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Evolution of carbon isotope composition of pore waters in the unsaturated zone: open or closed system conditions?

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Although radiocarbon is being used as a groundwater dating tool for more then four decades now, a number of questions related to this method still remains open. Among the problems not fully solved yet is the question of carbon isotope composition of the Dissolved Inorganic Carbon (DIC) pool in the infiltrating water recharging the given groundwater system. Two conceptual models addressing this question have been proposed in the literature: (i) the open-system model assuming that pore water in the unsaturated zone is in continuous contact with infinite reservoir of soil CO_2 of constant parameters (partial pressure, carbon isotope composition) during the process of dissolution of mineral carbonate phase, (ii) the closed-system model where the infiltrating water first equilibrates with soil CO_2 reservoir and then moves to the region where dissolution of mineral phase takes place, without contact with the CO_2 reservoir. Both models lead to very different carbon isotope composition of DIC pool, particularly with respect to radiocarbon content, and to different carbonate content of the pore waters. Although both models are widely used in interpretation of radiocarbon data in groundwater, studies addressing validity of both models under field conditions are rare.

The carbon isotope evolution of DIC pool has been studied in the unsaturated zone of ca. 8 m deep, consisting mostly of loess deposits and equipped with multilevel sampling system for both liquid and gaseous phase. The study area was located in southern Poland, in the vicinity of Krakow. From June 2005 to September 2006 the following

parameters were regularly measured at 7 different levels in the unsaturated zone: (i) parameters of soil CO_2 (partial pressure, δ^{13}C, radiocarbon content), (ii) parameters of pore waters (chemical composition, pH, δ^{13}C, radiocarbon content), and (iii) soil temperature. The collected data allowed a better insight into processes controlling isotopic composition of DIC pool in time and space. The chemical and isotopic evolution of pore waters at different levels of the unsaturated zone was modeled using PHREEQC code, assuming either open or closed-system conditions.

Due to strong seasonal fluctuations of CO_2 partial pressure in the unsaturated zone, pore waters taking part in the infiltration process were loosing carbon during winter months, when partial pressure of soil CO_2 is low, and gaining it again during summer months. For the investigated profile the given parcel of pore water was subject to ca. 20 such cycles before it eventually left unsaturated zone. Although those processes should occur close to chemical and isotope equilibrium, presence of some kinetic effects cannot be ruled out. This stems from the fact that, on micro scale, there is a possibility that the contact between pore water and soil CO_2 reservoir is broken for some period of time. Without detailed studies it is not possible to quantify the magnitude of this effect. Also, it is not possible to establish whether the given parcel of pore water passing in the course of its downward movement different “episodes” of open and closed-system conditions, has ever a chance to establish full chemical and isotope equilibrium. On macro scale, the observed changes in chemical and isotopic composition of pore waters in the course of their passage through the unsaturated zone suggested that open-system conditions prevail within the profile, however with some signs of gradual closing of the system toward the water table.

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