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Interactive comment on "Simple estimation of fastest preferential contaminant travel times in the unsaturated zone: application to Rainier Mesa and Shoshone Mountain, Nevada" *by* B. A. Ebel and J. R. Nimmo

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Interactive comment on "Simple estimation of fastest preferential contaminant travel times in the unsaturated zone: application to Rainier Mesa and Shoshone Mountain, Nevada"

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Anonymous Referee 1

Full Screen / Esc
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Interactive Discussion
Discussion Paper

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Author responses are in **Bold text**

General comments This paper, which is well written and organized, aims at estimating the fastest travel time for preferential transport of solutes through the vadose zone. Generally, I would regard this topic to be of high relevance and potential interest for the readers of HESS The study focuses on the downward transport of radionuclides through thick unsaturated zones to the aquifer at two areas within the Nevada Test Site (NTS), where several underground nuclear tests had been executed. Towards their goal, the authors first develop conceptual flow models for the two sites by reviewing and interpreting existing literature sources on lithology and subsurface structures. Then, they present and adopt a simple approach for quantitatively estimating the time required for transport from potential sources to the groundwater at these sites, in which continuous and intermittent supply of the preferential flow systems are differentiated. This approach, which is termed “Source-Responsive Preferential Flow model” (SRPF) in the present paper, had already been proposed in an earlier publication in WRR by one of the authors (Nimmo, 2007). However, several aspects of this methodology and its application to the two test sites still are at least worth discussing. I would agree with the authors that having a “simple model with minimal site characterisation” to predict contaminant travel times would be highly desirable, but in my opinion the methodology of Nimmo (2007) is questionable in some aspects.

Why should there be a central tendency for preferential flow velocities, such that a global “mean maximum velocity” can be defined? Why should a “universal effective rate” be globally valid?

Apart from the empirical evidence in the graphs and tables of (Nimmo, 2007), there are fundamental physical reasons to expect the maximum velocity of transport Vmax to vary little among media, especially in comparison to properties like hydraulic conductivity (K). Several of these are pointed out and discussed in

Section 4.2 of (Nimmo, 2007). Some additional ones are: (1) Vo represents a travel time, so it is not dependent on the abundance of pores of any particular size in a particular medium, as would be important for a prediction of flux. (2) It represents a maximum, and thus is not sensitive to all flow paths in the medium, but only one, the fastest. (3) Fastest flow tends to be gravity-driven, not capillarity-driven, and gravity is a constant in these problems. An analogy can be drawn to free-fall in air or water, the terminal velocity reached is approximately equal for a set of similar objects; in preferential flow liquid water is the object in question, which means differences may be minimal from case to case. For K, on the other hand, pore-size distribution, tortuosity, degree of saturation of the bulk medium, and other factors cause it to vary drastically from one medium to the next. Because these factors play a much smaller role when one confines one's objective to Vmax, it is reasonable to expect Vmax to vary much less among media than does K. An abbreviated version of our responses have been added to section 5.1 in the revised manuscript. This expected lesser degree of variation is obviously carried to an extreme with the assumption that Vmax is a constant, rather than a variable that varies modestly among diverse media. As we explained in the manuscript, 3907/12ff, the source-responsive model has been developed through a data-driven, or downward approach (Sivapalan et al., 2003). The process starts with what is known from direct observations, develops a simple underlying principle or generalization for those data, and then is refined and adjusted to produce a more accurate and versatile model. Though clearly untrue except in an approximate sense, a single value of Vo is adopted in the present early-stage development of the source-responsive model. With minimal complexity it is easier to discern what features of the model are working well, so that they can be retained and improved upon. It also serves to limit the degrees of freedom appropriately for application. This limitation helps to avoid creation of a many-parameter model that can fit essentially any data set just because of its many degrees of freedom. Later adjustments concerning the value

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Interactive Discussion

Discussion Paper

or universality of Vo, when additional data and information become available to support them, are desirable.

It can also be pointed out that the practice of lumping together a broad range of media for representation with a single parameter set is very common in unsaturated flow investigations. A prominent example is the widely used scheme of Carsel and Parrish (1988) that designates 12 sets of unsaturated hydraulic properties to represent all soils. All silt loam soils, for example, are lumped into a single box for the full suite of hysteretic unsaturated hydraulic properties.

The price paid in using a model with constant Vo is that it necessitates an understanding that the uncertainty of predicted results will be high, about an order of magnitude. In most quantitative scientific fields this would make it useless, but in unsaturated-zone hydrology, where complexities abound and hard data are scarce, this degree of uncertainty is competitive with many other approaches in current use. In the SRPF model, the universal effective rate, i0, relates to an input flux threshold for the generation of preferential flow. In a physical sense, this threshold rate may be a minimum flux required to begin and sustain gravity-driven film flow. That rate may depend on macropore wall roughness or aperture, but the ability of a simple approximation such as i0 to reasonably capture the timescales of this behavior by a pulsed-flow formulation suggests that there may be minimal variability in such a threshold rate, despite large differences in macropore characteristics and porous media, supporting the use of a global rate.

Can these “constants” be seen as variables that can be linked to some site-specific properties? How is the broad inter-site variability of subsurface materials and structures taken into account?

One of the main points of the Nimmo (2007) paper was that there was far less variability in first arrival times of solute via preferential flow than one would ex-

pect based on drastic site differences in unsaturated hydraulic properties and porous media type (soil and lithology). This was the fundamental realization of the Nimmo (2007) study, that temporal supply of water to preferential paths was more important than site properties for determining first arrival time. In a sense, this "Source-Responsive" paradigm can be viewed as a large sensitivity to boundary conditions rather than constitutive relationships. The approximation is then used to emphasize continuous vs. intermittent water supply and ignore the specific subsurface materials and structures, beyond basic analysis to determine if the subsurface presents conditions conducive to preferential flow. Certainly it is possible, especially if more travel-time measurements at more sites become available, to modify the Nimmo (2007) model to make use of site-specific properties to adjust the values of model parameters. Doing so would result in a more complex model producing results likely to be of less uncertainty. At many locations, including Rainier mesa, little is known about the properties of the unsaturated zone, so there is value in a simple model that does not require such property data and that can provide bounding values on the range of travel times that can be expected.

The authors of the present study adopt the approach as it stands, without providing critical discussion. Above all, they neither provide a validation of the SRPF models for the two study sites, nor any other discussion of the plausibility of their results, e.g. through comparison with results from other places of the NTS.

The revised manuscript adds (in the Discussion section) comparisons of the observed and predicted first arrival velocities for both similar lithologies (from the NTS and the Apache Leap site, which are tuffs) as well as comparison of observed and predicted first arrival velocities for 48 cases which are independent of the original data used in the development of the Nimmo (2009) model. Both the comparison against tuff lithologies from the NTS and Apache Leap and the new independent test suggest that that the SRPF model estimates are reason-

able and within the previously reported order-of magnitude agreement.

The estimated travel times are thus probably too approximate for delivering meaningful results. On the other hand, the application of the approach at the two sites does not provide new data or insights that would help to further explore the methodology.

We disagree with the reviewer that the estimated travel times are too approximate to be meaningful. In many cases, such as at Rainier Mesa, little unsaturated hydraulic property or fracture geometry information is available, thus making traditional contaminant transport approaches using Richards equation and the advection dispersion equations subject to equifinality and parameter uncertainty. This situation is precisely where the SRPF model may provide useful predictions, especially in the context of model abstraction and model ensemble predictions. The revised manuscript also adds independent testing, which the authors are convinced strengthens the case that SRPF model can provide order-of-magnitude agreement for first arrival times of solute.

The matter of how much uncertainty an estimated value can have and still be meaningful depends on the application the estimate is used for. To say that a travel time will be greater than one month and less than 100 years is a statement that may not be useful for a purpose like predicting aquifer water quality over the span of a decade, but for many purposes the designation of a broad but finite range is very useful. For example if regulations require different monitoring or remedial measures when possible travel times may be less than 500 years, this model result gives a direct answer. Likewise, if the regulatory criterion is 10 days, the model result gives a direct answer with the opposite conclusion.

Therefore, I would not recommend publication of the manuscript in its present form, but encourage the authors to resubmit the paper after major revisions.

We have made major revisions in response to this and other reviews, with the aim of making the manuscript suitable for publication.

Specific comments p. 3882, lines 21-22: “This work expands and clarifies issues from Ebel and Nimmo (2009) while building on that previous work.” It would be helpful to know which issues from that work are “expanded and clarified”. The manuscript seems to be a short version of the cited work of Ebel and Nimmo (2009), which is publicly available as USGS open-file report (http:// pubs.usgs.gov/ of/ 2009/ 1175/ of2009-1175.pdf). The authors could take advantage of this by summarizing the site descriptions in Section 2 and referring the readers to the report for all the details where necessary.

The revised manuscript removes as much of the site detail as is feasible and refers back to the Open-File Report by Ebel and Nimmo (2009) for the reader to find further details.

p. 3885, lines 6-10: Please include the distances to the saturated zone somewhere in the manuscript, for example in Table 1.

The revised manuscript shows the distances from potential radionuclide sources to the saturated zone in a new Figure (Fig. 4).

p. 3887, line 17: If much of the precipitation falls as snow (DeMeo et al., 2006), snowmelt would be a major contribution to groundwater recharge at the NTS. How would you account for the effect of snowmelt in your analysis?

Given that the intermittent-supply travel times are long (i.e. decades), the idea behind the SRPF model is that the total precipitation simply controls the amount of “on” time in the pulsed transport. Over a period of decades, the exact timing of precipitation (or snowmelt) delivery does not matter as far as the model is concerned. While this is clearly an oversimplification of process, it is consistent with the amount of data at the site (i.e. there are no detailed records of spatially-variable snowmelt). For the analysis presented here, apportionment of precipitation between rain and snow does not control estimated first arrival times, and therefore does not need to be included in the analysis.

p. 3890, line 4: “contaminant transport model” Later on in the text, you are using the term “travel time model”, which seems more appropriate. As correctly stated in the introduction, in Section 3.2 and in the conclusions, the SRPF model is not a transport model that could predict concentrations or fluxes.

The reviewer is correct, the SRPF model is solely for travel-time estimation. The revised manuscript eliminates any suggestion that the SRPF model is a contaminant transport model and makes it clear that the model can only be used to provide estimates of travel time for conservative solutes.

p. 3890, lines 19-22, and p. 3891 lines 9-19: If the travel times through the vitric tuff are short (1-6 years for 500 m), how do you come to the conclusion that preferential flow is “very limited” in this unit? The evidence you present is not so “limited” in my view, but clearly shows that preferential flow through fractures or fingers has to be expected for the vitric tuff, at least as a conservative assumption.

The point that the reviewer makes is valid, evidence suggests that preferential flow is indeed possible through the vitric tuff at Rainier Mesa and the conceptual model shown in Fig. 5 of the revised manuscript acknowledges this. However, the majority of previous efforts at Rainier Mesa, and additional efforts through the same Paintbrush tuff lithology at Yucca Mountain, have reached the conclusion that the majority of flow through the vitric tuffs is matrix-dominated and that a very limited amount of preferential flow occurs, most likely along fault systems. At Rainier Mesa, the vast majority of the working points and other potential contaminant sources are stratigraphically below the vitric tuffs, and provided some water is supplied to these potential sources preferentially or at a rate sufficient to initiate preferential flow, that is what is important to the SRPF model. We feel no change to the manuscript is necessary.

p. 3891 lines 19-25: “Philips et al. . . . water itself” Please consider skipping this passage, which in my opinion is not directly relevant for the study.

This section was put into the manuscript in response to a previous peer-review, where the reviewer felt that tritium was not an appropriate tracer of water movement. Our response may have introduced too much detail that is not directly relevant and we have removed much of this material from the revised manuscript.

p. 3892 lines 4-5: "Table 3 shows the large saturated hydraulic conductivity contrast between the zeolitic tuff matrix and the overlying vitric tuff matrix, with the zeolitic tuff potentially acting as a permeability barrier to vertical unsaturated matrix flow." The data in Table 3 do not show a large contrast in hydraulic conductivities between the two formations. The average value would in fact be higher for the vitric tuff, although only two values are given for each formation. Thus, this conclusion is not supported by the data.

Table 3 is confusing as originally presented, the lower saturated hydraulic conductivity value given for the vitric tuff (on the order to 10-10 m s -1) is for the "Grouse Canyon tuff" which is devitrified in some sections and densely welded in other portions. This would cause the saturated hydraulic conductivity to be far lower than for a truly "vitric" tuff, such as the Paintbrush Group. This type of lithologic detail is shown more clearly in the report by Ebel and Nimmo (2009), but not shown in this manuscript. The Grouse Canyon tuff also lies at the transition into the zeolitized tuff, so if there is a transition at this lithologic interface, it does not change the Nimmo (2009) SRPF estimates for the majority of potential radionuclide sources, which are stratigraphically beneath that transition. In the revised manuscript we have added this information to the table footnote and provided further references if the reader needs more information.

p. 3892 lines 20-28: It would be interesting if you could provide some numbers for the age observations.

In the revised manuscript we have added that the fracture water ages are on the order of 1-30 years and direct the reader to Ebel and Nimmo (2009) for further

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

information.

p. 3894, lines 18-19: ". . . the properties can be approximated from the similar lithologic units at Rainier Mesa shown in Table 3." Given the large variability over several orders of magnitude and the small amount of data presented in Table 3, I strongly doubt that the hydraulic properties at Rainier Mesa are characterised with sufficient certainty. Even less confidence can be put in the transfer of these properties to Shoshone Mountain.

The authors agree with the reviewer that very little unsaturated hydraulic property information exists for Rainier Mesa and Shoshone Mountain; it is possible that the level of characterization is insufficient to apply a traditional unsaturated flow (i.e. Richards Equation) and contaminant transport model (Advection-Dispersion Equation) without an overwhelming amount of uncertainty. It is the authors' opinion that this is exactly the type of situation where the lack of dependence on unsaturated hydraulic properties represents a strength of the SRPF approach. We are using the minimal hydraulic property information provided in Table 3 to establish a simple conceptual model that establishes the possibility of preferential flow, but the SRPF simulation approach does not require such data to estimate travel times. In the absence of any additional information at Shoshone Mountain, there is no alternative to assuming that hydraulic property data from Rainier Mesa are similar. It does not seem like a reach to assume that when the lithology is similar at the two sites then there is likely similarity between the active flow and contaminant transport processes. No change to the manuscript is necessary, in our opinion.

p. 3894, line 24: Where is the "upper carbonate aquifer" (or the lower, respectively) at Rainier Mesa? Is it the perched aquifer mentioned earlier in the text?

In the revised manuscript it is stated that the upper carbonate aquifer is the carbonate rock at Rainier Mesa shown in Fig. 5 and the lower carbonate aquifer

is the carbonate rock at Shoshone Mountain shown in Fig. 5.

p. 3895, lines 3-5: ". . . (ii) stable matrix flow dominates in the vitric tuff and siliceous rock, with a very small probability of preferential flow in these units." The probability for preferential flow in the vitric tuff unit does not appear to be so small, see comments on pages 3890-3892 above. Given the evidence presented in Sections 2.7.2 and 2.9.2, it rather appears that preferential flow cannot be precluded for any of the formations at NTS, except for the siliceous rock at Shoshone Mountain.

There is no evidence for preferential flow through the vitric tuffs at Shoshone Mountain, especially considering the smaller estimated recharge amounts there relative to Rainier Mesa. We are hesitant to say that macropore flow happens there, although the conceptual model in Figure 5 suggests that preferential flow is possible through major fault or fracture systems. We agree with the reviewer that the siliceous rock is likely the only formation for which preferential flow can be precluded. As at Rainier Mesa, the working points at Shoshone Mountain are stratigraphically beneath the vitric tuffs, so as long as any preferential flow or water supply at a rate sufficient to initiate preferential flow occurs through the vitric tuffs, then the SRPF estimates are acceptable as originally completed.

p. 3897, lines 23-25: "The spread about the 1:1 line of simulated versus measured fastest Vmax illustrates the approximate factor-of-ten accuracy of the Nimmo (2007) approach, and indicates definite uncertainty bounds." Please explain in more detail what you mean exactly by "approximate factor-of-ten accuracy", i.e. that 85

We agree with the reviewer and are now using "order-of-magnitude agreement" in the revised manuscript to describe the degree of model agreement with observed data.

p. 3899, lines 12-13: The estimated value for V0 seems absolutely crucial for the results of the SRPF approach. Why did you choose to take the geometric mean of the measured Vmax? As the objective is to estimate the fastest (worst case) travel

Full Screen / Esc

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Interactive Discussion

Discussion Paper

time of a solute, would it not be more appropriate to take the maximum of observed velocities? Of the values given in Nimmo (2007), this would be 480 m d−1, which were observed for a very similar lithology, namely fractured welded tuff at Yucca Mountain, located some 50 km to the southwest. Prediction with the SRPF approach for the case of continuous supply assuming 13 m d−1 would drastically underestimate this value and that by a factor of 36.

The term "maximum" unfortunately leads to some confusion. It is intended as referring to the maximum transport speed to be expected, among the many possible transport speeds and pathways possible in an unsaturated medium. Nimmo (2007) evaluated numerous specific cases where this maximum transport speed could be estimated independently from direct observations. The resulting values (for those cases where water had been continuously supplied) fell relatively close together, though naturally were not all equal. To the degree that it can be generalized that a single value Vo characterizes all media, it is best to consider the set of values from the different experiments as independent approximations of what that value should be. Thus to choose a best value for Vo, it is appropriate to take an average of the many different approximations of Vo. The geometric mean was used because the data approximated a lognormal distribution. Additionally, as stated in response to the comments by the second reviewer, we feel that using the maximum rate observed out of all the data could cause the model to be significantly wrong if an incorrect observed travel time was included in the study. For example, one accidentally contaminated sample by human error during field work could result in an extremely fast travel time, and if this travel time was the sole value used, thus cancelling out all other observed travel times, it could highly (and incorrectly) bias the travel time model. Using some averaging of first arrival times helps minimize the potential for such an error to bias the model by averaging across many data sets. We feel that the averaging approach makes the SRPF model more applicable to a broad variety of sites. In the revised manuscript we use the term "first arrival velocity" rather than maximum velocity,

which we feel is clearer and more appropriate.

p. 3900. lines 10-12: “To apply the model in such cases, a universal effective rate, i0 [L T−1], is hypothesized.” Is there any physical interpretation of i0 that would allow assessing its value with independent observations? Or is it just a fitting parameter to find some possible mathematical description of the observations by curve-fitting?

A physical interpretation of io that would allow its independent measurement, as the reviewer suggests, is highly desirable and definitely worth developmental effort. We have not yet done such development and are still reliant on the empirical evaluation from travel-time data of (Nimmo, 2007). Physically, the io parameter is the effective water input rate during the “on” pulses of hypothetical pulsed input rendered as the equivalent of a time-varying actual input rate. This is an important research topic in itself, and one which may lead to better understanding and quantification of io. At present, it could be viewed as a “curve fitting” parameter as the reviewer suggests. This is part of the “downward” or data-driven approach where process models are built using only processes supported by the available parameterization and evaluation data.

p. 3908, lines 1-2: “. . . demonstrates its role in the suite of models used to ask and answer difficult questions . . . ”The authors do not provide enough evidence (validation data, comparison to similar sites) that the SRPF approach can give plausible answers to the question of fastest travel times at the two test sites.

In the revised manuscript, we provide comparisons of simulated and observed VFirst Arrival for similar lithologies to Rainier Mesa and Shoshone Mountain as well as further SRPF model testing with an independent data set. While this data set is not comprehensive, it provides additional confidence in the approximate order-of-magnitude agreement between observed and simulated VFirst Arrival.

p. 3908, lines 13-14: “. . . these data do not rule out preferential radionuclide transport through the unsaturated zone at the timescales estimated in this study.” The reverse is also true, because this also means that there is no evidence for preferential radionuclide transport through the unsaturated zone. It seems quite astonishing that there is nothing to be found in the aquifer below the U12e and U12n tunnel working points, for which the authors show that significant exposure to radionuclides exists since several decades, argue that continuous water supply is given and the fastest travel time is estimated to be around one month (more exactly, somewhere between 3 and 300 days). In summary, there is no validation of the travel time estimates presented in this study; the evidence presented in Section 5.2 rather leads to the conclusion that the travel time estimates are far from plausible.

We acknowledge that preferential flow from potential sources to the carbonate rock has not been confirmed, but we disagree that the radionuclide monitoring has been comprehensive enough to rule out the estimated travel times in this manuscript. Sampling for radionuclide contamination in the carbonate aquifer at Rainier Mesa has been too sparse in space and time to definitively conclude that no radionuclides have reached the carbonate aquifer. If contaminated water enters the aquifer it is likely to be at particular positions where preferential flow paths intersect the water table. Any inflow of contaminated water that occurs is subject to substantial dilution when it enters the aquifer and would be further diluted if it is transported to the location of a sampling well. Thus any contamination entering the aquifer would only be detected if its flow path led it to the place and time of sampling and the dilution it undergoes during that transport is slight enough to keep its concentration from falling below the detection limit. A further limitation is that the monitoring is conducted approximately once a year (National Security Technologies, LLC, 2006, 2007, 2008), which would make it difficult to detect any contamination transported in the form of a rapid early peak, as is sometimes observed for preferential flow.

p. 3909, lines 16-22: “The SRPF model has the strong advantage of not requiring site-specific unsaturated hydraulic properties . . . “ At the same time, a great disadvantage

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Interactive Discussion

Discussion Paper

of the SRPF approach is that it is not possible to incorporate quantitative site-specific data, if available, in order to achieve greater accuracy.More traditional unsaturated-zone flow models could be parameterised following the same philosophy as was done for the SRPF model, i.e. using "universal" values for hydraulic parameters determined with some "global" dataset. But with these models, it would at least be possible to decrease the uncertainty of the results by incorporating site-specific data. In either case, tests against direct observations are essential to demonstrate the value of a theory or a model and to judge its accuracy.

The authors are not convinced that more traditional unsaturated-zone flow models could be improved with more universal values of parameters, because these models rely heavily on unsaturated K and other properties that do not have the sensitivity-minimizing characteristics of Vo noted above, hence they are extremely sensitive to variations of media. In practical terms, this sensitivity gives these traditional models greater vulnerability to uncertainty when applied in the unfortunately common situation that the characterizing properties are poorly known. For example at Rainier Mesa the preferential flow at issue is affected by a cubic km or more of rock with great hydrogeologic complexity. Where one does not know the site-specific properties, use of a model that requires them is a vulnerability rather than a strength. The idea that many traditional hydrologic models are overparameterized is not new. Jakeman and Hornberger (1993) noted that most hydrologic records are spatially and temporally sparse, and thus are insufficient for parameterizing and evaluating complex, physically-based hydrologic models. Perrin et al. (2001) suggest that the structure of distributed models prevents extraction of important parameterization information from sparse hydrologic time series, and that complex distributed models may have been developed with excessive confidence when not adequately tested. As stated in the manuscript, we do not intend the SRPF model to supplant more traditional flow and solute transport approaches where there is sufficient data to parameterize and evaluate those models. The SRPF approach is most useful in cases where

very minimal data exist or as another first-arrival time to compare in a model ensemble approach.

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Technical corrections p. 3908, lines 11: ". . . definite detection limits (about 330 pCi L$-$1) . . . "In Table 3, values of 5.3 or 8.1 pCi L$-$1 are listed. There must be some mistake.

The values of 5.3 and 8.1 pCi L$-$1 are given for gross beta contamination, which uses a different analytical technique than used for tritium (3H) and has a much lower detection limit. The limits referred to by the reviewer of 330 pCi L$-$1 are given for 3H. No change to the manuscript needed.

p. 3908, line 11 + Table 2: Please consider using SI units (Bequerel instead of Curie).

Curies are what the results are reported in at the Nevada Test Site; the revised manuscript includes the conversion factor from picoCuries to Becquerels in Table 2.

p. 3910, line 22: ". . . must be occur . . . " – Please check wording.

The typo is corrected in the revised manuscript.

p. 3929, Fig. 5: This is a slightly modified version of Fig. 2 in Nimmo (2007). It should be further modified to cover the entire range of observed maximum transport velocities reported in that WRR paper (up to 480 m d−1). Please remember that the material must be cited appropriately according to the AGU Usage Permissions.

The revised manuscript has "[after Figure 2 in Nimmo (2007)]." added to the Figure 5 caption. Also, the two cases from Fran Ridge, NV (390 m d-1) and Yucca Mountain, NV (480 m d-1) have been added to the revised figure, per the reviewer's suggestion.

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Discussion Paper