Geophysical Research Abstracts
Vol. 12, EGU2010-11485, 2010
EGU General Assembly 2010

Column experiment to study isotope fractionation of volatile organic contaminants in porous media under unsaturated conditions

Simon Jeannottat, Daniel Hunkeler, and Florian Breider
University of Neuchâtel, Centre of Hydrogeology, Neuchâtel, Switzerland (simon.jeannottat@unine.ch)

Pollution by organic contaminants such as petroleum hydrocarbons and chlorinated solvents is common in industrialized countries. The use of stable isotope analysis is increasingly recognized as a powerful technique for investigating the behaviour of organic or inorganic contaminants. Recently, compound-specific isotope analysis (CSIA) has proven to be an effective tool to confirm and quantify in-situ biodegradation by indigenous microbial populations in groundwater.In contrast, only few studies have investigated the use of CSIA in the unsaturated zone. In the unsaturated zone, the main potential applications of CSIA include the assessment of biodegradation and the fingerprinting of different sources of petroleum hydrocarbon or chlorinated solvents vapours. However, it has to be taken into account that isotope ratios in the unsaturated zone can vary due to diffusion and volatilization in addition to biodegradation. For application of isotope methods in the unsaturated zone, it is crucial to quantify isotopic fractionation resulting from physico-chemical and transport processes.

The study is focused on laboratory experiments that investigate the effect of vaporization and diffusion on isotope ratios. The effect of diffusion is carried out using a column experiment setup that can be considered to represent VOC transport from a floating NAPL towards the atmosphere. Furthermore, additional column and batch experiments will be conducted to better understand the effect of biodegradation. Volatilization is studied with an other experimental setup. In addition, a mathematical framework was developed to simulate the isotope evolution in the column study.

Since the initial experiments aimed at investigating the effect of vaporization and diffusion only, the column is filled with dry quartz sand in order to avoid perturbations of concentration profiles by humidity or adsorption on organic matter. An activated sand will later be used for the biodegradation experiments. A mixture of nine contaminants (pentane, MTBE, hexane, benzene, isooctane, methylcyclohexane, toluene, octane and xylene) that represents a wide range of hydrocarbons is emplaced in the column. Periodical measurements of concentrations and δ^{13}C values were carried out in the source chamber and at different distances along the column. A depletion of ^{13}C with distance is observed, which is due to faster diffusion of substances enriched in ^{12}C. The shift of δ^{13}C values towards more negative values is more significant during the first hours of the experiment. After some hours, the value stabilizes when a steady state is reached. These results fit well the analytical models. These results demonstrate that stable isotope profiles are reached under steady state conditions even though molecules with light isotopes only diffuse faster than molecules with heavy isotopes. This is an important finding for the application of isotope analysis to link VOC vapours to contaminant sources and to demonstrate reactive processes based on shifts in isotope ratios.

Further experiments will be conducted to study the isotopic response to diffusion, vaporization and biodegradation of chlorinated solvents (PCE, TCE) using quite a similar column setup. Stable hydrogen and chlorine isotopes will also be measured during the same experiments in order to better constrain the different processes and fingerprinting sources of contaminations.