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Mapping of natural CO_2 emissions before NECCS

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Abstract

The Fonts-Bouillants sector is a natural CO_2 emission site which belongs to the French carbonic province in the Nièvre department in the north of the Massif Central. In the context of the control of greenhouse gas emissions, this study aims to characterize the natural gas emissions on a site of industrial interest. The objective here is to produce helium while reducing the CO_2 impact. This site was therefore chosen to demonstrate the feasibility of the NECCS (Natural Emission of Carbon dioxide with Capture and Sequestration) technology aimed at capturing natural CO_2 emissions of geological origin. Various analytical procedures and techniques have been deployed on the Fonts-Bouillants site to monitor during field campaigns the concentrations and flows of gases from -10 m to the atmosphere. Raman and FT-IR measurements are carried out to follow the concentration of the gaseous phase at equilibrium with the aquifer at -10 m. Measurements at -1 m, on the surface of the ground, at +1m are carried out during seasonal measurement campaigns. Finally, maps of atmospheric gases are produced by infrared emission remote sensing. The results of a first measurement campaign showed that CO_2 was associated with nitrogen and methane in the water of the aquifer with relative proportions varying over time. Gas concentrations at -1m vary from 400 to 14,000 ppm for CO_2 and from 0 to 30 ppm for CH_4. At ground level, CO_2 concentrations vary from 500 to 10,000 ppm. They decrease at +1m to be distributed between 500 and 1000 ppm. Images obtained by infrared remote sensing do not reveal any plumes of CO_2 or methane above the site. These first results show the higher concentrations and flows of CO_2 in the northern part of the sector studied on either side of the Saint-Parize-le-Châtel fault. The most intense CO_2 flux is located in the North-East quarter and it makes it possible to estimate the mass of CO_2 emitted annually on a scale of 400 m^2 to 1700 tons. These results should be confirmed during the next campaigns of measurements. The Fonts-Bouillants site is proving to be a good environment for developing technologies aimed at deploying NECCS technology as well as the co-recovery of CO_2 associated gases.

Keywords: CO_2 mapping, natural CO_2 emissions, borehole, soil, atmosphere, NECCS

1. Introduction

European Climate Law, establishing the aim of reaching net zero greenhouse gas emissions (GHG) in the EU by 2050, sets an intermediate target of reducing GHG by at least 55% by 2030 compared to 1990 levels. Achieving this goal requires the implementation of conventional abatement technologies such as energy efficiency, the abandonment when possible of fossil fuel in transport, home heating and industry, and the deployment of CCS when the fossil cannot

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be abandoned. Achieving carbon neutrality requires offsetting residual emissions by increasing the share of natural carbon sinks and promoting so-called "negative" emissions technologies. Fuss et al. [1] lists 7 compensation technologies including afforestation and reforestation, Bio-Energy with Carbon Capture and Storage (BECCS), Direct Air Carbon Capture and Storage (DACCS), Biochar management, the enhanced weathering, ocean fertilization and carbon sequestration in soils. The study presented here completes the range of Negative Emission Providers by the NECCS (Natural Emission of Carbon dioxide with Capture and Sequestration). NECCS can be considered as an efficient Carbon Dioxide Removal (CDR) from the atmosphere because natural CO_2 emissions from the Earth mix with biological CO_2 to produce the concentration of pristine CO_2 of anthropogenic contribution. Prior to CCS, it is necessary to characterize and quantify the natural gas flux in order to calibrate the capture, transportation and storage units. The objective of the paper is to space-time CO_2 emissions mapping in the Saint-Parize-le-Châtel area in the Nièvre Department, France.

2. Site description

2.1. From the industrial point of view

45-8 ENERGY, France based company, is dedicated to the exploration and production of strategic industrial gases which are essential to the energy transition and the New Tech: helium and natural hydrogen. It focusses on short supply chains enabling human size local projects targeting a nearby consumption. The emergence of this sector is made possible thanks to a pioneering innovative geological approach supported by strong technological innovations. In collaboration with academic and industrial partners, 45-8 ENERGY conducts several Research and Development projects aimed at removing the technological barriers encountered, developing more eco-friendly technologies and strengthen its leadership. 45-8 ENERGY received its first Helium Exclusive Research License, named “Fonts-Bouillants”, on an area of 251 km^2 in the “Nièvre” Department, France. The project aims to capture at very low depths and product a gas rich in helium and carbon dioxide that naturally escapes to the atmosphere thanks to four thermomineral sources and a major geological fault in the sector. The CO_2 co-produced in the event of helium exploitation will be upgraded and / or mineralized. This pioneering co-valuation approach coupled with a disruptive supply chain that considerably reduces the carbon impact of the sector, makes a significant contribution to the ecological transition, awarded with the “GreenTech Innovation” label of the French Ministry of Ecological Transition. Multiple low-cost geophysical acquisitions (minimum footprint and easy-to-deploy) have been scheduled to have a precise vision of surface and subsurface geology. Reopening and testing ancient well over a long enough period have been made to assess flow rates, gas composition and reservoir behavior. Several wells were drilled during two exploration campaigns in 2021 and 2022, confirming the potential of the area for a future production.

2.2. Geology

The major natural sources of CO_2 in mainland France are located in the CO_2 hydrothermal provinces west of the Rhone valley (from Limagne to Languedoc). Other natural sources of CO_2 are known in the Alps, the Pyrenees, the southwest of the Massif Central, and in the Alsace region. During oil prospecting in the Drôme department, a 98% CO_2 deposit was discovered and then exploited by Air Liquide in Montmiral. Vents or rising gas are also detected in the North-East of the Massif Central near the coal basins. The Fonts Bouillants permit (PER) held by 45-8 Energy in Nièvre Department belongs to the latter environment. The PER is located at the southern edge of the Paris Basin, bounding the north of the Massif Central / northern edge of the Limagne and westward of the Morvan massif (extension of the Massif Central) (Fig. 1). It is composed by the most external sedimentary formations of the Basin from Triassic to Early (Lias) and Middle (Dogger) Jurassic. The basement is composed of Pre-Mesozoic Permian-Carboniferous Basins and granites and metamorphic terrains. In the valleys outcrop the Bourbonnais formation and the deposits of river terraces. The southern edge of the Paris Basin is a complex area where multiple lineaments inherited from the Variscan (Hercynian) orogeny meet. The perimeter of the Fonts-Bouillants PER nevertheless remains a relatively stable area. The main geological structures in the Fonts-Bouillants PER such as the Saint-Parize fault (Fig. 2) or the Neuville-lès-Decize horst are secondary structures, of small extent.

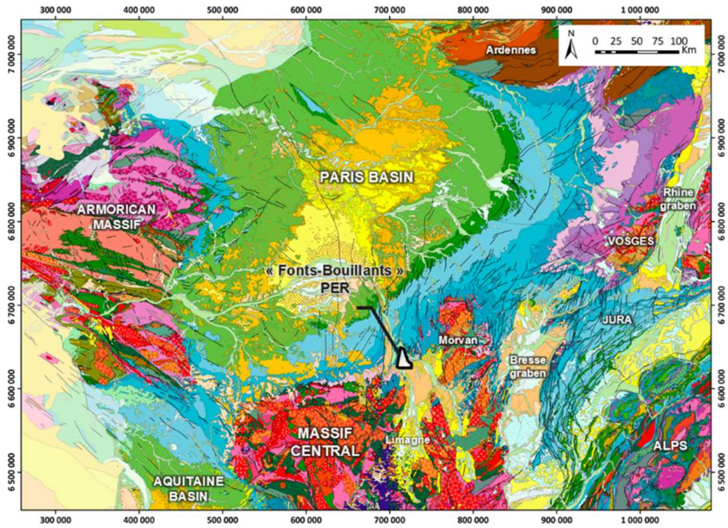

Fig. 1: Geological map of France at 1/1M (BRGM) with location of “Fonts-Bouillants” PER

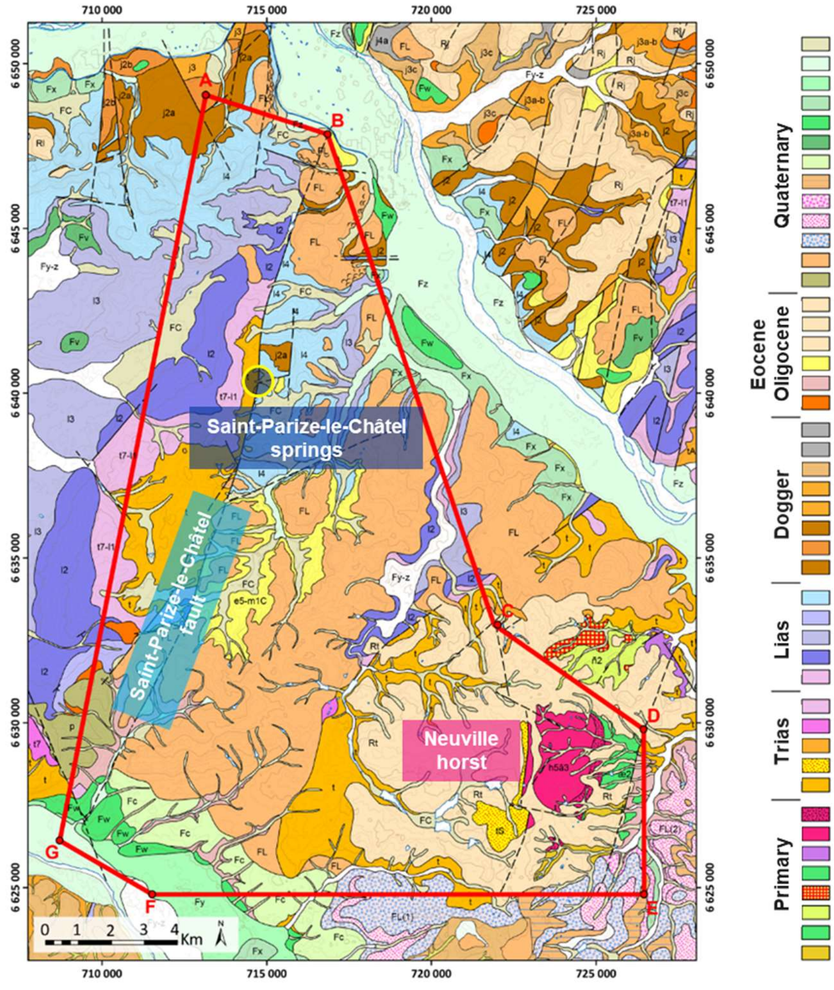

Fig. 2: Geological map of “Fonts-Bouillants” PER (from Geological map of France at 1/50 000 by BRGM)

The gas composition of the Fonts-Bouillants springs is dominated by CO_2 (93%), N_2 (6%) and CH_4 (0.2%). He content varies from 300 to 3000 ppm. In the Massif Central region, isotopic analyses of carbon from CO_2 carried out on all the thermo-mineral sources indicate a mantle origin [2]. The chemical analysis of spring waters gives a pH of around 5.8, and show the presence of bicarbonates (1200 mg/L), sulphates (1200 mg/L), calcium (800 mg/L), magnesium (80 mg/L) and chlorides (50 mg/L).

2.3. Context

The region has an important agricultural activity marked by livestock. The landscape is composed of a juxtaposition of pastures, cultivated fields and forests. The area of interest for this study is a mixture of pastures, groves, vegetable gardens and habitation over an area of approximately 40,000 m^2 over the Saint-Parize-le-Châtel fault (Fig. 3). The climate of the Fonts-Bouillants region is of the degraded oceanic type with a notable continental influence from the Loire and Allier plains. This results in cold winter nights and hot summer days with average temperatures ranging from 0 to 25 °C and a constant monthly rainfall of 60 mm. The year 2022 was marked by an intense drought with monthly precipitation well below normal (14 mm on average over the first 7 months) and maximum temperatures exceeding 40 °C. Some dry CO_2 emanation sites (mofettes) are observed all along the Saint-Parize-le-Châtel falt. Several mineral springs are located in the village of Saint-Parize-le-Châtel, and the most important is the Fonts-Bouillants spring, so called because of the large quantity of gas that escapes and gives it the appearance of a boiling liquid. It was once served at the Élysée Palace on the table of Félix Faure, then President of the Republic, in the ministries, prefectures and restaurants of Paris where it was highly appreciated. An unsuccessful attempt to reactivate the production of carbonated water, which had been stopped in 1975, took place and 5 shallow boreholes were drilled to find an aquifer with a higher commercial flow than historical sources. Most of the wells have been abandoned except for the SP2 well equipped with a strainer and kept for domestic water production. The SP-2 location is marked on Fig. 3 and Fig. 5b.

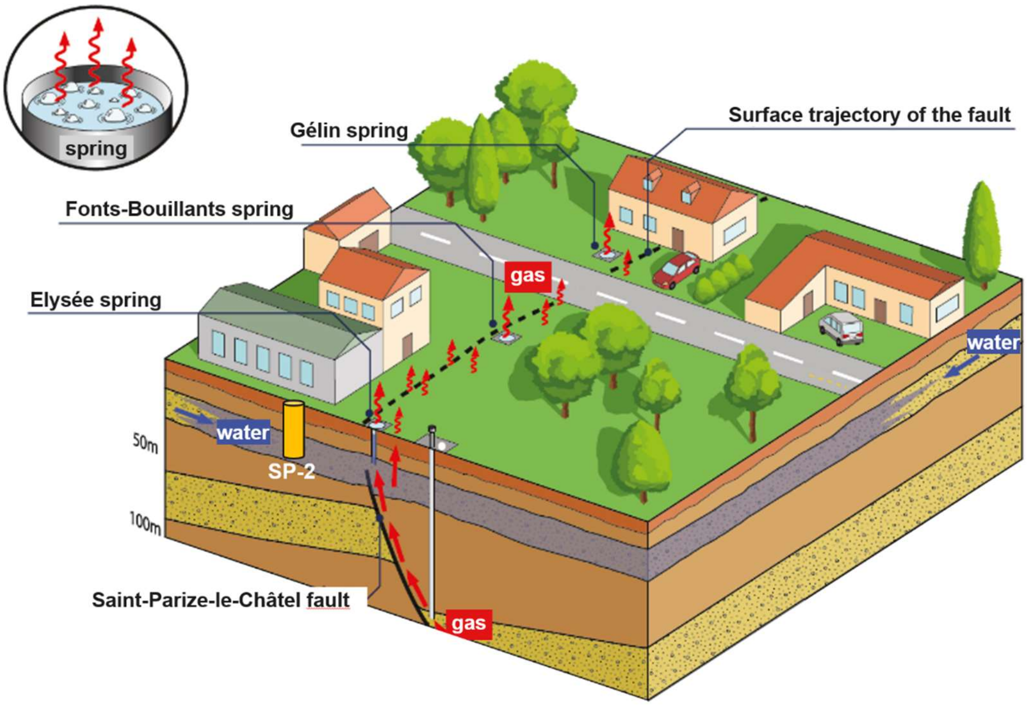

Fig. 3: Block diagram of the Fonts-Bouillants site where the study was carried out.

3. Experiments

The objective of the space-time CO_2 emissions mapping project in the Nièvre department is to quantify natural CO_2 emissions over a limited scope of the permit. This quantification leads to a mapping of concentrations and fluxes at the soil-atmosphere interface as it was done in previous studies [3, 4]. A well in a shallow aquifer (around 10 m) allows the variability of dissolved CO_2 and accompanying gas concentrations to be monitored remotely over a little less than a seasonal cycle. Finally, the atmospheric compartment is the subject of remote sensing measurement campaigns using 3-D mapping of the gas plume over the selected zones of kilometric extent. The approach is based on three stages.

3.1. Gas concentration measurements in borehole

Continuous measurement in a shallow submerged dedicated well SP-2, 25 m deep without evidence of gas bubbling at the West edge of the Saint-Parize-le-Châtel fault. Deployment of the ®SYSMOG system and coupling of FT-IR and fiber Raman measurements acquired every hour [5, 6, 7]. Regular gas sampling for quantitative laboratory analyses of trace gases and carbon isotopes are planned but not presented in this paper. The Raman measurements of the gases flowing within the gas circulation lines are performed using a Raman RXN2 spectrometer equipped with a 532 nm Nd:YAG excitation laser at 100 mW of power, and coupled with a gas probe (AirHead™, Kaiser Optical Systems, Inc) via a 10-meter-long optical fiber [5]. The air probe is embedded by a dedicatedly designed stainless steel gas cell equipped with a sapphire window and an array of mirrors to amplify the Raman scattering intensity that is connected to the gas circulation system. The peak area of CO_2, N_2, CH_4 and H_2O are measured by fitting Raman spectrum with Gaussian peaks after baseline subtraction using Labspec6 software (Horiba). The spectral data are transformed in molar proportions (mole %) from homemade calibrations and literature cross sections [8]. The infrared spectra were recorded using a portable infrared spectrometer ALPHA (Bruker Optics, Germany). A gas cell with a 5 cm path length equipped on both sides with zinc selenide (ZnSe) windows was placed in the sampling module and connected to the gas circulation module for the continuous analysis of borehole gases. The infrared spectra were acquired with 16 scans in the mid-infrared range (5000-600 cm^{-1}) with a spectral resolution of 1 cm^{-1}.

3.2. Gas mapping in subsoil environments

Interpolated maps of soil gas concentrations are acquired from CO_2 and CH_4 measurements at a depth of -1m and from CO_2 measurements on the soil / atmosphere interface and at +1m above the soil during 4 campaigns covering a seasonal cycle. In parallel CO_2 flows are recorded with a CO_2-flux chamber. The approach used to monitor gas concentrations at 1m depth in soil is an adaptation of the method used by Pironon et al. [3], Gal et al. [4] and Toutain and Baubron [9]. Measurements at -1 m in the soil are acquired after a micro-drill is made using a battery powered drill to insert a sampling tube through which the soil gas is pumped with a rate of 500mL/min to prevent the intrusion of air from the atmosphere. A stainless steel pipe is used for collecting gases (CO_2 and CH_4) that are measured on-site by FT-IR in the same way than in the borehole. Soil gas samples were taken by filling waterproof bags with a capacity of 1L using a GilAir Plus portable pump (Gilian-Sensidyne, USA). The gases were then analyzed by connecting the bags to a 5 cm optical path cell placed in a Bruker ALPHA infrared spectrometer. CO_2 concentrations of ambient air at soil surface and at +1m above the soil are quantified using a portable pSENSE RH (SenseAir, Sweden) CO_2 sensor to measure the CO_2 concentration (ppm), the temperature (°C) and the relative humidity (%). Calibrations of CO_2 concentration and relative humidity can be performed automatically under conditions defined by the manufacturer. The CO_2 flux is measured using a homemade flux automated dynamic closed chambers (20 x 20 x 20 cm), made of acrylic resin, installed on 5-cm-high bases inserted into the top soil to a depth of 2 cm. A pump was used to circulate gas from the chamber to an infrared gas analyzer (IRGA) measuring CO_2 (Li840, Licor, Lincoln, USA). Slopes of the linear variation in CO_2 concentrations over time were used to calculate CO_2 fluxes [10].

3.3. Reconstruction of atmospheric gas plumes

Recognition by 2D mapping of diffuse CO_2 emissions over a large area (L: 1km, l: 1km, h: 100m) and 3D mapping of the atmospheric plume of CO_2 in selected areas are obtained by deployment of infrared emission via the ®SIGIS/Bruker equipment combined with synchronous thermal cameras. Infrared emission spectra are collected using a scanning imaging IR system (SIGIS Bruker, called SIGIS in the text). Although this device has been described in details by [11, 12], the following quick reminder is done. The SIGIS spectrometer is based on the combination of a modified Michelson interferometer with cube-corner mirrors (interferometer OPAG 33, Bruker Daltonics, Leipzig, Germany) connected to a cooled single MCT (Mercury-Cadmium-Telluride) detector element and a scanning mirror. The system allows definition in an interactive way of the field of the infrared analysis constituted by a rectangular 2D pixelated grid (2058 pixel with X= 98 pixels and Y = 21 pixel). Then, the rotating head is held at a fixed position and the scanning mirror is sequentially set to all positions or pixels from the upper left corner to the bottom right corner in the relevant area. The entire infrared imaging remote-sensing device has been adapted to a small van-type vehicle [11]. Typical 2D measurement planes for CO_2 and CH_4 are expressed in probability of presence (correlation coefficient between 0 and 1) of the 2058 pixels.

4. Results

The results presented here correspond to the first measurement campaign carried out between May 16 and 19, 2022. The minimum temperatures varied from 13 to 16 °C and the maximum between 25 and 31 °C, the weather was mainly sunny without precipitation. The wind speed was quite constant over the period around 15 km/h, with a direction that greatly changed over the day.

The gas collected via the SYSMOG system corresponds to the gaseous phase at equilibrium with the water of the aquifer in the SP-2 well. We note a stabilization of the various indicators after 40 days when the vapor pressure reaches 2 bar. Figure 4 summarizes the evolution of four gases of interest. We note that CO_2, which represents 80 % of the gas collected on the first day, stabilizes at around 50 %, while nitrogen and methane have opposite behaviors. Their concentration increases to stabilize at around 45 % and 3.5 %, respectively. This stabilization occurs more rapidly (after 15 days) for methane than for nitrogen (after 40 days). The water vapor molar concentration remains almost constant around 2.5 %. The differences in behavior of CO_2, CH_4 and N_2 tend to indicate different sources coming to feed the aquifer over time. We can imagine a deep source linked to regional volcanism for CO_2, while N_2 and CH_4 can come from buried sedimentary rocks such as the carboniferous series very present in the region. Finally, the Saint-Parize-le-Châtel fault probably acts as a drain mixing gases of different origins.

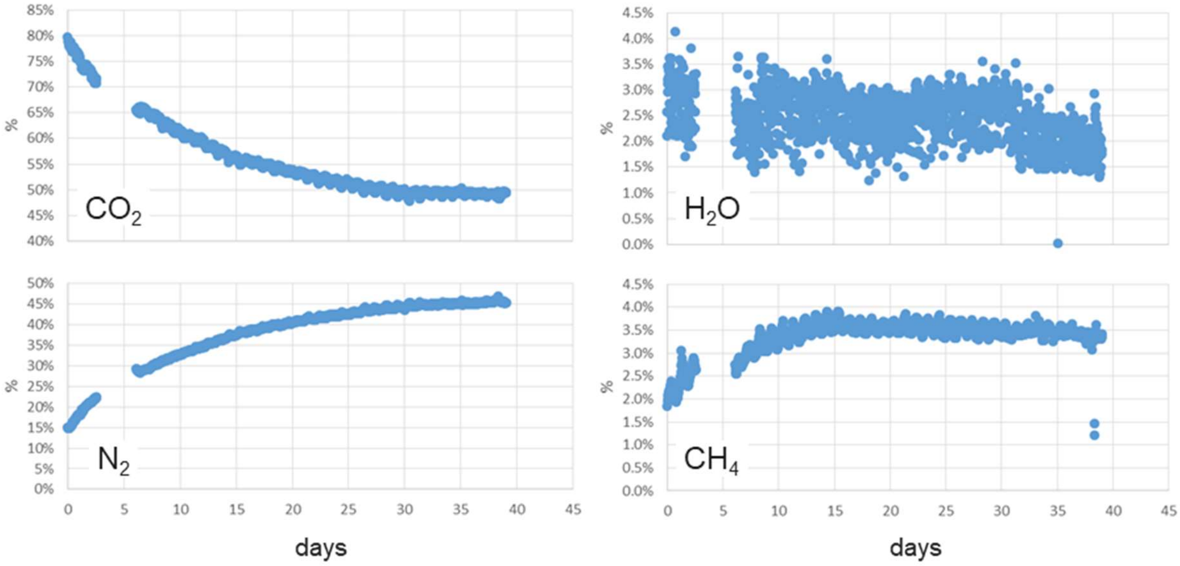

Fig. 4. Gas mole percentage from the vapor phase at equilibrium with the aquifer at 10 m depth in borehole SP-2.

Mapping of CO_2 at -1 m (Fig. 5a) shows levels ranging from 400 to 14,000 ppm, with concentration peaks in the northern part of the study area on either side of the Saint-Parize-le-Châtel fault. These concentration peaks are located in the zone of carbon-gas sources exploited in the past. The CH_4 concentrations vary from 0 to 30 ppm and the concentrations are distributed identically to CO_2 except in the southern part of the site. The close association between CO_2 and CH_4 measured at -1 m confirms the well measurements. Methane is a good indicator of gas of deep origin while the sources of CO_2 are more diverse. The gases detected at -1 m are therefore essentially of deep origin even if it is not possible to eliminate sources of CO_2 from the subsurface. Mapping of CO_2 at ground level (Fig. 6a) reveals concentrations between 500 and 10,000 ppm, the highest of which are located to the north of the site, on either side of the fault. It overlaps fairly well with the CO_2 and CH_4 concentration maps at -1 m. The CO_2 concentrations recorded at + 1m (Fig. 6b) reveal measurements that differ little from the regional atmospheric background noise and are between 500 and 1000 ppm. However, the concentration map shows higher concentrations in the northern part as well as for an isolated point in the south-eastern quarter of the sector. It superimposes fairly well on the maps at -1 m and on the surface. There is nevertheless a strong effect of dilution by the atmosphere because the concentrations measured at -1 m or on the surface of the ground are approximately 1000 times higher than those measured at + 1m.

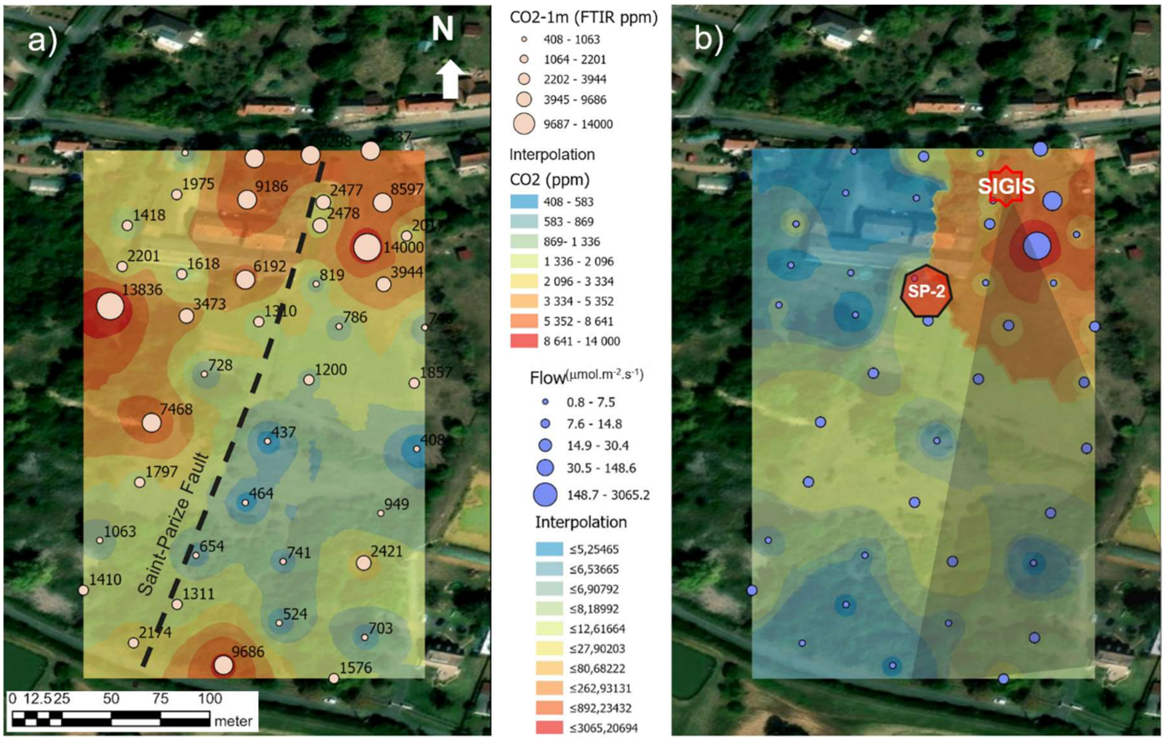

Fig. 5. Mapping of (a) CO_2 concentrations at -1m, (b) CO_2 flow measurements, on the site of Fonts-Bouillants. The red star on Fig. 5b indicates the location of SIGIS equipment and the cone of visualization is drawn. SP-2 borehole is located.

The CO_2 flux measurements (Fig. 5b) range between 1 and 3000 µmol.m^{-2}.s^{-1}. They show a maximum flow in the northeast quarter of the sector, to the east of the fault. The flows in the north-west quarter and in the south are weak to very weak. Control by the fault seems important. It exposes terrain of different nature and petrophysical properties that can govern the transfer of gas from the underground to the atmosphere. Finally, it should be noted that the surface temperature and relative humidity maps show an inverse correlation with a warm (25 °C < T < 39 °C) and dry (20 % < RH < 60 %) northern part and a southern part more humid (60 % < RH < 80 %) where the ground temperature is between 23 °C and 30 °C. These results are rather coherent because the gas emitted is a dry gas which can be responsible for a low relative humidity. However, a single measurement campaign is not sufficient to correlate the T and HR parameters with the site gas contents and flows. The annual seasonal report will make it possible to argue in a more rigorous way.

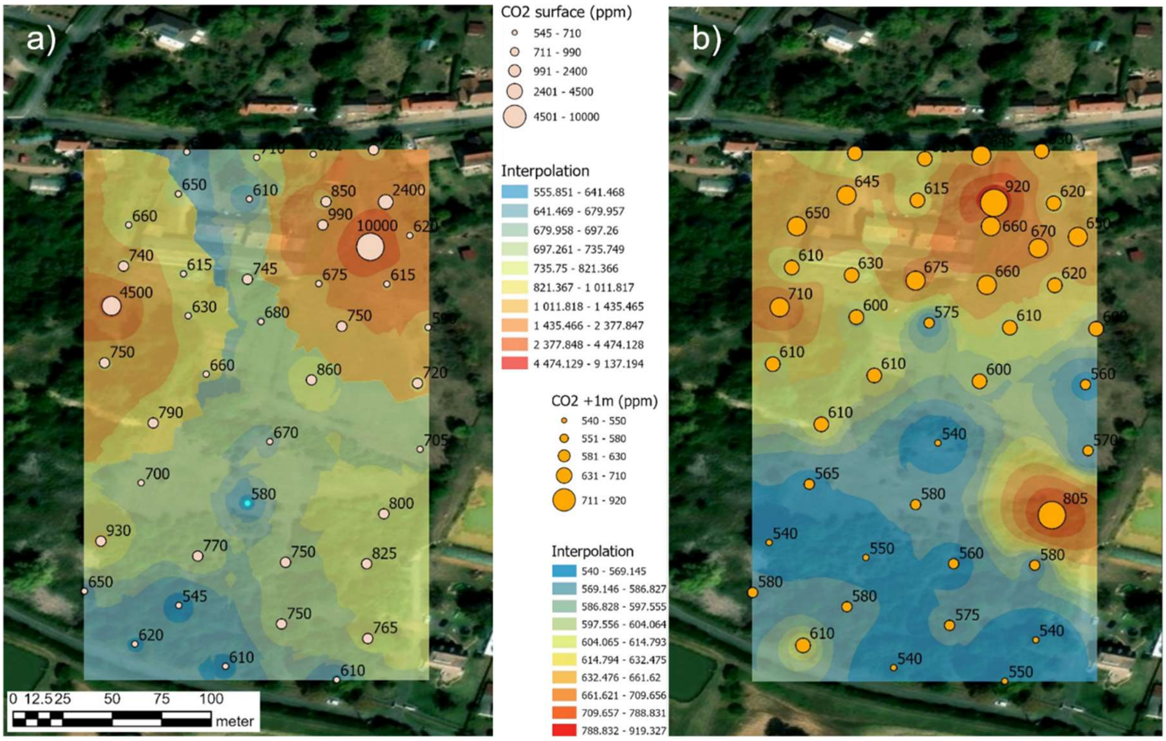

Fig. 6. Mapping of (a) CO_2 concentrations at soil surface, (b) CO_2 concentration at +1m, on the site of Fonts-Bouillants.

Instantaneous maps of atmospheric gases measured by infrared emission remote sensing by the SIGIS device were produced from the northeast quarter of the site (see location on Fig. 5b). Figure 7 shows typical 2D measurement planes of temperature (K) (Fig. 7a), CO_2 (Fig. 7b) and CH_4 (Fig. 7c) expressed in terms of coefficient of correlation. The temperature on the measurement 2D grid fluctuates form 291 K (sky) and 311 K (rest of embankments). Figure 7b confirms that no specific plume of CO_2 can be detected on the field investigated. Presence of CO_2 are more detectable in the sky because of the length of the optical column which is greater than 2000 m. Traces of CH_4 are observable at some points in the space of the 2D grid (Fig 7c) but the concentrations are too weak to be quantified (i.e. below the sensitivity limit of the SIGIS that is 2 ppmv). These results confirm the intense atmospheric dilution of the gases emitted from the soil in the atmosphere considering the weather situation of the first campaign of measurements at Fonts-Bouillants.

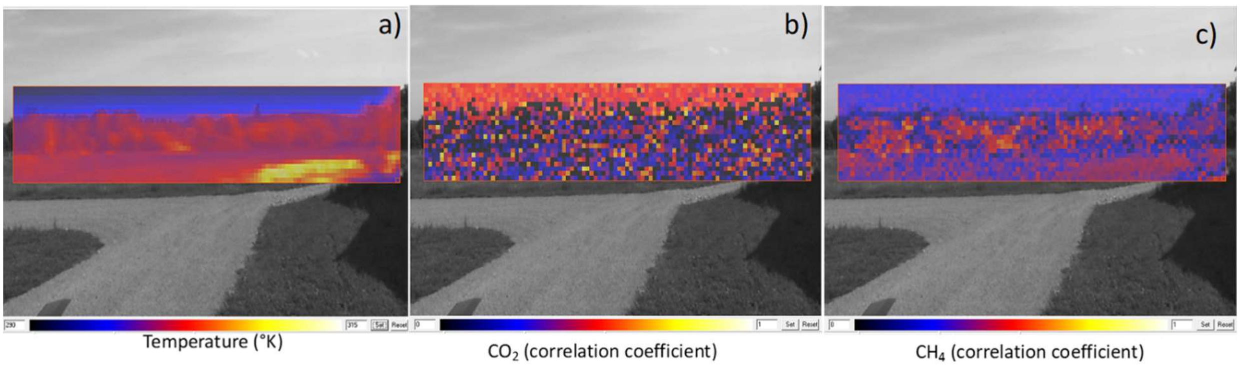

Fig. 7. SIGIS 2D measurement planes of (a) temperature (K), (b) CO_2 and (c) CH_4 expressed in coefficient of correlation.

Discussion

This first measurement campaign on the Fonts-Bouillants site (Nièvre, France) showed that the technologies deployed were suitable and made it possible to acquire consistent data series. Such a combinations of tools allow to have a vision in time and space of natural CO_2 emissions in a region where the gas fumes contain mainly CO_2 but also nitrogen and methane as well as trace gases such as helium. Maps at the scale of the site made it possible to locate the zones with high emissive potential and the zones where the gas flow is high (> 100 $\mu mol.m^{-2}.s^{-1}$). Thus the north-east quarter of the site combines both the highest concentrations of CO_2 and CH_4 at -1 m and the concentrations on the ground and at +1 m as well as the most intense CO_2 flux. We can conclude that the atmospheric dispersion of gases naturally emitted is great because the measurements at + 1m of CO_2 are close to the atmospheric background, despite a weak wind. In addition, remote sensing infrared emission maps (SIGIS) confirm the absence of gas pockets in the near atmosphere.

On the base of these first data, it is therefore possible to quantify a mass of CO_2 emitted naturally on the scale of one year over an area of 400 m^2 covering the high emission zone in the north-east of the site. From a maximum flux of 3000 $\mu mol.m^{-2}.s^{-1}$, we obtain a mass of CO_2 naturally emitted of 1700 tonnes, i.e. approximately 2000 times greater than the mass of CO_2 transferred to the atmosphere in the lowest emission areas. These approximations will be refined during the next measurement campaigns which will take into account climatic variations on the scale of a seasonal cycle.

This example shows that the NECCS technology (Natural Emission of Carbon dioxide with Capture and Sequestration) is much more effective than the DACCS technology which aims to capture diluted atmospheric CO_2 [13]. Large-scale deployment requires an inventory of naturally emissive sites and the development of capture technologies suitable for gases richer in CO_2 than the atmosphere. Figure 8 shows the layout of a NECSS unit for the capture of CO_2 naturally emitted on the site, the purpose of which is to capture CO_2 that leaks from the underground into the atmosphere. This unit consists of a separation module to purify the CO_2, compress it to transport it via carboducs to factories for use (sustainable mineralization from by-products or industrial waste for the production of recoverable materials) or to geological reservoirs for long-term storage. An important advantage of NECSS technology lies in the possibility of recovering gases co-emitted with CO_2, such as gases with a strong climate impact such as CH_4 or strategic gases such as helium or hydrogen.

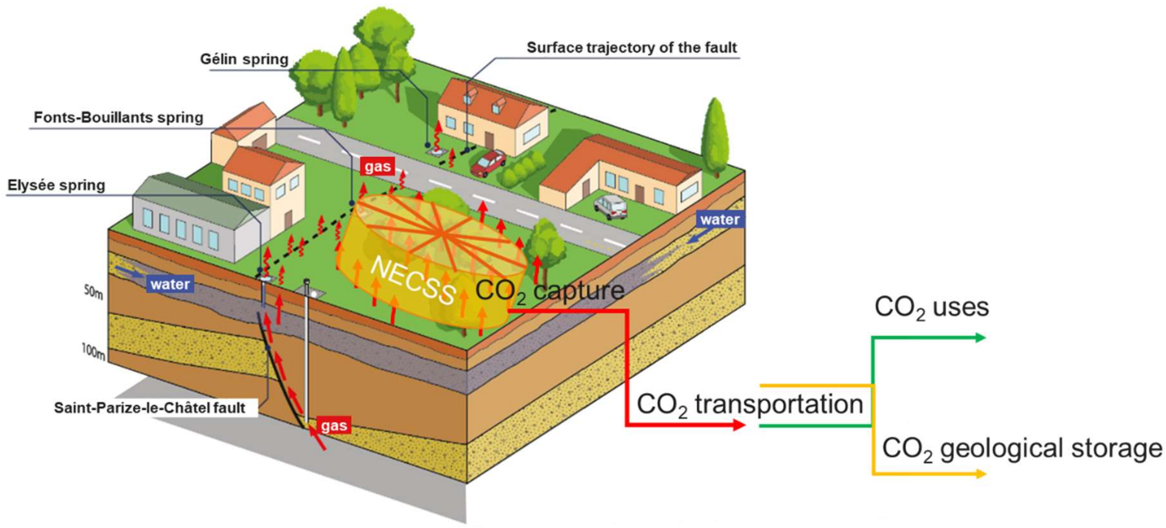

Fig. 8. Schematic view of the Fonts-Bouillants site and the hypothetical location of a NECCS unit for the capture of CO_2 emitted naturally to the northeast of the study area.

This already very complete approach to measuring gases on a geographical site of interest can benefit from improvements. Thus, in order to refine the tracing of gas sources, it would be good to couple the data acquired with isotopic measurements of carbon and oxygen of CO_2. In order to better trace the flow of deep origin, it would be useful to measure the flow of CH_4 and to record thermal images by drones (day and night) covering the entire site to better detect gas emission points.

Conclusion

The Fonts-Bouillants sector is a natural site of CO_2 emissions. It has been shown that the gas emissions are governed by the Saint-Parize-le-Châtel fault and the gas emissions have been characterized according to profiles ranging from -10 m, -1 m, 0 m, +1 m to the atmosphere by mixing different means and analytical procedures.

CO_2 is accompanied by major gases (CH_4, N_2) and minor gases such as helium. The CO_2/CH_4 and CO_2/N_2 ratios measured in the gas phase at equilibrium with the borehole water show that they vary with time and that these gases probably do not have the same origin. Mappings of CO_2 at -1 m, at ground level and at +1 m show that the areas with high emissions are located to the north of the sector studied on either side of the Saint-Parize fault. Nevertheless, the highest CO_2 flux is concentrated in the north-east quarter of the sector with flux values reaching 3000 μmol.m^{-2}.s^{-1}. If this flow was captured, it could make it possible to subtract around 1700 tons of CO_2 per year from the atmosphere on a reduced surface of 400 m^2. This estimate is maximum because the flow taken into account is the maximum flow measured on the study area. One of the objectives of this work should lead us to extrapolate these measurements to the scale of the Saint-Parize-le-Châtel fault, provided that we have a precise location of the mofettes.

This baseline strategy makes it possible to establish mass balances and assess areas favorable to CO_2 capture. In addition, these new data will make it possible to develop a methodology for the quantification of natural CO_2 emissions elsewhere in France and in the world with the aim of deploying the NECCS technology. Whatever the technology deployed, such an approach shows its interest for the quantification of CO_2 fluxes from the underground to the atmosphere. It could be recommended to apply it before any CCS operation.

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