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Denitrification in deep fractured aquifer investigated through batch experiments

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Fractured aquifers are characterized by extremely heterogeneous fluxes, water circulating almost exclusively in fractures. Exploitation of fractured medium leads to a reorganization of water fluxes and contribution of fractures. In western France a deep fractured aquifer located in crystalline rocks is exploited for tap water since 1991. The beginning of the exploitation leads to drastic changes in anion concentration. Chloride concentrations increase by 35% for 2 years and grows slowly since that time. But chloride concentrations remain lower than in a zone not influenced by pumping where fluxes are slower. Sulphate concentrations are still increasing since pumping initiation and nitrate concentrations reache a low level (from 16 to about 4mg/L) after only 418 days. Sulphate and nitrate concentrations and isotopic measurements allow to characterize an autotrophic denitrification process: nitrate reduction correlated to sulphide oxidation. It appears clearly that pumping, i.e. increasing fluxes, induces the conditions required for the biogeochemical reaction. However, the control of the physical fluxes on bacterial development is not understood.

In order to investigate the influence of water circulation in fractured rocks, batch experiments have been carried out. Fresh granite, altered granite and schist coming from borehole drilling were crushed and separated into two fractions: coarse grain fraction (1 to 4mm) and fine grain fraction ($<$1mm). In 250mL batch reactors, each fraction is mixed with water (1:3). The water is either nitrate-poor or nitrate-rich. Atmosphere was flushed with N2 to maintain field anaerobic conditions. Common anion concentrations (SO4, NO3 and Cl) were monitored during the experiment.

First, in finest rocks reactors, chloride concentrations increases by about 10 to 15%

within a few hours. Sulphate concentrations increase during the first day and remain relatively stable after. The highest values are found for the fine grain fraction. A clear correlation between Cl and SO4 is shown. For altered or fresh finest fractions of granite, nitrate concentrations decrease by 50% in 3 days and a nitrite peak occurs simultaneously. In coarse grain fraction reactors, nitrate concentrations decrease by only 10% whereas no nitrite peak can be observed. In schist experiment, the same anion concentration variations are observed in the fine and coarse grain experiment. However, the fine grain experiments show a denitrification of about 30%.

Crushing of rocks allows the fluids, contained in microporosity and fluid inclusions to contribute to surrounding water. Chloride and sulphate concentrations increase can be attributed to this process. Field evidences of high salinities indicate that such process might be important when very slow circulation occurs in fractured aquifers. The beginning of pumping can initiate circulation in some fractures that were initially not connected to the system and lead to element concentration increase. This suggests that even if sampling water is only 100m-depth, washing by rain have not erased signature of old circulating fluids. Pumping also initiated a denitrification process that seems to be related to surface availability and type of rocks. The re-organization of water fluxes and pathways by pumping changes availability of fractures, which increases surface reaction potential and leads to the change of water chemistry observed.