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## Reply on CC1

Michael Kilgour Stewart et al.

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Author comment on "Comment on "A comparison of catchment travel times and storage deduced from deuterium and tritium tracers using StorAge Selection functions" by Rodriguez et al. (2021)" by Michael Kilgour Stewart et al., Hydrol. Earth Syst. Sci. Discuss., <https://doi.org/10.5194/hess-2021-146-AC2>, 2021

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Reply to Nicolas Rodriguez (CC1)

CC1: We thank Stewart et al. for engaging in a discussion on our contribution. We also thank Francesc Gallart for contributing to this discussion. Here, we take the opportunity to clarify and re-iterate on several key points.

Authors: We thank Nicolas Rodriguez for constructive evaluation of our work.

CC1: While we agree with Stewart et al. that tritium is an extremely useful tracer in catchment hydrology, with a high information content (in the case of Rodriguez et al. 2021, higher than stable isotopes), we believe that the reasoning put forward in the comment is flawed. Indeed, in the comment, catchments (as highly dynamic systems) are treated as systems at steady state. Thus, the finding of different travel times can simply be an artifact of the mathematical approach chosen by Stewart et al. because the latter is nowadays known to be a hindrance to deriving more realistic travel times.

Authors: We agree that the assumption of steady state sometimes used with tritium measurements is not ideal for catchment systems. However, we think that even if the two isotopes were treated in the same way experimentally and mathematically (as done by Rodriguez et al., 2021), we would still expect different travel time distributions if the stable isotope input varied seasonally, and the tritium input had both seasonal and radioactive decay variations provided there was enough old water in the stream to reveal the difference. There is no a priori reason to necessarily assume steady state conditions (at least in the SH) because tritium is radioactive. If there are radioactive variations then tritium measurements at various flows allows identification of non-steady state flow conditions.

CC1: We would like to clarify on the claim "that no significant old water (beyond the range that can be resolved by stable isotopes) was identified by  $^3\text{H}$  in the Weierbach Catchment stream does not mean that such old water does not exist in other catchments". We agree that old water contributions exist in other catchments, and that those can be older than the ones observed in the Weierbach. However, we disagree with the notion that different

travel times are identified with both tracers, as this notion lacks any physical process-based explanation, and it can most likely be explained by limitations in the previously applied age-dating models for stable isotopes and tritium.

Authors: This is a restatement of the argument above. We do feel that there is a physical explanation (namely radioactive decay), although this tends to be masked at present by remnant bomb tritium with the Trier tritium input function (as explained in the Comment).

CC1: With increasing mean/median travel times and contributions of old water in streamflow, we can surely assume that the identifiability of model parameters solely based on stable isotopes becomes increasingly difficult.

Authors: Agreed.

CC1: However, the travel time of a water parcel physically has only one median age, and the parameters calibrated with stable isotopes can still suggest the presence of old water even if they are not clearly identifiable due to increased uncertainties.

Authors: This argument is not clear. Streamflow measurements involve mixtures of many water parcels and therefore have age distributions. Mixture of waters with different ages leads to attenuation of input patterns which are the basis of the stable isotope age determination method. This attenuation makes old water less visible to the method. The same could be said for tritium if the effects of radioactive decay are counteracted by remnant bomb tritium (in the NH). If not, the decrease with time due to radioactive decay should make old water more visible to tritium.

CC1: Furthermore, in the framework of time-varying travel times derived through SAS functions, the discussion on the differences between SH and NH tritium concentrations in precipitation is not completely relevant. We did not need the current tritium values in the stream to be very different from the water that was recharged decades ago. As explained in the discussion section of Rodriguez et al. (2021), we did not only rely on tritium radioactive decay to age-date water. The method we used can accommodate any tritium input signal. The tritium input signal will, however, affect how much information on water ages can be extracted with the method (we showed how this information can be quantified). We simply accounted for radioactive decay, and the decay likely allows identifying model parameters more precisely compared to stable isotopes for catchments with longer travel times and larger contributions of old water.

Authors: In fact, as shown in the comment there is at present little radioactive decay component of the tritium input variation at Weierbach Catchment because it is masked by lingering bomb tritium, there is only the seasonal variation matching the seasonal variation in the stable isotope input record. So the tritium and stable isotopes would be expected to give similar travel times. This issue of lingering bomb tritium makes it difficult at present to test the truncation hypothesis generally with the Trier tritium input record.

CC1: That being said, it would be interesting to evaluate whether the information content in the tritium input is higher in the SH compared to the NH. This can be tested in a more robust way than in this comment, and we invite Stewart et al. to apply our framework to test whether their suggestion holds against data.

Authors: We agree that this would be a valuable thing to do. However, this is beyond the scope of this comment and we do not currently have suitable SH datasets.

CC1: We do not support the recommendation in the comment by Stewart et al. to keep sampling tritium sparsely over longer periods. This will very likely bias the tritium data towards hydrological recessions, which by definition will more likely contain older water.

Also, this will most likely entirely miss the short-lasting events associated with younger water. We want to re-iterate here that findings from early work with tritium showed the potential of tritium for revealing young water contributions. These studies tend to be overlooked. Contrary to the suggestion in this comment, we encouraged sampling tritium across the full range of flow stages in catchments to avoid this potential issue.

Authors: Our position here is that we think long-term tritium sampling should be encouraged (as we recommended in the Comment) to take advantage of the changing situation with regard to remnant bomb tritium and radioactive decay of tritium, but in addition sampling should encompass the full range of streamflow rates of interest, and sampling of tritium should match that of stable isotopes as much as possible for comparison of tritium and stable isotope estimates of travel times via the SAS method. We agree that sampling tritium over high-flow events is informative and note that several papers have done that (Morgenstern et al., 2010; Cartwright & Morgenstern, 2018; Hofmann et al., 2018).

CC1: We perceive some circular reasoning in the submitted comment, which is problematic. In the comment, the TTD is assumed, to deduce what the tracer signal in the stream should be, to deduce that the tracer should not allow for discriminating young and old water in cases where the TTD is exactly as assumed (this point is not emphasized enough in the comment).

Authors: The point of the calculation in Fig. 1 of the Comment using the assumed TTD is not to represent any particular system, but to show approximately the effects of the lingering bomb tritium and the mixing of different-age waters on tritium concentrations in stream water. The criticism appears to be that our approach is not based on real data from a real system, but from an admittedly flawed simple model. However, any real system data would only represent one example from a large number of possibilities so it would be no more valid than any other example.

CC1: The real question is: is the assumed TTD realistic and accurate (especially bearing in mind the steady-state assumption)? A completely different TTD model (for example, multimodal, with both young and old water) could yield a very different perspective. More importantly, this reasoning is based on a steady-state assumption, while many more situations are possible in unsteady conditions. Virtually anything is possible in unsteady conditions, while the steady-state assumption is extremely constraining for deriving general conclusions on the link between TTDs and tracers.

Authors: There is nothing in the literature to show that a multimodal TTD model would allow better identification of old water by a seasonally-varying isotope (either stable isotopes or tritium affected by remnant bomb tritium) than a single modal model. The degree of mixing (e.g. the factor  $f$  for the EPM model) is the important factor for smoothing of seasonal variations.

CC1: The presented comparison between 2H and 3H in the comment is based on different sampling strategies, which by design target different portions of the TTD. It is not so surprising that the MTTs differ, especially with a steady-state approach. Travel times are highly dynamic, even if inferred from tritium only, and the same discharge can be associated with vastly different median travel times (for instance, see figure 9 in Rodriguez et al., 2018). We thus argued that a fair comparison between the tracers needs to use tracer data sets that are as close to each other as possible, in a consistent time-varying mathematical framework.

Authors: We agree that a fair comparison between the tracers needs to use tracer data sets that are as similar to each other as possible in a consistent time-varying mathematical framework. We believe that if this is carried out on a variety of catchments

it will show that tritium can identify old water components much more effectively than stable isotopes provided radioactive decay can be used for dating (as in the SH).

CC1: Other comments:

- L45-47: This “range” is a strong a priori assumption based on several limitations, and it is precisely what we questioned in our work.

Authors: We will change the wording here to a less prescriptive “young water”.

CC1: 2. L48-54: We already discussed about storage S in the Weierbach catchment (section 4.4.1 in our paper). Estimating MTT from  $S/Q$  with Q the catchment runoff is wrong. The total flux through the catchment needs to be used instead. Moreover, this method too is valid only in steady-state conditions.

Authors: We agree, but this is meant as a ball-park figure not an accurate estimate.

CC1: To conclude, recent work suggests that there is no absolute truncation issue. The perceived “truncation” may rather have resulted from what was a too restrictive conceptualization of tritium-based TTD estimations, as also suggested by recent progress in travel time research.

We fully agree with the authors of this comment on the importance of using tritium for travel time analyses, but we disagree on the fact that keeping this long-standing perception that tritium can, by default, show us this “invisible” old water will be helpful. This old water is invisible only if we choose to make it mathematically or numerically inexistent (of course there may be additional challenges of equifinality when working with a single tracer). We think that it is time to challenge our long-standing assumptions, to give up our limiting strong a priori assumptions, and to embrace the possibilities offered by the new theoretical frameworks and by the new sampling and measurement techniques. We would like to invite Stewart et al. to apply our proposed mathematical framework allowing for time variance and multimodality in TTDs to their available datasets for testing and quantifying their proposed claims. Our code is accessible online:

[https://git.list.lu/catchment-eco-hydro/composite\\_sas\\_model\\_2h\\_3h\\_weierbach](https://git.list.lu/catchment-eco-hydro/composite_sas_model_2h_3h_weierbach)

Authors: We agree that further comparisons of stable isotope and tritium measurements on catchments (analysed by the same methods) can only be helpful to reach a fuller understanding of travel times in catchments.

Morgenstern, U., Stewart M. K., Stenger, R. 2010: Dating of streamwater using tritium in a post nuclear bomb pulse world: continuous variation of mean transit time with streamflow. *Hydrology and Earth System Sciences* 14, 2289-2301.

Rodriguez, N. B., McGuire, K. J., & Klaus, J. (2018). Time-varying storage–Water age relationships in a catchment with a Mediterranean climate. *Water Resources Research*, 54, 3988– 4008. <https://doi.org/10.1029/2017WR021964>