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Reply to Anonymous Referee #2

by M.A Gusyev, D. Abrams, M.W. Toews, U. Morgenstern and M.K. Stewart.

General Comments:
This paper presents simulated tritium concentrations in river waters in five watersheds within the Lake Taupo catchment, New Zealand. Findings include differences in the shape of transit-time cumulative distribution functions for watersheds with partially penetrating streams and watersheds with streams that preclude underflow. In addition, the paper demonstrates that the relative magnitude of tritium among watersheds is not simply a function of mean transit time combined with constant tritium decay (p. 3093/lines 9-12) and that the relative magnitude of tritium among watersheds can change over time–one of the more novel contributions of the paper. The paper also presents a comparison of simulated transit time distributions generated by using particle tracking (MODPATH) and by using a solute transport model (MT3DMS). This methods comparison makes up the bulk of the abstract and conclusions but is not reflected in the title of the paper. It should be. Overall, there exists a bit of a disconnect between the title, abstract, and conclusions (and literature cited), which makes it difficult to sort out what new contribution to the literature is intended by the authors. Perhaps the title should be " 'Simulation of' tritium concentrations in river waters . . ." if the focus is to remain on the simulation methodology. On the other hand, perhaps the abstract and conclusion should contain simulated tritium results, such as those mentioned above, if the title is to remain 'as is'.
Nonetheless, once the different sections of the paper are tightened up and the authors clarify the main objective of the paper, publication is warranted as the paper contains interesting results.
Response: We thank the Anonymous Referee for the constructive comments and agree that the title needs to be adjusted. We followed the Referee's suggestions and propose an updated title of the revised manuscript:
"Simulation of tritium concentrations and groundwater transit times in river waters of the western Lake Taupo catchment: A comparison of particle tracking MODPATH and solute transport MT3DMS methods."

Specific Comments:

3084. The utility of simulated transit time distributions (CDFs, PDFs) should be apparent in the abstract given that most of the abstract focuses on how best to obtain one. For example, transit time distributions are necessary to understand tracer and contaminant output functions at discharge points.

Response: We agree with the Referee's comment and added the suggested text after "outflows" on page 3084 line 12: "These TTDs are necessary to understand the tracer and contaminant spatially-distributed source and output functions at river discharge points."

3085/20-23. It's not clear how this reference fits in as the manuscript is currently written. It's the Eberts et al. comparison that is relevant. What about, "For example, Eberts et al. (2012) compared simulated tracer concentrations computed using TTDs from particle tracking and LPMs for wells in four aquifer systems by using the Excel workbook TracerLPM (Jurgens et al. 2012), which can use either TTDs from MODPATH or LPMs as input to the convolution integral for obtaining tracer concentrations at wells. They found that . . ."

Response: We thank the Referee for this comment and modified the text as suggested: "For example, Eberts et al. (2012) compared simulated tracer concentrations computed using TTDs from particle tracking and LPMs for wells in four aquifer systems by using two Excel workbooks: an updated TRACERMODEL1, which evaluates a numerical non-parametric version of the convolution integral, and TracerLPM (Jurgens et al. 2012), which can use either TTDs from MODPATH or LPMs as input to the convolution integral for obtaining tracer concentrations at wells. They confirmed that similar tracer time-series concentrations in wells can be simulated by both DPMs and appropriately selected LPMs, but only particle-tracking DPMs identify spatially variable sources of tracers and contaminants in those wells (Eberts et al. 2012)."

3085/1. Tracer-based ages rather than tracer concentrations were simulated in many of the examples from the literature that is cited here. Thus, it is not strictly correct to state, ". . . have also been used to simulate isotope tracers at wells . . .". In other words, isotope tracers were not necessarily simulated for each of the cited works; rather, groundwater ages were frequently simulated. Consider something like . . ."have also been used to simulate isotope tracer concentrations and (or) tracer-based groundwater ages."

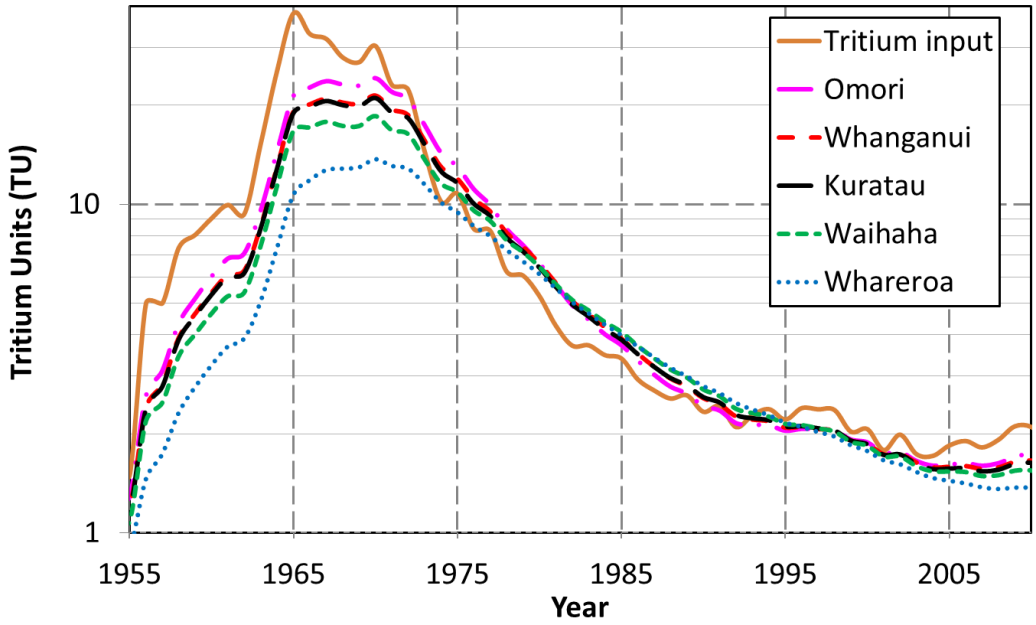
Response: We thank the Referee for the constructive comment and adjusted the text accordingly: "*have also been used to simulate isotope tracer concentrations and (or) tracer-based groundwater ages."*

3091/14. What "adjustment outlined in Abrams (2013)"? Is this the adjustment described in 3089/6-14? Can the adjustment be succinctly described here?
Response: Yes, this is a description of the adjustment by Abrams (2013). We replace the text for clarity by: "adjustment values estimated earlier to correct for the stream width in MODPATH, see Abrams (2013) for more details."

3092/18-25. It seems likely that the narrower range of travel times associated with the Omori catchment compared with the other catchments contributes to its relatively high peak concentration. In other words, the relatively high peak concentration for the Omori catchment may not simply be related to the relatively small amount of time that the tritium stays in the aquifer but also to the relatively small amount of mixing of waters with different tritium concentrations in the Omori, resulting in concentrations that are more similar to recharge concentrations compared with the other catchments.
Similarly, it seems likely that the relatively gradual decrease in tritium concentrations from 1970-1990 (fig. 3b) in the Whareroa catchment is, in part, the result of the wide range of travel times reflected in the baseflow, resulting in proportionately longer flush times.
Response: We thank the Referee for highlighting this fact and introduced an additional explanation in the text on page 3092 line 25: "However, it also appears that a narrower range of transit times is indicated by the small MTT and contributes to the higher peak of the output tritium concentrations in river waters. For example, the smallest MTT and a smallest range of transit times, as evident by the CFD (see Figure 2), result in higher tritium concentrations in the Omori catchment compared with the other four catchments. Similarly, the largest MTT of the Whareroa catchment is, in part, the result of the wide range of travel times reflected in the baseflow, resulting in proportionately longer flush times and the relatively gradual increase in tritium concentrations due to large amount of long transit time waters, which were recharged by pre-bomb tritium concentrations and had more tritium decay in the subsurface"

3092/26. Figure 3b does not show tritium in rain. Consequently, the reference to the figure at the end of this sentence does not seem appropriate.
Response: We combined Figure 4a and Figure 3a-c into one figure, which allowed us to compare tritium concentrations in the rain and river flows of five river basins, see the image below. The new Figure 3 indicates that the old groundwater can still have higher tritium concentrations than tritium concentrations in rain from the 1980s. This also indicates that extremely high tritium analytical

accuracy is needed and that normal accuracy is not sufficient. The log y-axis was suggested by Referee #1.



3093/7-13. This is an interesting result.
Response: We thank the Referee for this comment.

3098/16. The first sentence of the Concluding Remarks states, ". . . we presented an approach to calibrate the steady-state MODFLOW/MODPATH model to measured tritium concentrations in rivers waters at baseflows . . ." implies that the approach used in this study is new; however, the use of travel time distributions and the convolution integral for computing time-dependent tritium concentrations at discharge points is not new. (An example is shown in the reference by Jurgens et al. 2012.) Furthermore, much of the conclusions are focused on how to generate tritium concentrations with MODPATH transit time distributions. Again, such work 'in and of itself' is not new. On the other hand, the comparison of MODPATH and MT3DMS based transit time distributions does appear to be new, as do findings related to relations between simulated tritium concentrations among watersheds within a larger catchment.
Response: We consider our work "new" because the simulation of tritium concentrations in river waters with the particle-tracking MODPATH has not been reported in the peer-reviewed literature. We recognize that the particle tracking MODPATH with convolution integral has been frequently used to simulate tritium in wells. From the Referee's example, Jurgens et al. (2012) simulated tritium concentrations in *abstraction wells* with the use of LPMs and did not attempt

simulations in river waters. We also want to emphasize that other tracers such as nitrates have been simulated in rivers with the use of particle-tracking MODPATH, but the tritium concentrations in rivers have not. For example, Kauffman et al. (2001) obtained *nitrate concentrations in rivers* using particle tracking MODPATH with the synthetic nitrate input curves obtained from land use types. This is a particularly important distinction because of the inherent uncertainty in nitrate inputs to an aquifer that is not present with tritium. Therefore, we decided to stress this point by adding statements in the revised manuscript as well as adding a paragraph to emphasize the significance of this study:

on p 3084 line 1: adding a statement "The purpose of this study is to simulate tritium concentrations and groundwater transit times in river waters with particle tracking, and to compare the particle tracking results with solute transport results."

on p 3086 line 27: adding "with particle tracking MODPATH" after "the WLTC"

on p 3086 line 28: adding "and tritium concentrations" after "comparing"

on p 3100 line 16: adding one paragraph and one reference

"As a final remark, we would like to highlight the importance of tritium measurements in river waters for the calibration of particle tracking and solute transport models. For the model calibration, tritium has less uncertainty than other tracers because of its known input concentrations measured in precipitation, its chemical inertness in the subsurface, and its unresponsiveness to air exchange processes in river waters. In precipitation, tritium concentrations are independent of air temperature and pressure, and measured monthly at many ground stations of the Global Network of Isotopes in Precipitation established by the International Atomic Energy Agency (IAEA/WMO, 2014). For the subsurface processes, tritium, being a part of the water molecule, does not react with aquifer matrix or dissolved chemicals in groundwater. In river waters, the tritium concentrations discharged with groundwater remain unaffected by evaporation and air moisture as surface water bodies. Because of its radioactive decay, tritium concentrations in groundwater are still dependent on the travel times despite constant tritium inputs over recent decades in the Southern Hemisphere and can be determined using high detection accuracy of 0.02-0.03 TU. From now on, the tritium time series measurements in river waters of the Northern Hemisphere are essential for the model calibration especially after the tritium concentrations in precipitation return to pre-bomb levels in the next decades."

Reference:

IAEA/WMO. Global Network of Isotopes in Precipitation. The GNIP Database, 2014. Accessible at: http://www.iaea.org/water

Technical Corrections:
3084/26. Haitjema, 1995 is missing from the References, and Kaufman et al. 2001 . . . "Kauffman" should be spelled with two 'f's (here and in the References)
Response: We added the missing reference and corrected the spelling as suggested by the Referee.

3085/16. Something is grammatically incorrect . . ."MODPATH/MODFLOW allows one to evaluate age groundwater tracer directly . . ."
Response: We changed the wording of the sentence by "models allow".

3086/8. "groundwater flow MODFLOW" . . . groundwater flow is not necessary before MODFLOW
Response: We replaced the text as suggested by the Referee.

3086/15. The word "a" should not precede "groundwater flow models"
Response: We removed "a" as suggested by the Referee.

3087/20. SWS, 2012 is missing from References
Response: This is a typo and should be SWS (2010). We corrected the text as suggested by the Referee.

3088/18. SWS, 2013 is missing from References
Response: This is a typo and should be SWS (2010). We corrected the text as suggested by the Referee.

3090/8. Should have a reference for tritium decay rate
Response: We added the tritium decay and a reference Morgenstern and Taylor (2009), see our reply to the Referee #1.

3092/28. "tritium decay of 12.32 year" . . . should be "tritium half-life of 12.32 years"
Response: We changed the text as suggested by the Referee.