Soil and Geologic Formations as Antidote for CO₂ Sequestration?

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Rapid exploitation of fossil fuels is associated with significant CO₂ emissions. According to the Paris Agreement, total anthropogenic CO₂ emissions should be reduced to less than 20 Gt per year by 2050, even for reaching a "net-zero emissions" status by the end of this century to hold the increase in the global average temperature to "below 2°C above pre-industrial levels". The latest report of the Intergovernmental Panel on Climate Change (IPCC, 2018) showed that limiting global warming to 1.5°C above the pre-industrial level would significantly reduce the damaging impacts on ecosystems and human health and well-being compared to 2°C rise, ensuring a more sustainable and equitable society. Reducing CO₂ emissions to this extent will require "rapid and

far-reaching transitions". This means that remaining emissions should be balanced by removing CO₂ from the air. Faced with such a formidable challenge, carbon capture and storage within soil and geologic formations is considered to be one of the most promising and practical options (Beiser-McGrath & Bernauer, 2019). Although CO₂ can be transformed and stored in the forms of chemicals, industrial gases/fluids, or biomass, the sequestered carbon would return to the atmosphere within several months to decades. By contrast, CO₂ sequestration in soil and porous rock formations is considered to be a permanent solution, because the CO₂ could transform into stable carbonates below the Earth's surface (Fig. 1). It is estimated that the calcium and magnesium carbonates from mineral carbonation in soil and rocks are theoretically sufficient to fix all the CO₂ produced by the combustion of all available fossil fuel reserves (Leclaire & Heldebrant, 2018). Besides, the global geological storage capacity (8,000 to 15,000 Gt CO₂) completely fulfils CO₂ storage demands for several centuries (20 Gt CO₂ annually). Thus, CO₂ from the emissions of exploited underground fossil fuels then returns to the underground mineral sink as a long-term solution to limit climate change.

For the *in-situ* geological approach, pressurized CO₂ is injected into porous rock formations buried hundreds of meters below the Earth's surface. Natural carbonation is a slow process, but surprisingly, a pilot project of CO₂ injection in Iceland reported that after 2 years, more than 95% of the injected CO₂ in lava flows might have been mineralized into carbonates (Kintisch, 2016). The above research also suggests that a modification of the deep ecosystems occurs in response to the CO₂ injection, as evidenced by a bloom of chemolithoautotrophic bacteria possessing an additional ability to promote autotrophic carbon-fixation. *In-situ* CO₂ injection also has a valueadded application in enhancing oil recovery (EOR) from oil fields. It is estimated that more than 90% of the world's oil reservoirs are suitable for CO₂-EOR application, which could significantly reduce the net cost of geological CO₂ sequestration. For *ex-situ* mineral sequestration, Ca/Mg/Ferrich minerals in soil and rocks are carbonated in hydrothermal reactors with pressurized CO₂. Alkaline soil and rocks have a great potential for CO₂ sequestration via neutralization reaction. Solid mineral wastes remain one of the world's largest waste streams, and most of these wastes are in slurry form as a result of industrial washing processes. Some of these mineral wastes, especially basaltic and ultramafic minerals can trap CO₂. Recent research has reported that ground ultramafic fluids to generate hydrogen gas under low-temperature condition. Along the reaction path of the hydrothermal alteration of ultramafic soil and rocks, silicate mineral replacement reactions concomitantly release Fe for incorporation into the secondary oxide and carbonate phases. Therefore, alkaline soil and rocks, mining wastes, and secondary minerals can be regarded as precious resources for CO₂ sequestration.

The technical and economic feasibility of CO₂ sequestration in soil and rocks should be further assessed holistically. *Ex-situ* mineral carbonation in soil and rocks is a safe but expensive and energy-intensive scenario ($$47-87 t^{-1} CO_2 storage$). Accelerating the carbonation of minerals in soil and rocks requires mining, transportation, crushing, grinding, and safe-handling of the material, or pre-treatment of mineral wastes that are suitable for carbonation. Reducing the energy consumption of accelerated carbonation by utilizing renewable energy, improving the mineral-fluid interfaces, or introducing appropriate catalysts/bio-catalysts should be further investigated. By comparison, *in-situ* CO₂ storage in the underground sink would be inexpensive (from \$1-8 t⁻¹).

CO₂ stored for onshore, to \$7-22 t⁻¹ for offshore) and consume less energy. The major concern is the potential CO₂ leakage from the mineral mass. The latest research has predicted that over 98% of the injected CO₂ can be retained in the subsurface for over 10,000 years (Alcalde et al., 2018). However, the volume of minerals would increase by 1.74 times after carbonation, based on the carbonation of Mg₂SiO₄, which may induce cracking and change the stability of the geological structure. Therefore, the amount of CO₂ injection should be below the critical value based on the volume of the storage layer and the porosity of the minerals with a factor of safety. Although the injected CO₂ could be quickly transformed into biomass by chemolithoautotrophic bacteria in the subsurface, carbon storage as microbial biomasses is not desirable since long-term stability of the biologically-stored carbon cannot be ensured. Transforming the biomass to stable carbonates in the underground environment still faces some technical barriers. Achieving a breakthrough with these currently available technologies requires urgent interdisciplinary and international research collaborations, as well as support and incentives from government, industry, and the general public. Successful CO₂ sequestration in soil and rocks at a large scale could ease the tremendous pressure of intensive climate change, and realize a "net-zero emissions" or "carbon negative" society in the near future.



Fig. 1. Concept of *in-situ* geological CO₂ storage and *ex-situ* mineral carbonation.

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Notes

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