Assessment of the *in situ* biomethanation potential of a deep aquifer used for natural gas storage

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Abstract

In response to the challenges of sustainable development and the H_2 sector, it is foreseeable that H_2 will be stored into geological storage, such as deep aquifers. However, CO_2 evolves in deep aquifers because it may be naturally present there; it may also be a constituent of the stored gas mix, or could even be voluntarily stored in the context of the fight against global warming. Autochthonous microorganisms can consume them as sources of energy and carbon (methanogens, (homo)-acetogens and sulfate-reducers). This was already demonstrated in a previous experiment (Haddad 2022) and under operating conditions (Lobodice, Czech Republic ; Smigan 1990).

Understanding these mechanisms and quantifying them appear necessary to assess the modifications generated by this type of microorganisms on the properties of the gas. The methanogenesis reaction ($CO_{2\ gas} + 4H_{2\ gas} \rightarrow CH_{4\ gas} + 2H_2O_{\ liquid}$) induces a lowering of pressure, since 5 gas molecules are transformed into a single gas molecule: CH_4 (water being condensed at subsurface conditions). *In situ* biomethanation technique could represent a potential on several scales larger than conventional catalytic or biological methanation reactors, due to the very large reservoir volumes involved. Biomethanation in geological reservoirs would enable us to reduce our consumption of fossil fuels, so as not to emit more CO_2, while meeting the growing energy needs of a region and ensuring its independence from hydrocarbon-producing countries.

A deep aquifer already used as UGS was selected for this study. Formation waters from 17 control wells in this aquifer (Fig. 1) were sampled to assess the potential activity of indigenous methanogenic populations, as well as sulfate-reducers. Despite relatively low sulfate concentrations for a deep aquifer (0.025-1.35 mM), sulfate reducers were found at all sites targeting and quantifying the *dsrB* gene, which is characteristic of this metabolic

group (between $1.8 \cdot 10^1 \pm 2.0 \times 10^0$ and $1.3 \cdot 10^4 \pm 2.0 \cdot 10^3$ *dsrB* gene copy numbers.mL^{-1}). In contrast, methanogenic archaea based on the *mcrA* gene quantification were detected at only 10 of the 17 sites (up to $4.3 \cdot 10^2 \pm 8.3 \cdot 10^1$ *mcrA* gene copy numbers.mL^{-1}). The choice was made to focus the rest of the study on 7 of these 10 sites. The potential for methanogenesis was assessed on cultural tests with formation water alone or supplemented with calcite ($CaCO_3$), a mineral present in the formation. Results indicate that initial times and controls are controlled by the sulfate variable, since the latter was not consumed by sulfate-reducers. Biotic trials in the presence of calcite and H_2/CO_2 (abiotic controls and final times) are logically characterized by higher concentrations of calcite, bicarbonate and calcium, but this is not the case for trials in the presence of H_2 alone. We therefore deduce that methanogenesis took place mainly *via* gaseous CO_2, but that without the latter, calcite was a source of carbon for lithoautotrophs. Cultures incubated with H_2 as the sole gas phase have the highest methane concentrations, logically associated with the lowest sulfate concentrations (consumed by sulfate-reducers), the lowest *Eh* (probably due to the presence of sulfides) and more alkaline pH values up to 10 (which may have led to precipitation of carbonate and calcium ions). All the sites studied showed sulfate consumption and methane production. Analysis of taxonomic diversity (MiSeq; *16S rRNA* gene V4-V5) showed the dominance of three genera of sulfate-reducers with *Thermodesulfovibrio-Desulfovibrio-Desulfotomaculum* and methanogenic populations belonging to the *Methanobacterium* genus.

These initial results show a strong potential of *in situ* biomethanation for the deep aquifer studied. All these experiments were carried out at near-atmospheric pressure, and the results still need to be confirmed and refined in the laboratory under conditions that simulate real-life conditions as closely as possible (rock, pressure, nature of gases).

Keywords

Deep aquifer, Underground Geological Storage, *in situ* biomethanation, H_2 storage

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Conflicts of interest

The authors have declared that no competing interests exist.

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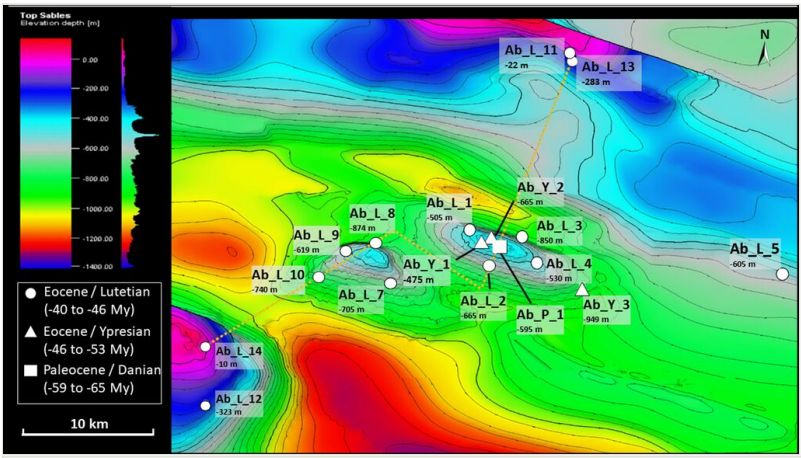

Figure 1.

Location map of the sampled sites of the studied deep aquifer used as UGS.