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Using carbon and water isotopes and noble gases to assess the origin of methane in fresh water aquifers in the south of the Netherlands

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Groundwater in the Dutch subsurface is known to contain substantial concentrations of methane of which the origin is not always clear. The Dutch subsurface contains relatively high organic matter contents which makes a biogenic origin plausible, however few studies have used water and carbon isotopes to deduce the origin of methane. In relation to possible future exploitation of deep shale gas resources, it is now considered important to assess base line quality of fresh groundwater in overlying aquifers from which drinking water is produced. Therefore, we sampled the raw water of 41 large public supply well fields in the south of the Netherlands which represents a mixture of groundwater of different ages and used the a discrete travel time distribution model (DTTDM, Visser et al. 2013, WRR) in order to quantify the age distribution of the mixture. Measurements included major ion chemistry, 3H, 3He, 4He, 18O, 2H, 14C, 13C-DIC and 13C-CH4 and the full range of noble gases. 13C-CH4 measurements were carried out using a Picarro G2201-i CRDS analyser. The heavier noble gases enable the calculation of the Noble Gas Temperature (NGT) which characterizes the temperature of past recharge conditions. The 14C apparent age of each mixture was derived correcting for dead carbon sources and included carbonate dissolution and methanogenesis as the defining processes. The 13C-CH4 measurements showed a range of δ-values between -70 and -100‰ which give a clear indication for biogenic methane. No clear relations between 13C-CH4 and 13C-DIC or the 4He/CH4 ratio were observed. However, clear spatial patterns indicated that more depleted values are grouped in specific areas. The 13δCH4 values did not show a clear relation with the age distribution of the pumped water, even though a large range of age distributions was observed including old water with an age of > 25 k yrs. We believe that spatial differences in organic matter contents, origin of the geological deposits and/or the methanogenesis process itself determine the methane isotope ratios in this part of the Netherlands.