Experimental study on groundwater and spring water dating method using SF_6 tracer during baseflow condition in headwater catchment

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Residence time, the elapsed time from when rain water are recharged into catchment until it exits at the outlet, is important hydrological descriptor of catchment subsurface flow path, storage, turnover characteristics of anthropogenic contaminants or solutes, and used as an indicator of catchment land use change effects. For estimating relatively young residence time of water output in small catchment or hillslope scale, seasonal variability of oxygen and hydrogen isotopes ($\delta^{18}O$, $\delta^{2}H$) or chloride ions (Cl^-) in rain water and their damping in output is commonly used. The method of anthropogenic gas tracer, i.e. chlorofluorocarbons (CFCs) and sulfur hexafluoride (SF_6), are also commonly used for groundwater dating, and the analytical apparatus has recently been introduced into Japan since about 2010's and application examples have been increasing. However, there are only few studies considering gas recharge conditions and what they mean, and particularly in Japan, each study uses different, not unified assumptions of gas recharge conditions. In this study, we focus on the effects of value setting of gas recharge conditions premised on output water dating using SF_6 tracers and test the effects of seasonal recharge temperature variation on estimated mean residence time. The observed value of dissolved SF_6 are used collected in small catchment nested in Kiryu experimental watershed (KEW) located in Tanakami mountain, Shiga Prefecture, Japan (5.99ha, N34°58', E136°00') in this study. For groundwater and spring water dating, FLOWPC (ver.3.2.) program, developed by Maloszewski et al., is used.

Following the Henry's law, dissolved gas concentrations at the equilibrium condition are derived from Henry's constant K_H and the gas molar partial pressure, both are parameterized by temperature, salinity and elevation at the time when the gas recharging to water had been occurred. In KEW, where the elevation width are relatively small (190-255m), the fluctuation effects of recharge temperature are remarkable on estimated mean residence time. Considering seasonal variation of temperature of gas recharge condition, dissolving atmospheric SF_6 recharged into catchment are numerically examined using various yearly seasonal temperature records of air, unsaturated soil layer, saturated soil and bedrock groundwater. The results indicate that calculated dissolving SF_6 input may have a wave of one-year cycle and suggest that dissolved SF_6 in current water outlet may also fluctuate up and down. In addition, it can be pointed out that even when dissolved gas concentration was detected in relatively high value it can be explained due to the fluctuation of output concentration. And the mean residence time estimated considering residence time distribution model of SF_6 input by FLOWPC program, for example of saturated soil groundwater located in catchment hillslope, is 30-70 months with width depending on the seasonality and applied type of temperature record.

In this study, we conducted numerical experiment using observed SF_6 concentrations in groundwater and spring water and revealed that considering temperature setting and recharging seasonality of gas tracer cause fluctuation of both input and output gas concentrations, and related value of mean residence time. However, the estimated value using the dissolved SF_6 or CFCs in each sample is a momentary value which may change in time, and only once sampling is insufficient to elucidate whole of catchment hydrological characteristics of rainfall-runoff response. In order to estimate mean residence time in headwater catchment, flexible approaches considering catchment time-variable hydrological characteristics of rainfall-runoff response and stream water chemistry are needed. We emphasize it is also needed to examine the uncertainty of the residence time estimated from this method and discuss the methodological limits carefully for the future hydrological study of residence time estimation using gas tracers.

Keywords: headwater catchment, mean residence time, sulfur hexafluoride, FLOWPC, residence time distribution model, gas recharge temperature