Can bomb-peak tritium persist in the transition zone? A case study from the German island of Langeoog

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ABSTRACT

Tritium has been used as a tracer in hydrogeological studies to date young groundwater. It was released in massive amounts to the atmosphere by nuclear bomb testing in the 1950's and early 1960's. Its activity in the atmosphere peaked around 1963-1964 and has been steadily falling since then due to its half-life of 12.32 years. On the northern hemisphere, where the atmospheric tritium concentration reached much higher levels than on the southern hemisphere, elevated tritium activities in groundwater persist, and thus can still be used to identify groundwater that has recharged during the post-bomb era.

Tritium has also been encountered in brackish and saline groundwater in coastal aquifers. Stuyfzand *et al.* (2012) found ^{3}H values of 30.3 TU in recently intruded North Sea water directly below the beach near the city of The Hague in the Netherlands. These high values are linked to the discharge from nuclear reprocessing plants in the UK and France (Nies *et al.*, 2010). Lower, but still above natural background, values ($3.3 < {}^{3}H < 23.9$ TU) were measured in samples from a wedge of brackish groundwater at the same study site. This wedge was therefore believed to have intruded during the period of maximum groundwater extraction around 1956. Bryan *et al.* (2016) studied a freshwater lens system on Rottnest Island off the coast of Perth in Western Australia. All but one saline groundwater samples had tritium concentrations below the detection limit. The one exception had a ^{3}H activity of 0.67 TU, which was believed to indicate relatively recent intrusion of seawater.

Sivan *et al.* (2005) studied the distribution of tritium in the mixing zone of a coastal aquifer south of Tel Aviv, Israel. Similar to Bryan *et al.* (2016), they interpreted the presence of measurable tritium in their saline groundwater samples as an indication for seawater intrusion during the 60 years prior to their study. However, they also considered the possibility that the tritium in the transition zone was not derived from the intruded seawater but from the overlying freshwater. That is, old, tritium-free seawater may obtain tritium by mixing with young, tritium-containing freshwater, especially if the freshwater was impacted by the bomb peak.

On the island of Spiekeroog, located in the northern part of Germany, Röper *et al.* (2012) detected virtually no ^{3}H activity in saline groundwater samples from depths of 60 m below sea level or more, and they inferred an age of over 70 years. Chloride concentrations and stable water isotope values indicated that the samples contained up to 35% freshwater, but apparently this mixing did not contribute appreciable amounts of tritium. By contrast, previously-unpublished data from the neighbouring island of Langeoog show that groundwater in the transition zone beneath the freshwater lens has elevated ^{3}H activities. Interestingly, the freshwater tritium activities have been found to decrease with depth to values < 2 TU at the base of the lens (Houben *et al.*, 2014), while in the transition zone tritium values increased to up to 5.7 TU again (Figure 1c).

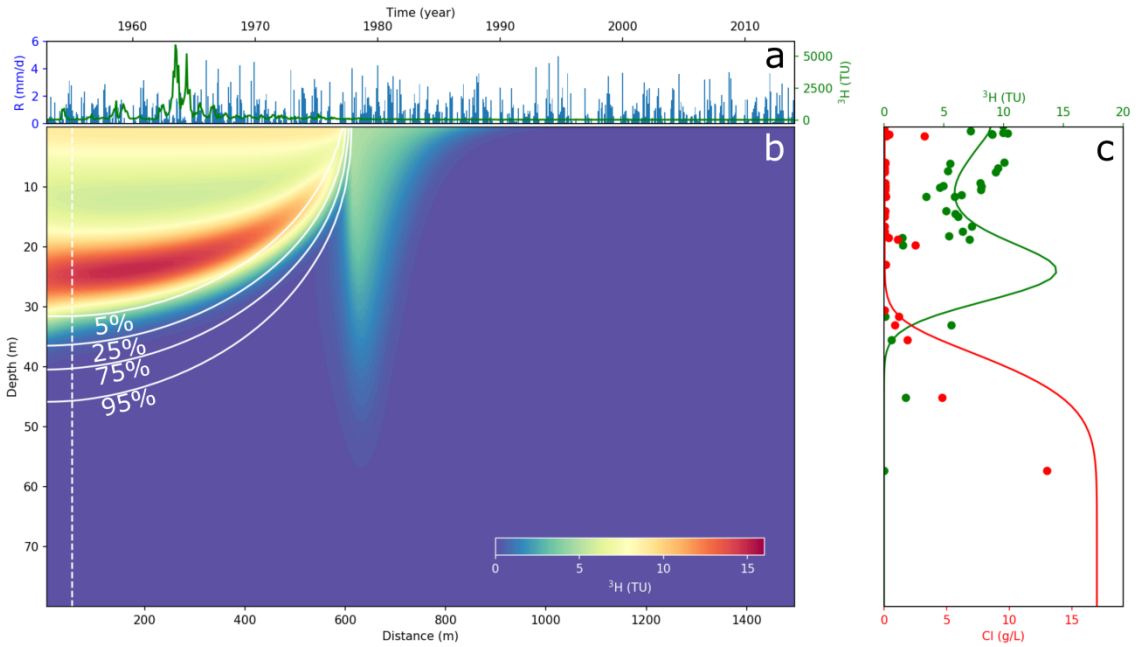

Figure 1. Graph showing the monthly recharge rate (blue) and ^{3}H activities (green) versus time. (b) Modeled contour plots of the ^{3}H activity (colours) and percentage of seawater (white contour lines). (c) Graph showing the ^{3}H activity (green) and chloride concentrations (red) as a function of depth along the white stippled line in (b). Lines indicate model results, data points represent samples from various locations across the lens from *Houben et al. (2014)*.

Numerical simulations were conducted to explore the possibility that the tritium in the transition zone is a remnant from the atmospheric bomb peak. Groundwater recharge was calculated based on rainfall and evaporation data and was varied using monthly time steps as described in Post & Houben (2017). A two-dimensional section across the freshwater lens was considered. The transient simulation spanned the 61 year period from 1 January 1953 (the earliest year for which the atmospheric tritium input could be estimated) to 31 December 2013. The meteoric water recharging the freshwater lens was assigned a ^{3}H activity based on measured activities of nearby rainfall stations of the global network of isotopes in precipitation (GNIP) network (IAEA/WMO, 2016). To generate a continuous input time series, the partial records of the German stations in Stuttgart, Braunschweig and Cuxhaven as well as the Dutch station in Groningen were collated. For the period prior to 1961 the data from Ottawa, Canada were substituted. The overall trends measured at the different stations are similar, but there can be appreciable differences in the ^{3}H activity values for any given month. Despite the uncertainty thus introduced, this approach was considered to be the most appropriate to generate the longest-possible input time series required for the model simulations.

The initial conditions for the model were determined by simulating the same 61 year period with all model parameters unchanged except for the ^{3}H activity of the meteoric recharge, which rather than being variable was assigned a constant value of 5 TU. The latter simulation also served to provide the initial concentrations for groundwater salinity.

Preliminary simulations show that it is indeed possible for atmospheric tritium to migrate to the bottom of the circa 35 m thick freshwater lens and mix with seawater within the duration

of the simulation period (Figure 1b). The tritium bomb peak itself is still located within the freshwater part of the lens, but some tritium has mixed with the brackish water in the transition zone. The model under-estimates the thickness of the lens to some degree (Figure 1c), which is presumably due to the adopted recharge rates being too low. Nevertheless, the general trend of the ^{3}H activity with depth across the lens as well as the elevated ^{3}H activity at the upper limit of the transition zone displayed by the data points are also visible in the model simulations (Figure 1c).

Great uncertainty exists though about the effect of abstraction, which has not yet been considered, because of the two-dimensional nature of the model, yet it is believed to be an important factor on Langeoog. It is therefore not possible to draw site-specific conclusions for the island, but the model simulations are general proof-of-concept that tritium in the transition zone can be used to study mixing between fresh- and saltwater in coastal aquifers. This means that (as long as the bomb-peak can be detected) tritium may be used to infer timescales of mixing, but potentially also to better constrain the dispersivity in numerical models of coastal aquifers.

The model simulations also showed the development of a zone of elevated ^{3}H activities below the seafloor, which protrudes much deeper than the base of the freshwater lens. This is attributed to the intrusion of seawater, which is driven by the loss of solutes from the system, as dispersive mixing causes salt to be sequestered into the seaward directed groundwater flow that discharges near the coastline. The shape of this tritium plume shows that the intrusion can have a strong vertical component and thus that the determination of seawater intrusion velocities based on tritium activities measured along a shore-perpendicular transect needs to be done with great care. Interpretations that do not take this vertical motion into account could yield erroneous results. To our knowledge, field evidence of this deep zone has not been documented so far.

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