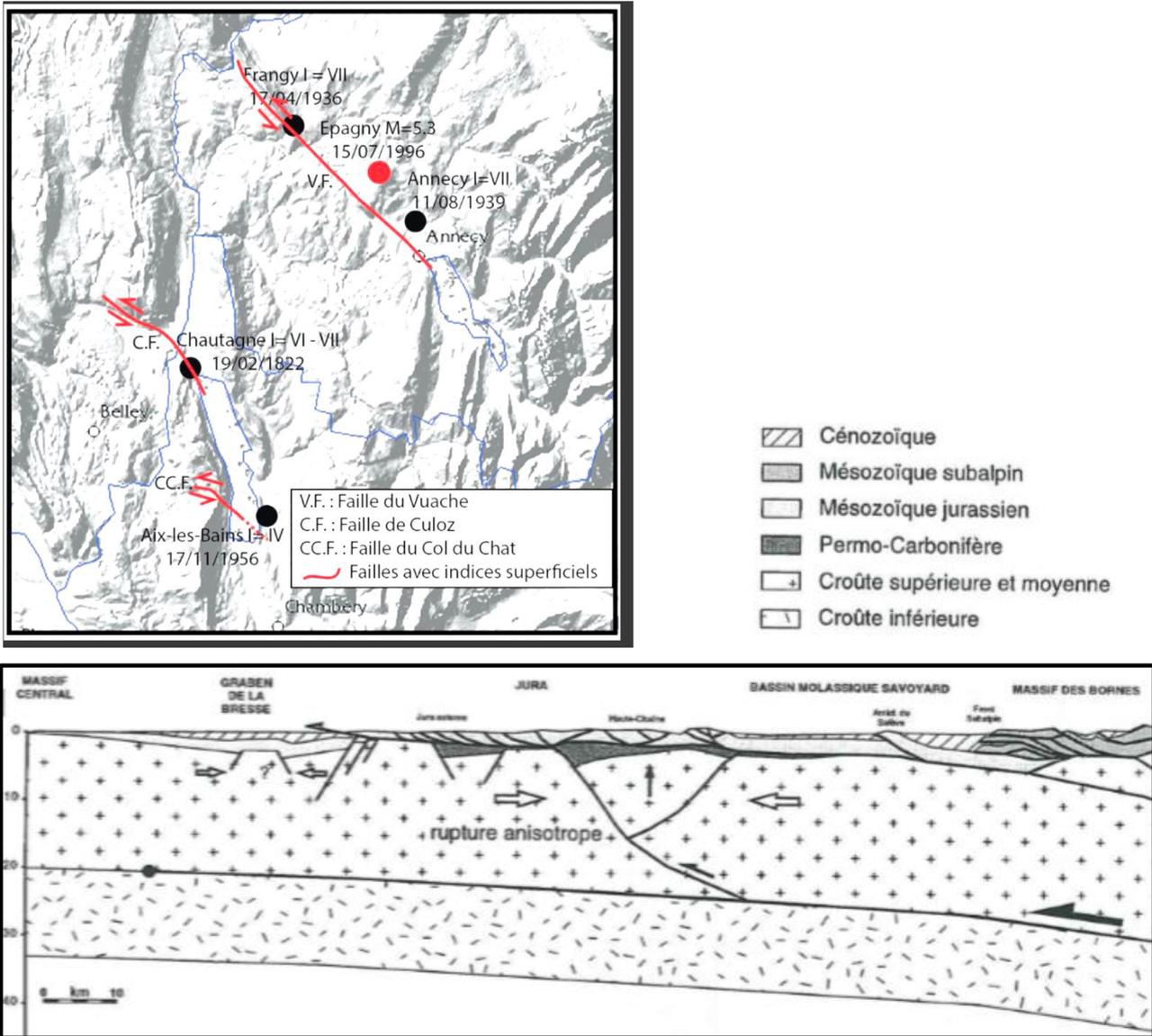


Fig. 5. Recent earthquake data (De La Taille C., 2015). Ecors profile cross-section (Philippe Y., 1995) - see location Figure 2.

uplifted. Jouanne *et al.* (1994), estimate a lateral displacement rate of 4 mm/y for the basement, involving thrust which underlies the Jura. This feature is consistent with Torcieu and SREP4 wells data: the Muschelkalk (98 m thick) and Buntsandstein (48 m thick) drilled in Torcieu are missing in SREP4. So, the moving forward should be still active;

- the decollement located above shaly Muschelkalk, at the base of the evaporitic and lagunal facies of Keuper (Gypsum, shale and dolomite) generates a second slipping plane, base of a unit including Liassic pieces as drilled 80 m along in SREP4 well, 10 m in SREP5 and 0.4 m in Torcieu well with oolothic ferruginous oolithes;
- a third slipping plane, flanking the Liassic pieces, represents the base of a Keuper unit including removed pieces of dolomitic Lettenkhole reservoir near the base. It delineates with second, a liassic decametric thick section, highly perturbed by frictions and able to establish a vertical

and transversal drain to the upward reservoir. This feature is interpreted here as part of a reverse anticline flank, as observed by drilling in S1, 2 and 3 wells (Keuper at surface, then Lias and Dogger at TD) 4 km Southeast of the field (Schoeffler, 1941).

Dogger limestone in front of the thrust (at TD for SREP5 well), above the basal slipping plane, is supposed to be part of the front brechia (fractured zone located under the thrust), rest of the overturned anticline.

The structure shape of the top of the field, based on the first gas occurrence depths on the different wells of the field (see Tab. 1) looks like an anticlinal with a top not far from SREP3 and a possible closure up to 60 m. But the effective closure is 10–15 m for base Sinemurian limestone (“calcaire à Gryphées”) 120 m above the reservoir.

Well SREP1, is located in another lowered compartment and did not find gas.

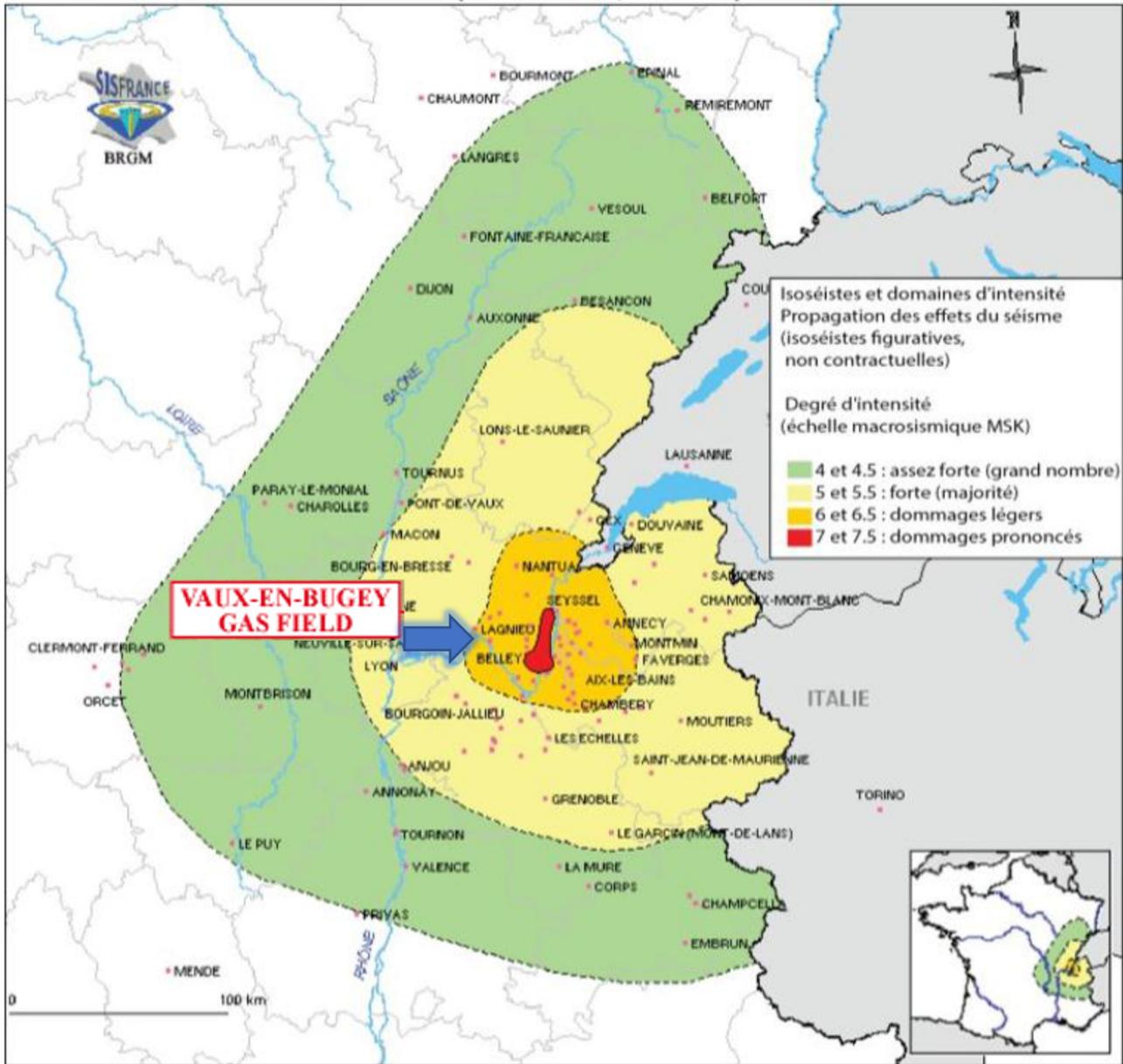


Fig. 6. Chautagne seismic event (1822). MSK intensity VII-VIII, magnitude (5,5-6). BCSF source.

The gas reservoir corresponds to Keuper dolomitic limestone (one or two layers) of few meters and even less. These horizons are interlayered with marls or shales and gypsum. This situation may lead to over-pressured reservoirs. This is the case for SREP2 at 222 m below ground level and perhaps for Pagniez-Bregi well. (see after).

2.2.2.2 Gas infilling

Gases have been probably trapped in a two steps mechanism (Fig. 11).

The initial area of gas trapping (trap 1) should be located along the marge of the Bresse Basin or northward, from Permo-Carboniferous Basin or deep faults. From post Oligocene uplift, the nearby Triassic Buntsandstein reservoir

(48 m thickness at Torcieu well) in preserved faulted tilted blocks, could be charged.

During late Miocene basement inversion or post Pliocene thrusting till present, faults are reactivated and a secondary migration to Keuper dolomitic formation was possible (trap 2).

To define when this last migration occurred, previous or post thrusting, is debatable and will be revisited in chapter 3.2 with pressure data from field production. Nevertheless, the described architecture of the thrust, fractured, with possible migration pathways vertically and laterally, together with oil and gas shows within the base of the decollement (Fig. 9) argue for a recent infilling.

An alternative scenario is to consider that the gas accumulation is the remaining of an older degassing taking place some 10 km Northeast.

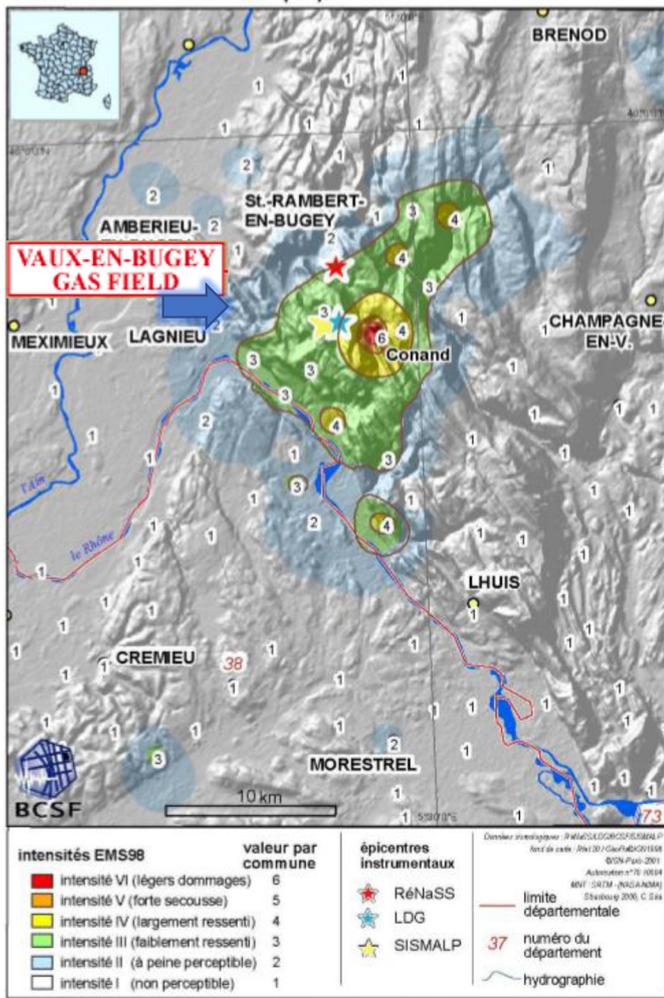


Fig. 7. Conand seismic event (2006). Magnitude (3.7), epicenter depth = 1–2 km. BCSF source.

3 Vaux-en-Bugey gas contain

3.1 Volume produced

SREP3 is at the top of the gas zone but the poor thickness of the reservoir (0–10 m) in this well, could explain the fact that the gas production of this well was very poor.

Even with some uncertainty, authors give information on the depletion of the field (Schoeffler, 1941; Bonte, 1948; Charpy, 1990). The main uncertainties are the volume of gas released during the discovery well (Pagniez-Bregi) blow-out and the production during the last period (1956–1961). Figure 12 presents a reconstructed production history, based on the hypothesis of a gas release of 2.6 million m³ (10 days of blow-out) and a zero production between 1956 and 1961. The blow-out on SREP2 is estimated to 6 million m³ by Bonte (1948).

3.2 Pressure depletion

Pressure values are even more uncertain than volumes. They are measured in kgf/cm² at the well head during

production and the dates corresponding to the different measures are not precise. Some values correspond to well SREP2 and others to well SREP5 or SREP3 (used to measure pressure). Some values correspond to the pressure in the upper layer (SREP5) or the deeper on or an average of the two values.

Considering the low level of pressure, z factor is near 1. It is the reason why the Figure 13 is a P/volume cross plot.

Two values are particularly interesting:

- the pressure on SREP2 before and after blow-out (*i.e.* production of 6 million m³). The gas venue started at 221 m with a well full of water (Schoeffler, 1941) That means that the reservoir pressure was 22 bars or more. We choose this minimum value to report in on the graph;
- the value of 15 kf/cm² is given after stabilization and pressure build-up (Schoeffler, 1941).

This cross plot suggests three important results:

- first, due to all uncertainties, the curve is not a real right but a clear linear trend could be drawn. That means that there is no water driving effect, that no pressure support is provided by an aquifer. It is consistent with the description of the reservoir: dispersed dolomitic beds with vacuoles plugged by marls and gypsum. A measure of pressure on SREP5 in 2018, performed by the authors, gave a value of 1 bar (absolute) at the well head. This very low value, almost 6 decades after the end of production, if representative, would confirm that the reservoir is isolated;
- secondly, to fit with the trend, specially the two measures of pressure before and after SREP2 blow-out, the discovery pressure (which we do not know) should be around 10 bars over the hydrostatic pressure. This is consistent with an isolated and uplifted reservoir and could explain the violence of the two blow-outs. The estimated initial pressure, 10 bars over the hydrostatic, could be explained by a 100 m structural uplift during the thrust transport;
- thirdly, the total volume of gas initially in place is around 22 million m³ (the approximative crossing point between the depletion right and the axis of gas produced) and the recovery factor is very high: around 20.73 (million m³ of gas production)/22 (million m³ of gas initially in place) = 94%.

3.3 Gas quality

Gas quality was measured several times on the well Pagniez Bregi and on production well SREP2 (Bregi, 1909; Schoeffler, 1941). Table 2 presents these data and recent data which will be discussed hereafter.

These analyses show the contents of alkanes (C₁ to C₄: around 90%), nitrogen (N₂: around 5%), of carbon dioxide (CO₂: 0.5 to 5%), hydrogen (H₂: 3.5 to 5%) and noble gases specially helium with a content of 0.096% (Lepape, 1958).

The analysis mentioned by Locherer (1927) is different: it mentions a zero-hydrogen content, which is not consistent with the Pagniez Bregi analysis (1909) and the Schoeffler one (1941).

To try to check H₂ content, a calculation on calorific value was done. A global calorific value of 9500 kCal/m³ is mentioned by Locherer (1927). This value seems globally consistent either to Locherer analysis without C₅+ gasoline or with Schoeffler analysis taking into account C₅+ calorific value. This approach cannot allow to conclude clearly.

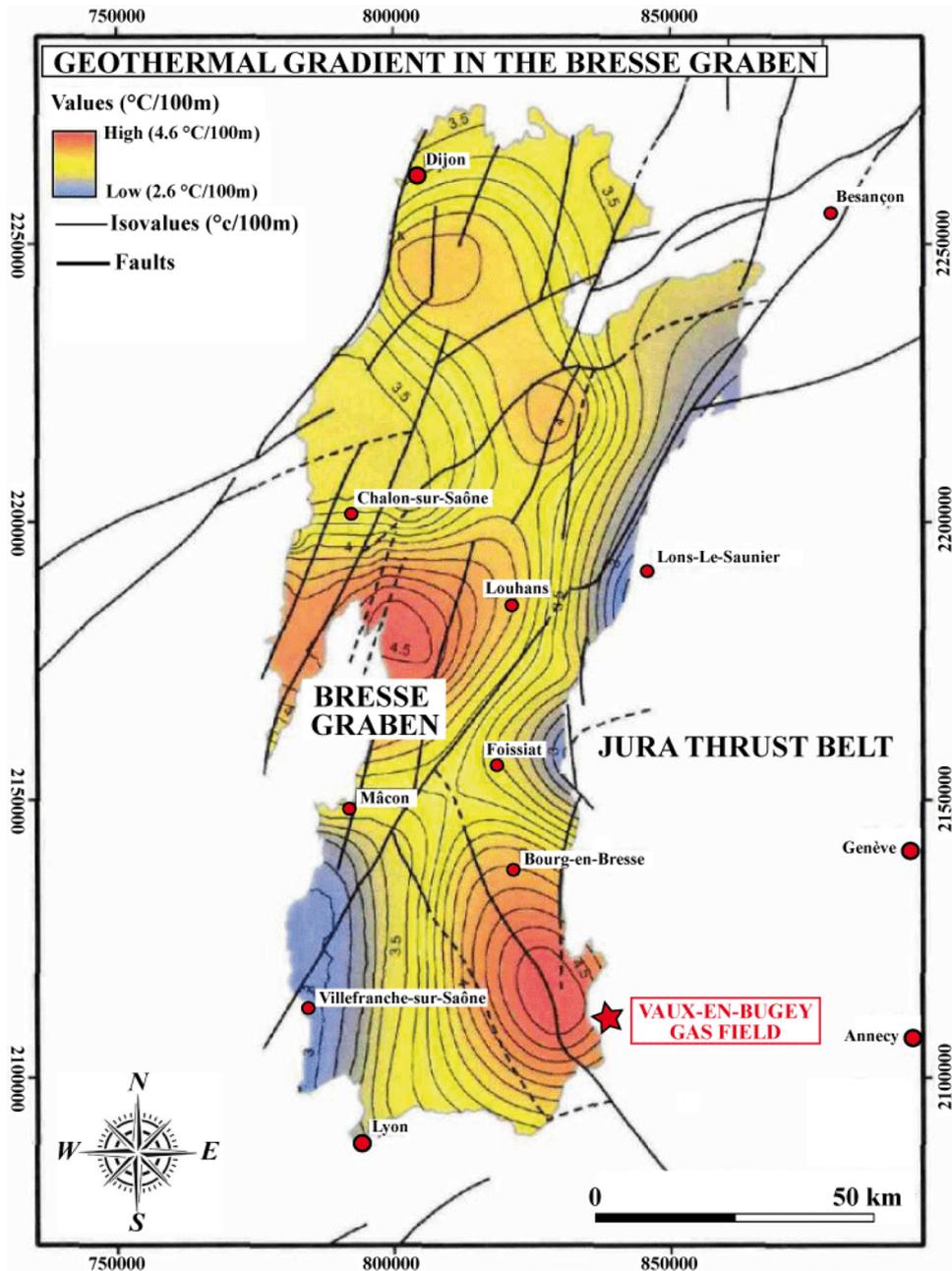


Fig. 8. Geothermal gradient in Bresse Basin (BRGM, 2008).

Nevertheless, the authors do not trust Locherer data a lot, because it is different than the two other ones and because Locherer does not mention the origin of his data.

Based on a total volume of gas initially in place of 22 million m³ and an initial content of around 5% of hydrogen, Vaux-en-Bugey gas field had contained 1.1 million m³ of hydrogen.

No H₂S content values are available during historical production period but it was reported that the gas smelled heavily during production period which suggests a significant content of H₂S (Bonte, 1948). Moreover, sulphur found in the cracks of the reservoir cores was noticed (Schoeffler, 1941). This sulphur probably crystallized from H₂S in contact with oxygen, the oxygen being supplied by mud or air during coring and drilling.

4 Discussion on gases origin

A schematic charge processes is presented in Figure 11 for gases accumulation. The origin of the different gases of Vaux-en-Bugey field needs to be discussed since the different components have not necessarily the same origin. A special focus is done on hydrogen and helium.

4.1 Methane and alkanes

Still exists a debate concerning the origin of natural hydrocarbon gases as methane and light ones till pentane (Etiopie and Schoell, 2014). Most of the time, generation of

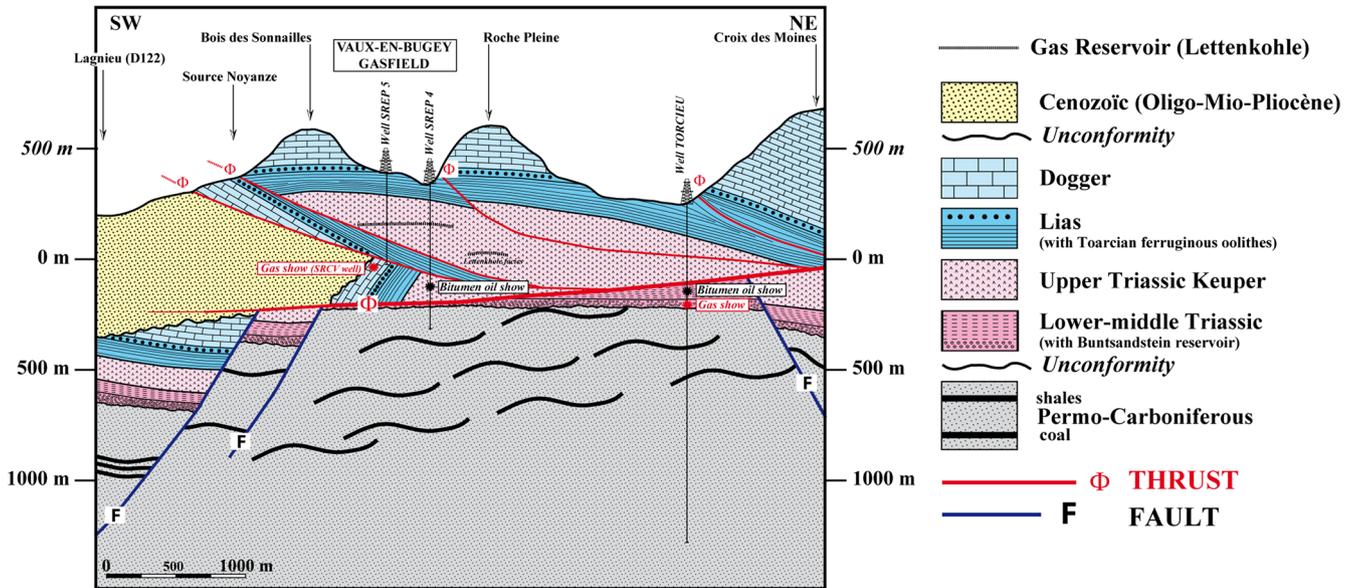


Fig. 9. Schematic interpretative cross-section.

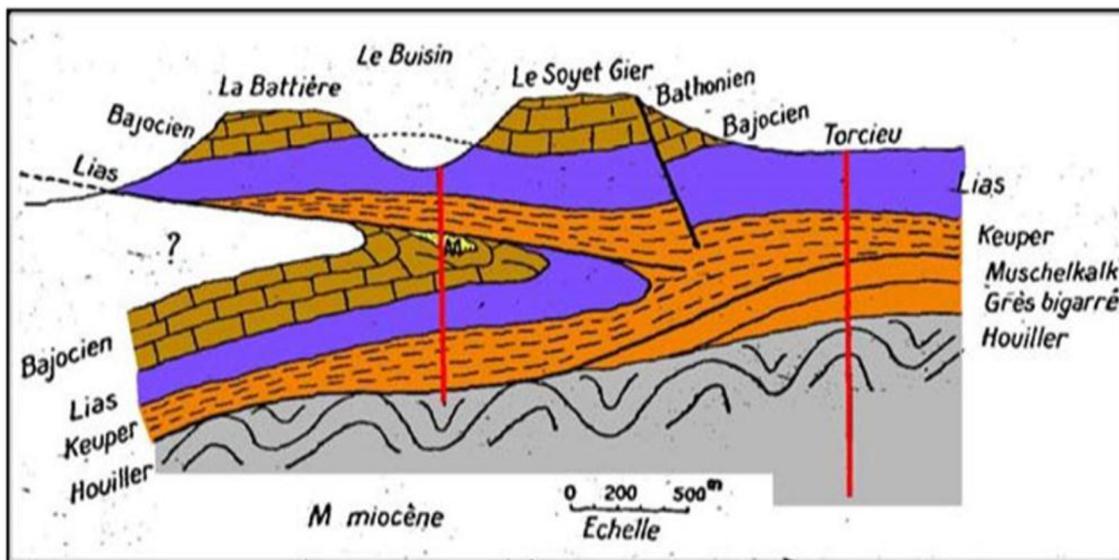


Fig. 10. Schematic interpretative cross-section (Schoeffler, 1941).

such gases could be explained by the microbial or thermal evolution of organic matter concentrated in sedimentary rocks. This process, named biotic seems well adapted to Vaux-en-Bugey gas field.

Carboniferous and Permian sources rocks are present to the South in the Bas-Dauphiné Basin and also to the North. In Charmont well (NE of Vaux) carboniferous coals are 52 m thick and for Chatelblanc well, maturity modelling suggests onset gas generation during early Cretaceous for Autunian source (Pullan and Berry, 2019).

At present time, recorded Tmax (the range is 435–470 in the oil window) in the Bas-Dauphiné Basin and basin modelling for Stephanian coals, over 1000 m deep (Guellec et al., 1990), explain easily a methane generation. The quality of these sources is high with petroleum potential of 20 to

180 kgHC/t for Stephanian coals, 5 to 500 kgHC/t for Stephanian bituminous shales and 5 to 120 kgHC/t for Autunian bituminous shales. In Chassieu well, representative of the basin, total thickness of source rocks is around 30 m (Blanc et al., 1991). Methane probably comes from Stephanian coal more deeply buried and light hydrocarbons from Permian (Autunian) shales. Early generation of oil and methane from Permian-Autunian bituminous shales and from Stephanian coal measures and bituminous shales, started at the end of Cretaceous at least (Blanc et al., 1991). After the Oligocene pre-rift phase, part of the source rocks buried deeper got more mature in the Bresse Basin and also within the Jura chain, uplift of coals during Miocene alpine phase could produce methane by decompression.

It has to be mentioned that the Autunian hot shale formation occurrence is far from Vaux location (60 km). This formation is

Table 1. Vaux-en-Bugey markers table.

Well	Years	Ground level m/sl	Total depth m/gl	Top trias m/sl	Top Buntsandstein m/sl	Top Permo-Carboniferous m/sl	Gas occurrence (m/gl) and production m ³ /d	First gas occurrence m/sl
Pagnez-Bregi	1905–1908	336	221	254.0	NR	NR	–188 and –221 with 960 m ³ /d	148
SREP2	1921	340	223	261.0	NR	NR	–217 with 10 000 m ³ /d and –222 with 110 000 m ³ /d	123
SREP3	1922	339	409	266.0	NR	NR	–179 with 500 m ³ /d	160
SREP4	1923	342	662	266.0	NR	–242	–241 with 1500 m ³ /d and –280 with very weak gas	101
SREP5	1924	390	354	279.0	NR	NR	–270 with 900 m ³ /d	120
SCRV	1919	343	492	280.0	NR	NR	–206 with 116 m ³ /d and 380	137
Torcieu	1917–1919	263	1652	59.0	–184	–232	–458 with weak gas	–205

(Source: from Schoeffler, 1941).

Nota: gl=ground level; sl=sea level; positive depth figures means above the reference; NR=not reached.

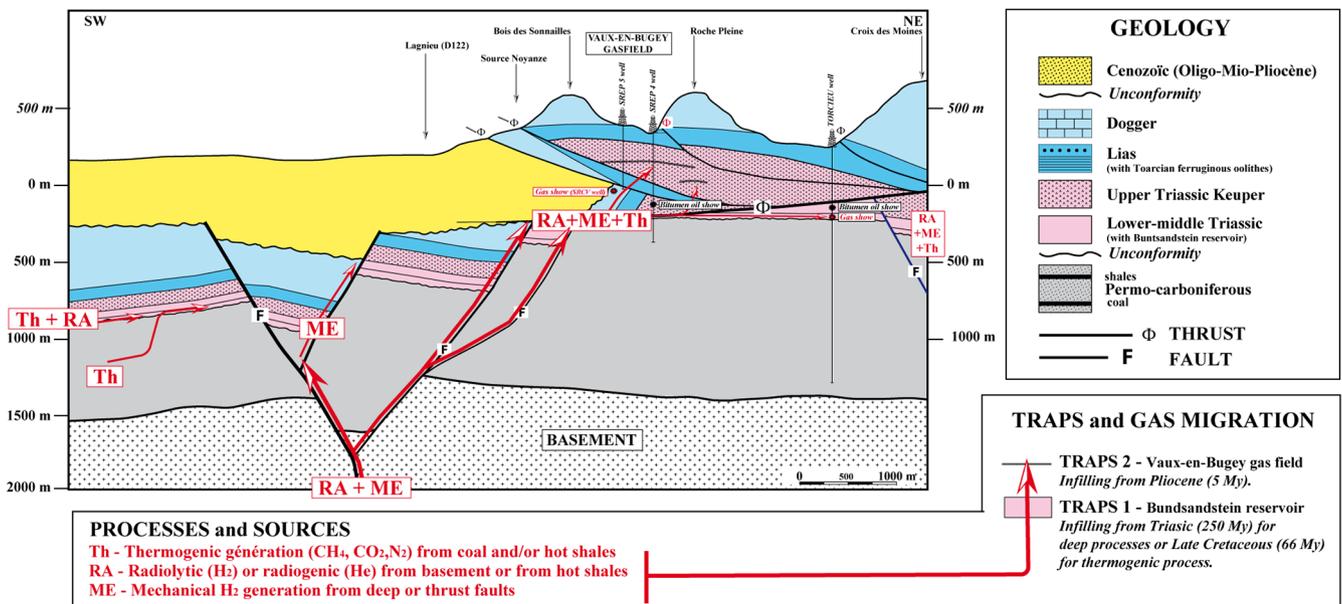


Fig. 11. Schematic charge processes (Vaux-en-Bugey gas field).

dipping toward the North-East, outcropping in Givors, and reach 1000 m deep at St Priest (20 km East to Givors) according to Mariton (1981) and Gudefin (1980). This horizon is unknown northward of the basin, only mentioned in Blyes 101 well (–975 m/sl for top Permo-Carboniferous) and missing in Cormoz well (East of Ambérieu). This Paleozoic formation is 500 m deep in Vaux SREP4 and Torcieu wells but without the hot shales and coal facies as in Torcieu well (1157 m drilled Paleozoic section for Torcieu well. See Tab. 1).

Evidence of thermogenic methane system could be validated for Blyes 101 well, with a gas show (composition not available) during drilling, in Buntsandstein reservoir: this well is located on the South-East border of Bas-Dauphiné Basin (see Figs. 2 and 3).

A biogenic origin for methane is not relevant for Vaux-en-Bugey case study, as gas composition shows upper alkanes concentration up to 8% (refer Tab. 2). Assessed biogenic accumulations get almost pure CH₄.

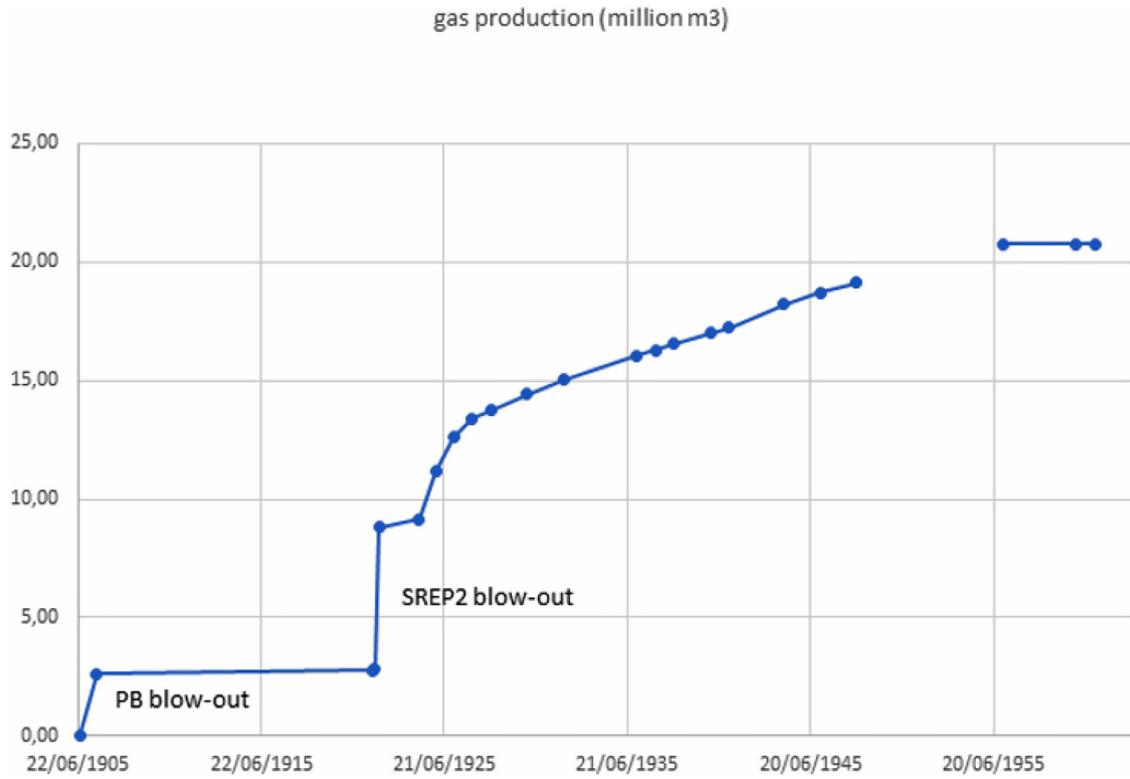


Fig. 12. Production history of Vaux-en-Bugey gas field.

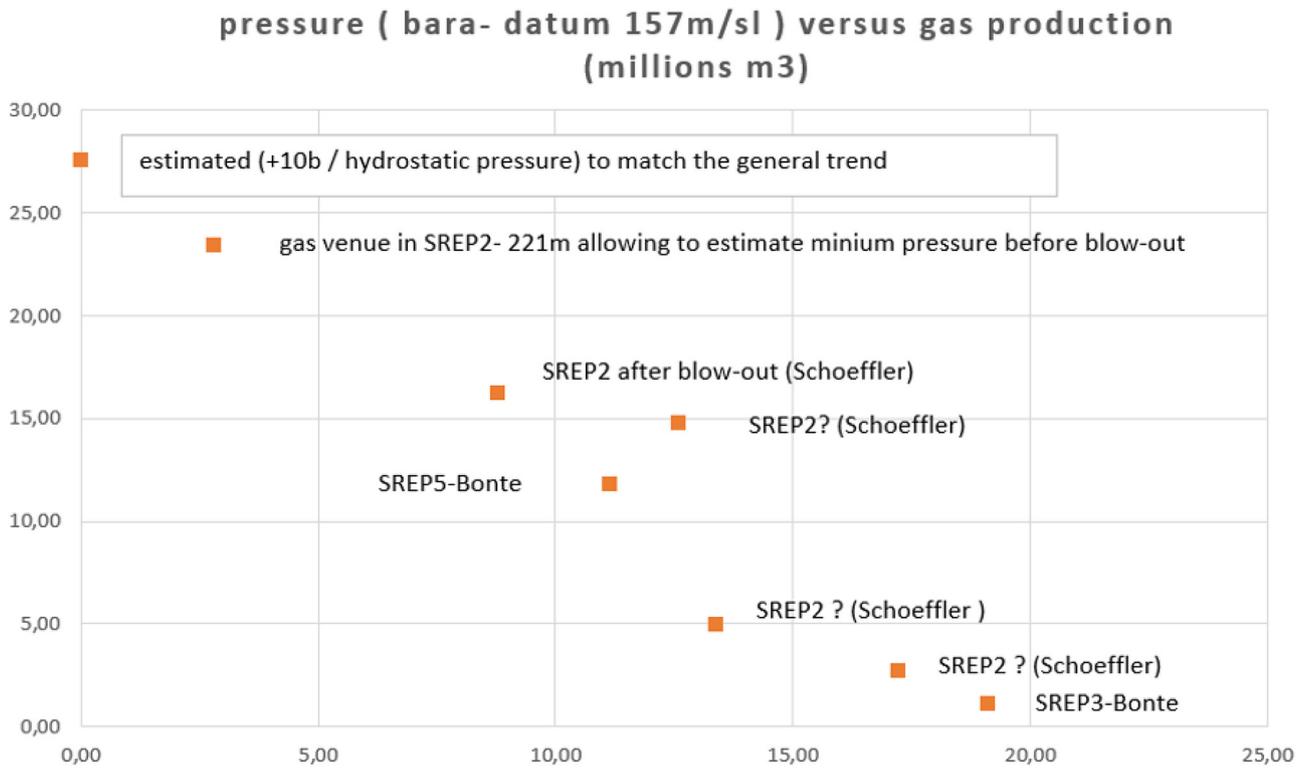


Fig. 13. Pressure versus production cross-plot for Vaux-en-Bugey gas field.

Table 2. Vaux-en-Bugey gas quality data.

Date of the measure	Well	CH ₄ (%)	H ₂ (%)	C ₂ H ₄ (%)	C ₃ H ₆ (%)	C ₄ H ₁₀ (%)	Combustible gases	CO ₂ (%)	N ₂ (%)	Ar, Kr, Xe (%)	He, Ne (%)	H ₂ S (ppm)	Reference of the analysis	Comment
1906	Pagniez-Bregi	90.13	3.69	1.08	0.00	0.00	94.90	0.43	4.66			NA	Bregi (1909)	Measurement corrected from an air pollution, detected by oxygen
1926	SREP2	79.27	5.24	4.91	3.17	0.14	92.73	2.36	4.91			NA	Lebeau mentioned by Schoeffler (1941)	
1933	SREP2	NA	NA	NA	NA	NA	89.28	5.2	5.41	0.016	0.096	NA	M. Lepape mentioned by Schoeffler (1941)	
09-avr-18	SREP5	NA	0.52	NA	NA	NA		4.9	NA	NA	NA	136	Deronzier	
25-juil-18	SREP5	NA	0.47	NA	NA	NA		4.9	NA	NA	NA	123	Deronzier & Giose	

Measurements are in % or ppm of volume.

NA = non available.

It must be mentioned that CH₄ could be produced by bacteria or chemical reaction (Sabatier reaction) from CO₂ and H₂. This mechanism is discussed in 4.2 and assessed as poorly probable during the depletion phase of the field.

As a conclusion: alkanes thermogenesis should be efficient for the field.

4.2 Carbon dioxide

CO₂ in the subsurface could have different origins (Jeandel, 2008).

Some of them are inorganic:

- mantle or magma degassing as in Montmirail (Drôme, France) CO₂ field (Jeandel, 2008; Crossey *et al.*, 2009);
- metamorphism of carbonates;
- dissolution of carbonates.

Some other processes are organic: kerogen maturation, metamorphism of coal, biodegradation of oil and gas.

Dioxide carbon saturation is around 5% in Vaux gas. In the context of Vaux-en-Bugey, CO₂ presence could be explained either by the organic maturation process or by mantellic origin and migration through deep faults.

4.3 Nitrogen

Nitrogen in the subsurface could have different origins:

- atmospheric origin through subsurface water;
- organic material maturation specially coal, nitrogen atoms being provided by organic molecules as amino-acids. This gas is generated at higher temperature than methane (Littke *et al.*, 1995);
- mantle or magma degassing (Ballentine and Lollar, 2002).

The atmospheric origin is not possible for Vaux-en-Bugey because its reservoir is tightly closed with no water flowing. The two others hypothesis are realistic.

4.4 Helium

This so rare element (5.23 ppm in the air – 960 ppm in Vaux-en-Bugey gas), seems particularly well represented in Jura. Not only in Vaux gas field (0.096% volume) but also in the Grozon gas well located Northward (in Lons-le-Saulnier area) with up to 1.34% volume (Ricour, 1956). As a comparison, remind that in the large Panhandle helium field (US-Texas) content varies within the field, from 1.3 to 0.1% (Gage and Driskill, 2003).

As helium is generated from radioactive decay of Uranium, Thorium and Potassium in mineral grains, the candidates as a source are old formations to allow a long radioactive process.

The Permian hot shales (270 My bp), previously cited, drilled in the “Bas Dauphiné” Basin, East of Lyon is a good candidate. Around 60 wells have been drilled in the area: they delineate a SW-NE oriented basin with middle Stephanian coaly formation at the base and just above an Autunian bituminous shale formation. Bed thickness of bituminous shale could reach 10 m and total thickness of the formation is more than 200 m by places.

As helium is solved preferably in gas than in water (Brown, 2010; Byrne *et al.*, 2018), helium is transferred first from solid grains into pore water and then flushed by gaseous methane migration. Autunian formation complies with this scheme: there are beds of white coarse sandstone (“gore”) at the base of the formation and in between bituminous shale beds. Moreover, as old, stagnant water collects more helium than young, hydrodynamic water, the “Bas Dauphiné” Basin, almost completely buried, may be relevant.

The helium may also come from the basement containing radioactive minerals and migrating by faults. This origin is realistic according to Vaux structural situation, described above: limited sedimentary cover (around 1500 m) and deep faults to allow migration.

The helium, if produced by radioactive decay of very old minerals, either of the basement or hot shales, has been first stored in subsurface water then flushed by the natural gas phase.

4.5 Hydrogen

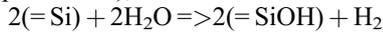
4.5.1 Different mechanisms of H₂ theoretically available

Present theories for H₂ generation in the subsurface, are still in an infancy.

The following processes have been identified or proved *in situ*:

- serpentinization of ophiolite (Fe²⁺ hydro-oxidation of olivine) (Deville *et al.*, 2010; Malvoisin, 2013; Etiope and Schoell, 2014);
- radiolysis of water due to the presence of radioactive minerals (Lin *et al.*, 2005);
- reaction of water with ⁴⁰K (produced by radioactive decay of ⁴⁰K) (Gregory *et al.*, 2019);
- mechano-radical generation or cataclasis due to faulting (Hirose *et al.*, 2011).

Experimental studies support that frictional mechanism liberates H₂ from a range of silicate and non-silicate rocks, associated with water, and even dry basalt (using crystallographic water);



Numerous field studies give a clear association between earthquake activity and high fluxes of hydrogen (also Radon and CO₂). In Japan, Sugisaki *et al.* (1983) records concentrations up to 5000 ppm (0.5%) within fault gouges of Nagoya District. For historic earthquakes, he plots several thousand ppm (several percent volume) for all historically active faults related to the earthquake. Lower values (< 200 ppm) are recorded within faults where the associated earthquake is unknown, classified as “prehistorical active fault”. In China, Wang *et al.* (2018), precise that China began to observe the dissolved hydrogen in the 1970s: A scientific deep drilling exploration project has shown high value of hydrogen concentration several days before earthquake, at a very long distance up to 680 km from the epicentre:

- micro-organism hydrogen production in aerobic and anoxic conditions. For the anoxic conditions the following processes are possible: fermentation of organic material, nitrogen fixation, carbon monoxide oxidation, phosphide oxidation, acetate oxidation (Gregory *et al.*, 2019).

Others mechanisms seems realistic or possible but not clearly observed or demonstrated for subsurface (some hundreds of meters deep):

- degassing of primordial hydrogen (Larin, 1993);
- hydro-oxidation of Fe²⁺ of siderite at moderate temperature Milesi *et al.* (2015) demonstrate that the reaction is possible but at high temperature and high pressure;
- hydro-oxidation of pyrite. This mineral is very stable and no hydro-oxidation (without oxygen) has been reported (Toniazzi, 1998);
- hydro-oxidation of Fe²⁺ in biotite. This reaction is currently studied by J. Murray, B. Fritz *et al.* with real data of the granite of Soulz-sous-Forêts. It is possible but will produce significant quantities of hydrogen only if the hydrogen is regularly withdrawn from the system;
- reaction of H₂S with FeS (pyrrhotite) leading to pyrite formation and H₂ released (Drobner *et al.*, 1990);
- H₂ generation by thermogenesis from organic matter, is not considered, based on the shale gas production. Hydrogen is

quite never recorded. The best value is 0.07% in Barnett shale (Johnson *et al.*, 2015).

Others mechanisms have been observed in industrial contexts but they seem unrealistic *in situ* because of the elements or the level of energy they need:

- manufacturing gas from coal or wood;
- hydro-oxidation of iron metal (Fe) with a high temperature (chemical reaction which lead to discover H₂ gas by Lavoisier);
- generation of H₂ while an iron tool is acting in water, needing high energy (while drilling or grinding rocks) (Bjornstad *et al.*, 1994);
- reaction of acid (H₂SO₄ for instance) on iron powder (the process used to prepare H₂ for airship balloons during early 20th century (Bidault des Chaumes, 1914) or leading to corrosion of steel equipment;
- methane pyrolysis (CH₄ >> C + 2 H₂). This reaction has been recently improved by catalysis and proposed as a process to manufacture hydrogen at 600 °C (Upham *et al.*, 2017);
- pyrolysis of oil shale (like Colorado oil shale) at 510 °C gives better generation rate for hydrogen than for methane (Burnham, 2010);
- more over H₂ may be generated by oxygen corrosion of steel (containing Fe) equipment or similar effects due to cathodic protection (corrosion of the anode or generation of H₂ in case of too high potential applied to the equipment).

4.5.2 Possible mechanisms for Vaux H₂ generation

The origin of the hydrogen of Vaux-en-Bugey field is not clear. Isotopic data should help to confirm some hypothesis but they are not available. Based on the geologic knowledge of one of the most studied area in France, the Jura, several hypotheses are proposed among the list presented above:

- hydro-oxidation of Fe²⁺ of minerals was the first tentative interpretation. No ultrabasic rocks (with concentrations of divalent iron Fe²⁺) which could produce hydrogen in the crust are reported around Vaux location. The basement is a standard igneo-metamorphic type basement. Texas and Mali case studies have Precambrian rocks nearby, not in Vaux-en-Bugey area;
- iron mines have been operated nearby (Mazenot, 1936), in Villebois for instance, and ferruginous oolitic Toarcian facies is confirmed in well Torcieu (20 m above the decollement plane). But the data available on this Liasic ferrous horizon, (Cayeux, 1922) show only Fe³⁺ components as hematite. In other iron mines of the same horizon, Western (Mont Du Lyonnais province) or Southern (Isere province) siderite (FeCO₃) is reported (Cayeux, 1922) as component of the cement between oolites. Thus, it is not totally impossible that siderite does exist in Toarcian ferrous horizon and had been oxidized to provide H₂ to Vaux field but it is not very probable;
- as position and composition of granite within the basement is unknown in the area, the biotite hydro-oxidation process remains possible along deep basement faults but not documented;

- the deep crustal outgassing of H₂ along faults is a possible mechanism as deep faults provide communication between deep crust and Vaux formation. The hydrogen migration from deep crust could have started very early along old Hercynian faults reactivated by Oligocene distension or alpine inversion during late Miocene and possibly ongoing;
- the radiolytic process (due to radioactive decay) is possible. It would probably be linked with the helium formation and migration from deep crust mentioned above. [Lin et al. \(2005\)](#) proposed a cross-plot between He content and H₂ content in subsurface water. A line is drawn for a pure radiolytic generation and different spots representing different geological contexts where Helium and H₂ has been measured.

Calculating a H₂ and He water content from the content in the gas (5 and 0.096%), the pressure of the field (30 b) and the respective Henry constants (5.10^7 and 10^8), the dot corresponding to Vaux formation water (H₂ = 8.10^6 μmol/L and He = $3.2 \cdot 10^5$ μmol/L) takes perfectly place in the high corner of the plot, not far from the radiolytic/radiogenic production trend. This fact is consistent with a radiolytic generation of hydrogen and radiogenic generation of helium from the same source.

- reaction of water with ⁴⁰Ca, should be considered. This effect is probably the origin of hydrogen contained in North Germany gas fields due to Zechstein salt ([Gregory et al., 2019](#)) but is not probable for Vaux-en-Bugey because the Keuper does not contain apparently potassic mineral (as sylvinitite);
- anaerobic fermentative hydrogen production in the reservoir is not very probable: since organic content of the dolomite reservoir is supposed to be poor, a biogenic generation of methane is excluded (refer [Sect. 3.1](#));
- steel corrosion effect cannot explain H₂ measured at the beginning of operation or during discovery of the field (no oxygen in the subsurface, brand new casings and tubings, and no cathodic protection);
- the last mechanism to consider is frictional mechanism. Many details on the quaternary tectonic history are reported by [De La Taille \(2015\)](#) to emphasize the faulting activity in this active thrust belt ([Fig. 5](#)): as the Chautagne (Ambérieu) seismic event (1822), MSK intensity VII–VIII, magnitude (5.5) ([Fig. 6](#)). Also, a very superficial seismicity has been recorded in South Jura, like in Conand earthquake (near Vaux-en-Bugey, 3.7 magnitude but epicentre in-between 1 and 2 km deep) ([Fig. 7](#)), and so affecting thrust sediments. It seems established now that crustal thrusting also take place in the chain: related highest intensities are predicted up to 6.7. For [Jouanne et al. \(1995\)](#), induced movement of rocks in front of the Jura is estimated to be 4 mm/year.

According to [Hirose et al. \(2011\)](#), the hydrogen concentration could reach 1.1 mol/kg of fluid. Considering a surface of 1 km² (approximate size of the field compartment) and a thickness of 1 mm, the number of moles of H₂ that could be generated could be $1.1 \cdot 10^6$ to be compared to 3.10^4 moles of H₂ present in the field. This approximate calculation demonstrates that this mechanism seems able to explain the H₂ generation in Vaux.

According to such a mechanism, a very young source of H₂ is possibly ongoing. To proof this process is still operating at present time is difficult because the field gas quality has been changing during the last decades, probably due to secondary processes as described in the next chapter.

4.6 Hydrogen sulphide

As explained above, hydrogen sulfide was present since the discovery of the field. Now the content measured is 130 ppm. This gas could be produced by different mechanisms but specifically from H₂ as described in 5.2.

5 Discussion on recent gas quality measurements

Well SREP5 is still available for measures and it is an opportunity to compare historical data and recent data, decades after discovery and production. This comparison is useful to understand what happens to hydrogen in the subsurface in the long term, either in natural gas fields or in artificial deposits (underground gas storage reservoirs).

5.1 Recent gas quality measurements

[Table 2](#) presents recent measurements of gas quality (2018) performed by the authors on well SREP5. These measurements have been performed with BIOGAS 5000 (Geotech product, Atex certified for CH₄, O₂, CO₂, H₂S) for all gases except hydrogen and with Portasens II (Analytical Technology product certified) for hydrogen content.

The pressure at the well head SREP5 is the atmospheric pressure (according to an approximative measurement based on hydrostatic level performed by the authors).

To obtain stabilized values of H₂ content (without variation within half an hour), one or two hours are needed.

The hydrogen content of the gas is lower than the initial values ([Tab. 2](#)) and it should be examined if this value is representative of the gas remaining in the reservoir.

The down hole connection between the well and the reservoir is unclear. The well is made of a 9" casing and a 7" tubing equipped with a screen is supposed to be inside the well but the annulus between both tubes is not isolated.

The H₂ content measured may be due to casing-tubing corrosion. This scenario is realistic. The measurements were performed after around one hour of decreasing concentration period and half-an hour of stabilization. The gas volume produced by the well was estimated to be around 30 l/h by different methodologies: size of the flame ([Etiope, 2015](#)-chapter 2) and 2 types of meters giving a maximum (40 l/h) and a minimum rate (27 l/h). Gas flow with such a rate needs several hours to renew entirely the volume of the well which is around 8.8 m³ (354 m of tubing 7"). Such a flowing period was not possible before the measurements. Thus, it is not excluded that even stabilized, the measured H₂ content is different than the concentration of the gas in the reservoir.

The initial measurement is around 0.1% higher than the stabilized measurement. As the flow is laminar in well, we could calculate that in half an hour, the 38 first cm of the tubing are withdrawn. The higher part of the production casing and

the tubing (around cm) are not buried and then submitted to temperature differences and condensation of water and hydrocarbon during very cold periods. This leads probably to a higher internal corrosion in this part of the well and a poor surface status. The excess of hydrogen produced during this stabilization period could be estimated to 0.0075 L. This may correspond to hydrogen due to corrosion, considering one day of this corrosion at a rate of 2.5 $\mu\text{m}/\text{year}$ on 75 cm of 7" tubing. This rate of internal corrosion is lower than some references of total corrosion (internal and external) on wet gas production wells (Patroni, 2007) but seems realistic according to the situation of the well.

The conclusion of these analysis is that the concentration measured may be higher than the representative concentration in the field. This possibility does not change the general conclusion drawn from these new data:

- CO₂ content is similar to the historical values (around 5%);
- H₂ (hydrogen) content has decreased to around 0.5% or even less if the corrosion scenario described above has to be considered.

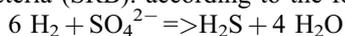
5.2 Discussion on the evolution of hydrogen content

To interpret this change in hydrogen content, several hypotheses could be proposed.

The first point to clarify is the homogeneity of this concentration in the gas field. The recent data are measured on the well SREP5. The value of 5% H₂ correspond to the well SREP2 and the value measured on well Pagniez Bregi was 3.7%. We do not have any information on the measure (0%) given by Locherer (1927). It is the reason why doubting of it seems reasonable.

Gravity segregation of H₂ in the reservoir may exist and explain these differences. Actually, the depths (below sea level) of the swallower gas layer are not varying consistently: +148 m above sea-level for Well Pagniez-Bregi, +123 m above sea-level for SREP2 and +120 m above sea level for SREP5. On the basis of these data, this hypothesis is not confirmed.

Hydrogen could disappear in the subsurface due to bio-chemical reactions (Marcogaz 2016; Gregory *et al.*, 2019). One of them is the sulphate consumption by sulphate-reducing bacteria (SRB): according to the following reaction:



No formation water analysis is available but gypsum horizons are very close to the reservoir and thus it is very probable that the formation water contains SO₄ ions.

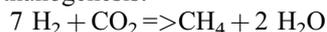
An approximative material balance on SO₄ consumed and H₂S produced confirm the feasibility of the scenario: the water formation volume, even without water influx with a reasonable SO₄ concentration (some mg/l) could be able to supply the SO₄ needed to consume the main part of the hydrogen of the field. It is possible that the major part of H₂S created had been withdrawn during production, and the remaining was dissolved in formation water but it is difficult to perform a mass-balance calculation because the initial H₂S content in the gas is unknown.

The question is to clarify when the reaction started: after gas migration to trap 2 (the Buntsandstein formation water

does not contain sulphate), during gas depletion, after gas depletion?

Some H₂S was probably in the gas before the discovery of the field (see above-sulfur crystallization in reservoir cores) but the main H₂S production reaction may have start since the discovery and is contributing to H₂ decrease. The drilling process may have provided SRB or feed SRB with nutriments (Gregory *et al.*, 2019).

Another possible reaction able to consume hydrogen, is the methanogenesis:



This reaction is possible at high temperature. This reaction is suspected to be possible at low temperature due to bacteria. (Buzek *et al.*, 1994; Panfilov, 2010; Gregory *et al.*, 2019; Ranchou-Peyruse *et al.*, in press). If this reaction had happened in Vaux-en-Bugey reservoir, the CO₂ content would have decreased. Table 2 values does not demonstrate clear decrease of CO₂ content. But CO₂ content in gas is directly linked to another geo-chemical equilibrium with carbonate and may change independently of methanogenesis. In conclusion, the methanogenesis reaction is not demonstrated but not totally excluded.

8 Conclusion

The case study of Vaux-en-Bugey field would contribute to a better understanding of natural hydrogen origin and geo-chemical mechanisms when stored in the underground.

Even in a well-known geological context as Jura is, the origin of hydrogen in the gas remains uncertain. Vaux-en-Bugey appears to be a case of hydrogen accumulation in continental crust which is not linked with a nearby ophiolite serpentinization mechanism which is the main described one.

Several hypotheses have been considered. According to the authors the more probable mechanism is water radiolysis for hydrogen and radiogenic origin for helium, from deep basement source (or possibly nearby Autunian hot shales) migrating along old and still active Hercynian faults. The other probable mechanism is mechano-radical hydrogen generation, cataclasis, due to friction along shallow or deep faults.

Timing of the gas reservoir infilling could not be solved properly. Nevertheless, the authors prevail a better probability for gas charge happened during thrusting, which allowed migration from a former gas accumulation in Buntsandstein.

Isotopic analysis of hydrogen in CH₄, H₂ and of associated He would give useful data.

Recent analysis of the gas strongly suggest that the hydrogen content has decreased since the discovery of the field, a century ago. The corresponding mechanism is probably the consumption by SRB (sulphato-reducing-bacteria). Water analysis and geochemical reservoir modelling would confirm this hypothesis.

Supplementary Material

Figure S1. Seismic profiles (Rocher *et al.*, 2004).

Figure S2. Torcieu well field log, 1917–1919 (St Gobain archives).

Figure S3. Vaux-en-Bugey wells location map, 1924 (ENS Lyon archives).

The Supplementary Material is available at <http://www.bsgf.fr/10.1051/bsgf/2020005/olm>.

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They thank Saint-Gobain Archives to provide original unpublished data (Torcieu well field log. Fig. S2) and G. Dromart for providing historical documents on the field (Fig. S3).

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