Geophysical Research Abstracts
Vol. 15, EGU2013-7510, 2013
EGU General Assembly 2013

The CO_2 Vadose Project - Buffering capacity of a carbonate vadose zone on induced CO_2 leakage. Part 2: reversed numerical simulation with PHREEQC

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The interest in CO_2 capture and storage as a method of reducing CO_2 emissions has underlined the need for more knowledge regarding the geological storage capacity. Because the ultimate failure of geologic CO_2 storage occurs when CO_2 seeps out of the ground into the atmospheric surface layer, it is of primary interest to understand how much vadose zone could buffer a CO_2 leakage. To assess the buffering capacity of the carbonate vadose zone with respect to this diffuse CO_2 leakage, numerical simulation using PHREEQC were performed with data obtained from CO_2 leakage experiment.

One of the aims of the CO_2-Vadose Project is to perform an experimental release of CO_2 and associated tracers (He and Kr) in order to study CO_2 transport and geochemical reactions along the carbonate vadose zone. Experimental site, which is a cavity of about 9 m^3 located at about 7 m in depth in a former underground limestone quarry in Saint-Emilion (Gironde, France), was set up with more than ten gas probes around the injection cavity in order to follow CO_2 concentrations before and after injection thanks to micro-GC and infrared analyser. Micro-climatic parameters were also recorded by a weather station at the site surface and around the injection room (barometric pressure, relative humidity, temperature). About 11 m^3 of gas mixture was released in the injection room and different concentrations of CO_2 were observed inside and all around the cavity, in limestone.

At the end of the gas mixture injection, the observed CO_2 concentrations were about 90 % in the experimental cavity. A few meters away from the source, CO_2 concentrations varied from atmospheric level (about 400 ppm) to about 11,000 ppm. Numerical simulations were done with PHREEQC to understand the kinetic and thermodynamic equilibrium of reactions occurring in limestone, to figure out how the carbonate vadose zone could buffer this CO_2 leakage. Field characterisation data, moisture content data, pore-water analyses results and CO_2 concentrations observed during experiment were integrated to the simulations to determine how much calcite was dissolved and in which time scale. Results from numerical simulation show that reaction reached equilibrium in few hours, mainly because of the low thickness of irreducible water layer in macropores, where gas transfer occurred.