Authors' response

Dear Editor and Reviewers,

We hereby enclose a new version of the manuscript after revising it using your valuable input and constructive criticism of the two Reviewers. The provided material starts with a letter to the Editor followed by a list with significant changes. Thereafter, there is a point-by-point response to each of the comments of the Reviewers on the previous version of this manuscript and a marked-up manuscript version of the old manuscript, where all revisions can be seen. For clarity all previous comments have been included unabridged in black text below, while our response is provided in red.

Response to Editor Decision

Reconsider after major revisions (further review by editor and referees) (09 Jul 2020) by Conrad Jackisch
Comments to the Author:
Dear Elin and co-workers,

Thank you again for submitting your manuscript to our special issue. Thank you also for your replies to the Reviewers' comments. You have drafted ways to take up their constructive criticism in the revisions.

I think it is also worthwhile to revisit our abstract of the special issue for that. We hope to find contributions which really aim at the intertwined relations of landscape properties and landscape functioning. As you have discussed in your replies, working out the novelties towards indicators of changes, methodological implications or restrictions etc. would be a way forward. So far, I feel reluctant to see a shifted focus on baseflow alone to be sufficient. But in combination with your suggestion to work out the role of snow coverage and the interrelation with the catchment's characteristics this appears more plausible to me.

You have stated that the Kryklan catchment is one of the best instrumented and understood boreal catchments. From the point of view of our special issue, this could be transformed into a large advantage to really track the value of this data to improve our understanding. Given the situation that MikeSHE and particle tracking is not the novelty and that it is neither the findings about MTTs/TTDs, maybe turning the argument towards what is the added information and where does it actually come from could be a way to also include a critical view regarding the state of our hypotheses and tools in hydrological sciences. During the discussions among the Guest Editors we found that much of our intentions with the special issue can be seen as closely related to the Critical Zone/Hydropedology concept of Henry Lin et al. but for the landscape/catchment in our case.

I hope you can align your revisions to these thoughts and would be pleased to receive your revised manuscript in due time.
All the best.
Conrad

Reply: We like to thank the Editor for these comments and suggestions. Reviewer #1 and #2 also indicated that we need to highlight more clearly the novelty of this study, so this is one of the major concerns we had to address in a revised manuscript. As stated in our initial reply to the Reviewers, the original manuscript was organized around the hypothesis that MTTs are controlled by catchment size. However, based on the comments of the Editor, both Reviewers and the available literature, we agree that the results of this study could be presented in a better way. We omitted the hypothesis as a rationale for the study and instead focused on other more novel aspects of this work.

In the new manuscript, we emphasized the implication of the northern landscape setting with prolonged dry winter conditions, followed by an intensive flow spring flood, dominating the hydrology. The seasonality of the catchment gives a unique opportunity to investigate seasonal changes in travel times and young water fractions and linking seasonal changes to catchment characteristics. We used isotope and base cation data to further test the credibility of the model results with respect to seasonal travel times and young water fractions. We linked the travel times to distinct catchment characteristics on a seasonal basis. We added the seasonality aspect in this version of the manuscript since we believe that this is a novel extension to the previous results. This extension will hopefully be of interest and give a new insight to intra-annual travel times for catchments with significant and distinct seasonality. Especially the winter and spring seasons have a distinct impact on stream chemistry in this northern setting and presumably the boreal landscape at large, and we believe that linking and explaining these effects and how they can be connected to catchment characteristics can give new insights to the functioning of boreal catchments.

To our knowledge, Krycklan is one of the most well-investigated and well-instrumented catchments in the boreal region with a large database of empirical observations that allow multiple ways to test the predictions of hydrological models. In our opinion, the discussion about MTTs, TTDs, and their variation at different scales in the landscape is far from finished. We believe that this study contributes significantly to the understanding of the hydrology of the snow-dominated boreal forest landscape, the coupling between hydrology and water quality, and, not least, the role of travel times in a changing climate. We argue that it is far from evident that MTTs and TTDs would look the same, and be controlled by the same factors, in different types of landscapes and seasons, so we believe that this type of studies from different areas of the world is necessary to improve our understanding of these matters.

We sincerely thank you and the Reviewers for your input and constructive criticism, which improved the new version of the manuscript. We hope that these changes will make you reconsider this manuscript for publication in Hydrology and Earth System Sciences (HESS).

List of significant changes:

- **The manuscript is no longer restricted to the winter baseflow but includes seasonal and annual travel times and young water fractions.**
    - The seasons included are three distinct seasons of the Krycklan catchment:
        - winter low flow - (no new recharge to the groundwater as the precipitation falls as snow and remains frozen)
        - spring high flow (snowmelt occurring late March to mid-May)
        - summer (season impacted by evapotranspiration, here defined as Jul to Sept, to reduce the impact of spring snowmelt (spring) and early snow (winter)).
    - New results for the 14 monitored sub-catchments:
        - Seasonal changes in travel times and young water fractions
        - The role of mires for young water and overland flow
        - Governing factors changes between seasons
    - Tables have been edited in the new manuscript:
        - Tables include seasonal observations and model results
        - Table 6 is new and includes seasonal correlations to catchment characteristics
    - Figures have been edited in the new manuscript to provide more in-depth analysis and presentation of the results:
        - Annual and seasonal results for catchment characteristics have been added
        - Figure 2 is new and is used to explain the theories used to connect seasonal stream chemistry to travel times and young water fractions.
        - Figure 5 is new, with an extended version in the Appendix. It shows the intra-annual variations of groundwater ages for the investigated sub-catchments. Compared to the annual mean times that were discussed in the previous version these graphs provide a more complete description of how the travel times vary both between different types of catchments and between different seasons

- **The introduction and discussion were changed.**
    - The introduction was extended using reference suggestions by Reviewer #1.
    - The introduction and discussion were updated to reflect the new scope of the manuscript and the new results that are presented. We hope that the extension of seasonality is a good step forward for a more interesting take on travel times. We believe that the snow-dominated Krycklan catchment with distinct seasonality gives an extra novelty aspect to these results.
    - The hypotheses regarding that catchment size is the mayor impacting characteristic has been omitted, and the manuscript is focused on explaining and understanding the seasonality of travel times and young water fractions in the snow-dominated boreal landscape.

- **Reviewer #1 and Reviewer #2 both requested an extension of the method section.** The method section in the new version of the manuscript was therefore extended to include a more in-depth description of the hydrological model and particle tracking. Figure 1 has been changed to include a soil map and a topography map. Figure 3 is new and is used to help explain the particle tracking method.

- **The abstract and title was changed accordingly to reflect the changes listed above**

Response to Reviewer #1

Introduction comment from Reviewer #1

- In this study the authors used a physically-based distributed model in combination with a particle tracking approach to determine groundwater travel times in the well-studied Kryklan catchment in northern Sweden. They compared the modeled mean travel times (MTTs) with average winter values of stable isotopes, pH, base cations and found significant correlations for all of them. Furthermore they tried to relate MTTs to certain catchment characteristics. The only strong and significant correlation they found was between MTTs and the fraction of low conductive sediments.

- Reply: We would like to thank Reviewer #1 for his/her constructive criticism and suggestions which helped improve the new version of the manuscript. The main concern raised was the need to improve the analysis and discussion part, which, we agree has improved the manuscript further. Our explanations and responses to all the Reviewer's comments and questions are listed below. All the comments have been included unabridged in this document, together with our response following each comment (red).

- The use of particle tracking approaches to determine travel time distributions with numerical models becomes more and more common in catchment hydrology. And although some of these approaches still suffer from certain simplifications (missing dispersion component, particles disappearing after temporary exfiltration, etc.) they can already shed light on general catchment dynamics. Having said that I am missing a more detailed description of both the model setup with Mike SHE and the particle tracking approach in particular (one or two additional figures would not hurt).

- Reply: We understand the Reviewers concerns regarding the method sections. A main reason for this is that the present model is based on the model that already has been thoroughly described in a previous paper by Jutebring Sterte et al. (2018), which is cited in the manuscript. However, we realize the need to provide the readers of this manuscript with a more proper background. We therefore incorporated a schematic figure in the method section as well as adjust the text to make the method description easier to follow and understand. Please see Figure 3 in the new manuscript which is an additional part of the particle tracking procedure. We have also added a more descriptive text of the Mike She water flow model in the revised manuscript and there is also an extended version of Figure 1.

- Language, style and structure throughout the manuscript are quite good and easy to follow.

- Reply: We appreciate the specific language comments listed below which helps to increase the understandability and language quality.

- In the introduction the authors present their hypotheses regarding the relationships between travel times and catchment characteristics. However, it seems that the review of relationships that have already been determined in former studies is a little short. I would like to point out that it is already quite well established that MTT saretime-variant and going along with this the strength of relationships between MTTs and certain physical catchment characteristics changes as well (this should be discussed at least in a little more depth than just mentioning it in line 49 once). Also research over at least the last 10 years has stressed the fact that it is in most cases a combination of multiple characteristics that control a catchment's MTTs. A comprehensive (short) summary of the state of art would be really helpful.

- Reply: We appreciate the Reviewer's comment and have added a section that better describes the state of art regarding this in the introduction of the revised manuscript. We have read the suggested literature and used these to extend the introduction and discussion. Reviewer #2 also addressed this issue and we do agree that our hypothesis used in the original version of the manuscript was not the best way to present the results of our study. We added the concept of seasonality in the new manuscript, i.e., we aimed to quantify the age of the water for three district seasons and try to find explanations to intra-annual changes in travel times for different sub-catchments.

- I am also missing a more detailed analysis of the modeled travel time distributions. I am especially intrigued by the very steep initial rise of the cumulative distributions. Given the fact that the TTDs are related to baseflow conditions, this seems particularly puzzling to me. Therefore I would like to see more explanation and discussion in this part. A figure of the modeled (not cumulative) TTDs would also be interesting/helpful.

- Reply: The groundwater in Krycklan is relatively shallow, especially around the mires. The till has a marked decreasing hydraulic conductivity with depth. The main flow of water occurs therefore in the shallow part of the till, resulting in fast groundwater flow especially during snow melt in the spring. TTD:s is the travel time of the groundwater for the whole simulated period, and thereby the steep initial rise of the TTD:s are mainly connected to the spring snowmelt. This emphasize the importance of conducting thorough investigations of TTDs and MTTs in different climate regions. We assume that the "true" baseflow can be represented by the groundwater reaching the streams during the winter period since there is no new input of precipitation to the soil during this time. This period also has an older age (gMTT). We added a figure with monthly groundwater age distribution and the partition of groundwater and overland flow (Figure 5) and changed the TTD figure (Figure 4) to show the particle age distribution for some streams.

- The discussion and conclusion sections are straightforward and quite easy to comprehend. This may, unfortunately, be due to the fact that the results are not really new. Catchment size is not related to MTTs; hydraulic conductivity, slope and flow path length are related to MTTs. In the end I am left wondering whether there is enough novelty (maybe relating pH and base cations to MTTs?). A more in depth discussion of the modeled TTDs could help or maybe additional analyses on the interdependencies of how different catchment characteristics combined control MTTs. Still, I believe that in the end the authors will manage to add more analysis and discussion to justify a publication in HESS.

- Reply: We extended the evaluation regarding the TTDs and the chemistry data and gave more focus to the seasonal change of mean travel time for different sub-catchments. We also added a figure that show the monthly groundwater age distribution to the streams Figure 4. Additionally, we added a section in the introduction part that highlights the unique set of data and models available, which enables a coupled analysis of chemistry and hydrology in order to understand the complex integrated surface-subsurface water system of the Krycklan catchment. It should be noted that in contrast to many previous studies this study was conducted in a snow-dominated catchment in the boreal forest region, which implies that one could expect differences in the hydrology and transit time distributions compared to more temperate regions where the majority of previous work has been conducted. We believe that the extended winter season conditions allow great opportunities to address the role of the base flow, its TTDs and relationship to water isotopes and water chemistry. Furthermore, we must also emphasize the comprehensive background data that was used for the setup and testing of the results, e.g. continuous discharge measurements and measurements of stable water isotopes and stream chemistry at 14 streams. Combined this should strengthen the reliability of the modelled TTDs.

Specific comments from Reviewer #1

-Reply: We agree with most specific suggestions and comments from Reviewer #1. However, some sections of the introduction and discussion were re-written with the help of this input and some sentences might end up being different in the revised manuscript.

Abstract:

- Line 23: '...to investigate...'
- Line 23: Better write: '...the travel time of the input to 14 [...] sub-catchments via groundwater to the stream...'.
- Line 30: Move this sentence up to Line 27 just after '...stream water.'.
- Line 31: I would add whether these were positive or negative correlations.
- Line 34: I would not call this a 'landscape characteristic'. Maybe better call it 'physical catchment characteristic'.
- Line 35: '...to positively correlate with MTT.'

- Reply: Major adjustment to the Abstract have been made to fit the new introduction, result and discussion section. Thereby, the sentences that the Reviewer have commented on has been changed. We have tried to account for the suggestions in the new text.

Introduction:

- Line 50: These referenced papers deal mainly with mean travel times, not with travel time distributions. Appropriate references for variations in TTDs would be for example Botter et al. (2010), Heidbüchel et al. (2012), Hrachowitz et al., (2013). Line 87: Again, I would not use 'landscape' factors since the term landscape refers to the visible landforms rather than to physical properties or subsurface features.

- Reply: We like to thank the Reviewer fort the suggested references. We have read them and used them to extend the introduction.

- Introduction: This section lacks the mention/discussion of previous work that is very relevant to the study. Especially concerning research on the connection of travel time and catchment characteristics. Your hypothesis is that catchment size correlates with travel time, but what about any other catchment properties? So far travel times have been related to slope, flow path length, soil thickness, antecedent moisture content, hydraulic conductivity, just to name a few. I recommend expanding the short introduction a bit more to include a brief discussion of potential travel time controlling factors (Ameli et al., 2016; Birkel et al., 2016; Haitjema et al., 1995; Heidbüchel et al., 2013; Hrachowitz et al., 2010; McGuire et al., 2005; Yang et al., 2018).

- Reply: We appreciate the Reviewer's suggestion and used them to extend the introduction using the suggested material. We also focused more on the connection between stream chemistry compared to model results. The long winter with no input of precipitation to the groundwater makes it possible to distinguish the baseflow from the discharge affected by new input of water to the system. Krycklan has a significant amount of empirical data and long-term observations that provides a unique opportunity to analyze a northern snow-dominated catchment.

Methods:

- Line 106: '...type OF regolith...'?

Reply: The sentence: "Connected by a network of streams, the different sub-catchments have distinct characteristics, which allow for an evaluation of the effects of catchment characteristics on hydrologic transport, **including type regolith**, vegetation, and differences in topography." was changed to: "Connected by a network of streams, the different sub-catchments have distinct characteristics, which allow for an evaluation of the effects of catchment characteristics on hydrologic transport, **including soil type**, vegetation, and differences in topography."

- Line 123: I would like to see a figure of the stratigraphy and how it is displayed in the model.

- Reply: We changed Figure 1 to include a map over the typography and a soil map. The manuscript also includes a more descriptive text about the Mike SHE model in the method section.

- Line 172: Are these winter GW travel times travel times of particles that entered the catchment during the winter or exited the catchment during the winter (are they from 'forward' or from 'backward' TTDs)? This is a very important and interesting question since it could be very different catchment characteristics that control forward or backward travel times.

- Reply: We agree that this could be better described to make it clearer. It is the age of particles exiting the catchment through stream discharge during winter regardless when they entered the groundwater as recharge, i.e. it is backwards TTDs. We hope that this was made clearer in the new version of the manuscript. Note that practically all precipitation falls as snow during the winter so typically there is little or no addition of water to the system during this period. Therefore, there is no addition of particles to the model during the winter months. Consequently, we doubt that it would be meaningful to trace the small amounts of water that might be added during the winters in these systems. Figure 3 was added to make the explanation of the particle tracking easier.

- Line 215: According to this equation a mire coverage of 100% would result in an infinitely large adjusted cation concentration which is quite unrealistic (this is equivalent to a scenario where mires do not contribute any cations whatsoever). How do you justify this relationship?

- Reply: The adjustment of the cation concentrations is based on observations of how mires affect the stream water concentrations of a wide range of elements. This has been described in detail by Lidman et al. (2014), which is cited in the manuscript. A general pattern was that the concentration of predominately weathering-derived elements such as the base cations decreased with the mire coverage. For the base cations, which do not sorb particularly strongly to peat, the decrease corresponded well with the decrease in mineral soils. This was therefore interpreted as dilution by mire water, since weathering does not occur in peat. For example, if 20% of the mineral soils in catchment was replaced by 20% peat, the weathering should be expected to decrease by approximately 20%, eventually causing approximately 20% lower concentrations in the runoff. (In practice, somewhat higher specific discharge from mires would increase the dilution effect, but then a slight addition of base cation with the precipitation would counteract this effect so the net effect is approximately 1% dilution by 1% wetland.) This is illustrated by Na in Fig. X with data from Lidman et al. (2014), where these matters are discussed in more detail for Na and other elements. We believe that this justifies the need to adjust the base cation concentrations for the effect of the mires in order to make a fair comparison to the modelled transit times, which include the mires.

In principle, however, the Reviewer is correct in stating that this adjustment would be problematic in cases where the mire coverage approaches 100%. One reason, as the Reviewer points out, is that the adjustment would lead to unrealistically high concentrations if the mire coverage were close to 100%. This is, however, only a theoretical problem since the mire coverage in the investigated sub-catchments never exceeds 44%. There is therefore no need to make such unrealistic and also unsupported adjustments. We find it hard even to envisage how a catchment with much higher mire coverage would be able to develop in this area, but since no such sub-catchments exist in the present study we believe that this discussion could be left for another context. Another reason why we would be reluctant to use this type of adjustment for such extreme cases where the mire coverage was close to 100% (had it for some reason been necessary) is that it would require extrapolation far outside the observed range. In this case the relationship to the concentration is significant between in the range 0-44% mire coverage. The observations were made in the same streams that were investigated in this study so there is clearly a justifiable relationship between base cation concentrations and mire coverage.

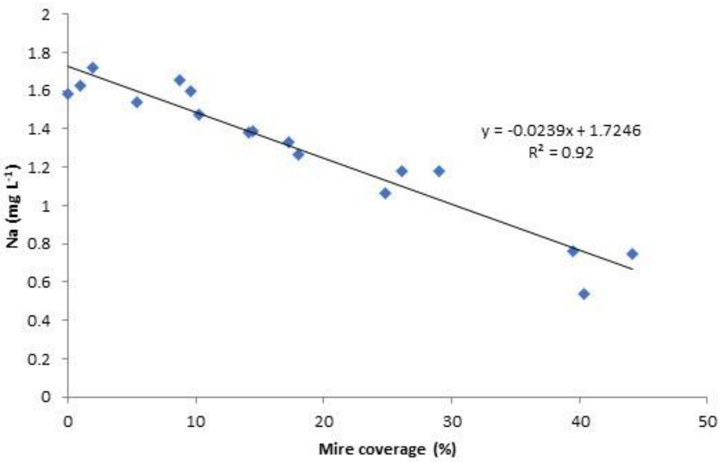

Figure X - Average Na concentrations in the investigated streams as a function of the mire coverage. Data from Lidman et al. (2014).

- Line 220: What exactly do you mean by this (‘...less impacted...’, ‘...also considered.’)?

<u>- Reply:</u> What we meant to express was that there is a fundamental difference between the water isotopes, on the one hand, and the stream chemistry, on the other. The interpretation of the water isotopes is based on the characteristic variation in the input signal throughout the year. Consequently, the important parameter is the variation in the water isotope signature in the runoff, as given in the manuscript. The annual average, however, is not a meaningful parameter. In the case of pH and base cation concentrations, the major drivers are processes within the catchment, e.g. weathering rates and transit times, and it is therefore meaningful to look also at the annual averages in this case. We have made major changes in method section 2.3 to describe this better.

- Line 239: But the simulated specific baseflow is not included in Table 4.

- Reply: Information about the specific discharge was omitted from Table 4 at a late stage. It must however still have been left in the text and this was corrected in the revised manuscript.

- Line 243: Again it would be important to know whether these are forward or backward times.

- Reply: We agree with this comment and checked and adjusted the entire text accordingly. To answer the question, it is backwards times, i.e. the age of a particle when it reaches a stream and exits the model, regardless of when they were first introduced to the groundwater. Since the system is frozen during the winters, there is only negligible addition of water. Hence, it would not be very meaningful to study the forward times for the winter period. The section regarding particle tracking (section 2.4) has been edited and the Figure 2 is new and is used to help to describe the particle tracking part of the study.

Results:

- Line243: I would some how indicate that the means are geometric instead of arithmetic when you write MTT (maybe something like 'gMTT'?). Because one will automatically assume that MTT is the arithmetic mean of the travel times.

- Reply: Thank you for the comment, we adjusted this in the new version since it will improve the clarity of the text and we decided to use the suggested gMTT.

- Line 245: This is not a complete sentence (verb missing).
- Line 248: 'Over the course of a year...'; '...may enrich or dilute...'
- Line 259: Winter baseflow or winter groundwater fraction?
- Line272: Instead of 'decreased' I would rather write'...became more negative/became more enriched in the lighter isotope...'
- Line 277: '... averages...'
- Line 304: 'Percentage of low conductive sediments...'

- **Reply:** The result section now includes seasonal travel time results. This change also changed most of the text in this section.

- Line 292-294: It would be good to add 'positive' or 'negative' before 'correlations', so you immediately know the direction of the relationship.

- Reply: We agree that this would add to the manuscript and make it easier for the reader to directly understand the relationship without having to look at the figures. This have been changed in the new version.

- Line 297: How do these equations show that LCS is the most significant parameter?

- Reply: The Reviewer has a valid point here. It is hard to justify based only on the equations in questions that LCS is the single most important parameter for controlling the MTTs in the investigated catchment.

This should clearly be rephrased in a more stringent way. The message we were trying to convey is that it seems hard to get around the impact of the LCS when looking at the MTTs throughout the landscape. As in this example, it is difficult to replace LCS by any other parameter and obtain equally good correlations, which indicates that it probably plays an important role for controlling MTTs. However, this section has been removed in the new manuscript to give space to seasonal travel time results and comparisons to catchment characteristics.

Discussion

- Line 314: '...on the other...'?
- Line 318: '...and a gamma distribution transfer function (convolution) method...'
- Line 318: 'In a conceptual, non-distributed modeling study...'
- Line 319: Some more details would be helpful ('another travel time distribution technique').

- Reply: The discussion section now includes an uncertainty/limitation part, a discussion towards seasonal travel time and chemistry results, as well as a discussion towards seasonal travel times and catchment characteristics. These changes required major changes of the text in the discussion section We agree with the comments from the Reviewer and had them in mind when writing the revised result section.

- Line 361: This section is a bit unstructured. You start out by stating that slope and hydraulic conductivity are the main factors controlling MTTs, then you state that the fraction of LCS is the most important factor adding travel distance to the mix before arguing that the steeper small C20 behaves differently maybe also because of the fluvial deposits fraction... Since these are some of your main findings it would be good to clarify the section. Also, what about the fact that you released the particles at the top of the transient groundwater table? That means that depending on the groundwater table a larger or smaller part of the regolith was not taken into account for the MTT calculations. If the water table was high a larger proportion of the regolith was considered, if it was low the particles started somewhere else in the profile. What are the implications of this? Would that influence your results?

- Reply: Please check the Figure 1, Figure 2, method section 2.1 and the new discussion 4.1 in the new manuscript. Most of the calculated groundwater table has a level between 0-3 m below the ground. The vertical discretization of the saturated zone follows the soil layers and are a few meters in depth close to the surface and increasing in thickness with depth. Note that the horizontal grid-size is 50*50 m and therefore the MTT will be bias to long travelling groundwater. Peatlands are generally the areas with an average groundwater table above 1 m, while the till areas have a depth between 2-3 m in average. However, at C14, the deep esker has a lower water table than on other locations in the catchments. The esker has the highest vertical hydraulic conductivity, and thereby the fastest route from infiltration to recharge. Although the horizontal distance is still larger than the distance to the groundwater table and the vertical hydraulic conductivity is large, one should note that this could impact the results and give C14 and C16 older MTT than if the phreatic surface was at the same level around the whole catchment. It can be noted that the areas were the MTTs mainly could be suppressed by this limitation in the modelling tools are areas with relatively long MTTs. This effect therefore somewhat counteracts the general patterns that were observed in this study. Hence, the observed patterns are not a result of the differences in groundwater depth. On the contrary, we would have expected patterns that are even more distinct if it had been possible to account for the time it takes for the water to percolate to the groundwater surface. We added a discussion about this (section 4.1), since it is an interesting subject and relevant for the interpretation of the results and try to make the section more structured.

- **Line 380:** It would be good to point out more the novel aspects of your work. What did you find out that was really new? The fact that Darcy's law works? Catchment size has been ruled out as a MTT control since quite a while now. Or is it mainly a study that confirms the applicability of the particle tracking of your model by comparing results to time series of pH, base cations and stable isotopes?

- Reply: Reviewer #2 also indicated that we need to highlight more clearly the novelty of this study so this is one of the major concerns we have tried to address in the revised manuscript. The original manuscript was organized around the hypothesis that MTTs are controlled by catchment size, but based on the comments of both Reviewers and the available literature, we agree that the results of this study could be presented in a better way by omitting this hypothesis as a rationale for the study and instead focus on other more novel aspects of this work. This includes emphasizing the implication of the northern landscape setting where the hydrology is dominated by prolonged winter conditions, and a dominating spring flood. Krycklan is one of the most well-investigated and well-instrumented catchments in the boreal region with a large database of empirical observations that allow multiple ways to test the predictions of hydrological models.

In our opinion, the discussion about MTTs, TTDs and their variation at different scales in the landscape is far from finished, and we believe that this study could contribute significantly to the understanding of the hydrology of the snow-dominated boreal forest landscape, the coupling between hydrology and water quality and, not least, the role of winter base flow in a changing climate. It is far from obvious that MTTs and TTDs would look the same and be controlled by the same factors in different types of landscapes and climates, so we believe that this type of studies from different areas of the world are necessary to improve our understanding of these matters. We further believe that the highlighting of the role of seasonal travel times in the revised version of the manuscript, is particularly suitable for the boreal landscape due to its pronounced seasonality. We also tried to emphasize better the large amount of data that we used to set up and test the model, e.g. the continuous discharge measurements in the streams and the extensive monitoring of water isotopes and stream chemistry. Combined we believe that this background data significantly strengthens the credibility of the conclusions we present.

Figure specific comments:

- Figure 1: Why do you call the sub-catchments subareas in this figure? Also, all subcatchments (not only the green ones) connect before reaching the main outlet at C16.

- Reply: We changed sub-areas to sub-catchments in Figure 1. Yes, all streams connect before they reach C16. However, the color code was used to illustrate which areas connect before they reach the white area. We tried to clarify this in the figure text.

- Figure 2: I am confused by the y axis label. Why does it start with 100% and then decrease to 0%? Isn't this a cumulative TTD (that should start with 0%)? Also, you never mention how you construct this TTD. Do you record the time the particles that arrive in the stream in the winter needed to travel through the catchment (backward TTD) or do you record the time that particles that are released during winter need to travel through the catchment (forward TTD)? I am curious since the cumulative TTDs exhibit a shape I would not have expected since the initial rise is very steep although you use a logarithmic x axis. For a groundwater TTDs in particular, this extremely high initial values are quite unusual. How do you explain this?

- Reply: Thank you for pointing this out! There was an error in this figure, the number of particles should start at 0 % and increase to 100 %, so the y-axis should change, see Figure 4 in the new manuscript. The figure now shows the annual age distribution of particles reaching the streams of four example sub-catchments. The snowmelt affects the groundwater travel time age and therefore there is a rapid start (Table 5). Given that a substantial portion of the annual precipitation leaves the catchments during a few weeks of spring flood, we do not think that the initial steepness of the distribution is surprising. This is probably one of the characteristics of boreal systems compared to other types of systems and emphasizes the importance of conducting this type of studies in different climate regions.

- Figure3: So if youre place the geometric MTT with thec ommonly used arithmetic MTT, how does that change the correlations? In case they become weaker you could argue for the general use of the geometric MTT.

- Reply: The arithmetic MTT is often used when looking at the gamma convolution distributions using isotopic data. It has been argued that the MTT should not be used for MTT older than 5 years because the amplitude signals are lost (Kirchner, 2016). Therefore, the aMTT is less impacted by the "old tail". For particle tracking or modelling methods the median or the geometric MTT may describe the bulk of the particles better than aMTT, because one particle (even though there are hundreds of particles in total) with a very old age may cause significant differences in the MTT estimation. This is undesirable, since the age of these very old particles becomes increasingly uncertain and are very hard to validate. The gMTT is less dependent on a few old particles but does still take them into account. The aMTT and gMTT are also well correlated and would probably give similar correlation results (Fig. Y).

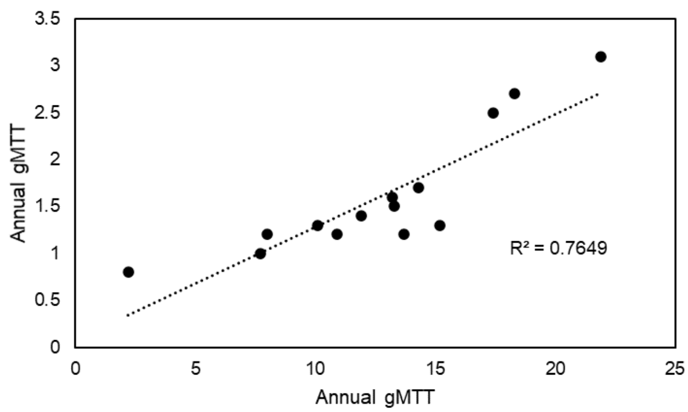

Figure Y – Correlation between gMTT (geometric mean) and aMTT (arithmetic mean) of particle tracking results

Kirchner, J. W.: Aggregation in environmental systems – Part 1: Seasonal tracer cycles quantify young water fractions, but not mean transit times, in spatially heterogeneous catchments, Hydrol. Earth Syst. Sci., 20, 279–297, https://doi.org/10.5194/hess-20-279-2016, 2016.

Supplements:

- Supplements S1: Check again for typos and misuse of words throughout the supplement (in particular in the table captions and the table foot notes).

S1, line 3: What does that mean that the table includes both non-transformed and log-normal transformed data? Which is which?

- S1, line 4: What happened to the other 44% of particles?
- S1, line 8: ‘Artesian’ mean? And why ‘back-transformed’?
- S1, line 19: ‘...as well as the precipitation (P)’
- S2, line 3: What does ‘The statistics are based on...’ mean?
- S2: The tables are not that helpful. Many of the abbreviations are not explained

- Reply: The appendix has been completely changed to fit with the new manuscript regarding seasonal changes. It now includes a description of the statistics used and an extended version of Figure 5.

Add to literature:

- Literature Ameli, A. A., Amvrosiadi, N., Grabs, T., Laudon, H., Creed, I. F., McDonnell, J. J., and Bishop, K.: Hillslope permeability architecture controls on subsurface transit time distribution and flow paths, J. Hydrol., 543, 17–30, https://doi.org/10.1016/j.jhydrol.2016.04.071, 2016.
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- Yang, J., Heidbüchel, I., Musolff, A., Reinstorf, F., and Fleckenstein, J. H.: Exploring the dynamics of transit times and subsurface mixing in a small agricultural catchment, Water Resour. Res., https://doi.org/10.1002/2017WR021896, 2018.

- Reply: We thank the Reviewer for these literature suggestions which was used to especially improve the introduction section. We read these suggestions and incorporated them were they seemed to be most fitting.

Response to Reviewer #2

Introduction comment from Reviewer #2

- In the manuscript "Linking groundwater travel times to stream chemistry, isotopic composition and catchment characteristics," Sterte et al. analyze the drivers of catchment travel times across catchments in norther Sweden. They use a physical hydrology model combined with particle tracking to generate transit time distributions and compare this to isotopic and stream chemistry observations. Overall, I think that the study is well done and the manuscript is well written and easy to follow.

Reply: We would like to thank Reviewer #2 for his/her constructive criticism and suggestions, which we helped to improve the manuscript. The main concern seemed to be the introduction and hypothesis, which we, having read the Reviewer's comments, agree benefited from being re-worked to progress the manuscript further. Our explanations and responses to all the Reviewer's comments and questions are listed below (red).

- However, I do have some significant concerns about the manuscript in its present form. My most serious concern is that it's not clear what the novelty of this study is that would warrant publication in HESS. All of the methods used here are well established and the idea that catchment travel time distributions are driven by catchment characteristics is not new. The authors start from the hypothesis that catchment area is the main driver, however previous research has already indicated that many drivers will be important, so disproving this hypothesis does not seem to be the best angle to take here. I would suggest the authors consider what portions of their findings are the most novel addition to the body of literature in this area and organize the manuscript around this rather than the area hypothesis.

Reply: Thank you for this comment, it helped to make a better manuscript. We received a similar comment from Reviewer #1. Based on these comments, we re-focused the manuscript, and in the new version, better highlight the novel aspects of this study. This includes emphasizing the implication of processes in the northern landscape setting where the hydrology is dominated by prolonged winter conditions followed by snowmelt. We do that by studying one of the most well-investigated and well-instrumented catchment systems in the boreal region. Both Reviewers have pointed out that the hypothesis that the catchment area is the main factor controlling the travel time distributions is obsolete and that the manuscript, therefore, should not be organized around this idea. Having considered the remarks of the Reviewers and studied the literature thoroughly, we feel inclined to agree that this was a mistake. We believe that the manuscript has benefited from the revision, where this idea does not have such a central place. As Reviewer #2 remarks below, this should not be the storyline of the manuscript, particularly as neither the modelling results nor the observations supported the area hypothesis.

- Even if the area hypothesis is what guided the study in the first place, this does not need to be the storyline of the manuscript. Along the same lines as my first comment I think the introduction could use significant revision. As it stands it is a very broad overview of the topic but I would like to see a more thorough review of previous finding directly relevant to this work that can clearly motivate the novelty of this study and the gap it is filling. Similarly I think the discussion section would be more powerful if it provides a better evaluation of how and where results form this study add new information/disagree or provide additional corroboration to existing studies.

Reply: Thank you for your comment. We have expanded the introduction to include a section regarding relevant works related to this study. We received some very helpful reference suggestions from Reviewer #1, which were incorporated in the introduction with regards to relevant research. We also agree that the catchment size hypothesis used in the original version of the manuscript was not the best way to present the results of this study, and this hypothesis has therefore been omitted. Both the introduction and the discussion were changed accordingly. We focused the study on the connection between seasonal travel times and stream chemistry (base cations and isotopes). There is a unique opportunity to distinguish the baseflow in the streams due to the prolonged winter, which gives little to no new input of water to the system and then comparing it to spring (high flow) and summer. The boreal systems are sensitive to climate change, and to have as much of a base understanding of these systems as possible is important to be able to evaluate changes in the future. Shorter snow seasons and changes in the amount of precipitation can change travel times in the future, which can have an impact on weathering and biological processes. We have in this study showed a strong relationship between isotope mixing and base cation concentrations, on the one hand, and groundwater travel times on the other. We have already seen changes in the climate and in the hydrology in the last couple of years in Krycklan, making this a pressing issue.

- For the most part I think the paper is very clearly written, however the description of the modeling approach is a bit confusing and could use some more details. For example the term simulation is used to refer to both the hydrology model and the particle tracking portion which can be confusing. This section could be helped by a figure or a schematic to illustrate the approach I think.

Reply: Reviewer #1 also requested a schematic figure, and we agree with the Reviewers and like the suggestion to better describe the modeling procedure (Figure 3). We aimed to give more distinctive terms to the hydrology modelling part and the particle tracking part of the manuscript. One reason for the confusion is probably that much of the hydrological model was described in a previous paper. Although this paper was cited, we realized that we need to explain the model setup in more detail also in this manuscript so that potential readers can grasp the general idea of what has been done without reading the previous paper (see method section 2.2).

Specific comments from Reviewer #2

Reply: We agree with the most specific suggestions and comments from Reviewer #2. However, some sections of the introduction and discussion will be re-written whit the help of this input, and some sentences might end up being different in the revised manuscript.

- I think the catchment numbering could be done in a more intuitive way so its easier to separate unique outlets (i.e C12-15). I would suggest giving each of these outlets their own letter and then numbering points within them potentially by drainage area, that way itis easier to compare when points are within the same drainage or not.

Reply: We understand the concern regarding this comment. However, we hesitate to change the names of the sub-catchment since they all have been included in many previous peer-reviewed papers, which argues against introducing new names in this manuscript. It would make all comparisons with previous papers from the Krycklan catchment unnecessarily difficult and confusing. We hoped that the colors of figure 1 will help to distinguish which sub-catchments were connected. However, we also added a color code in table 1, which will further connect the different sub-catchments in figure and text.

- Line 138-140: This is confusing are you trying to say that the hydrologic model is run first and then the particle tracking is applied to the outputs of that model?

Reply: Yes, the model flow field is run first and, then one year of the flow field is cycled for 1000 years with particle tracking applied to the flow field results. We will add the schematic figure (see Figure 3) to clarify these steps and be more direct that the hydrology model output used are the results from the model is used in Jutebring et al. (2018). We will also add a more in-depth description of the water movement model in the revised manuscript (method section 2.2.).

- Section 2.3 – this is really more a description of scenario design than numerical methods. I would consider renaming.

Reply: We appreciate this suggestion from the Reviewer and will incorporate the suggestion and change the name from "Numerical methods" to "Establishing travel times - Particle tracking"

- Line 148: 'several years' is very vague can you be more precise.

Reply: We agree with the Reviewer and have tried to be more precise in the revised manuscript. It is one year of the Mike SHE flow results that has been cycled for 1000 years in order to allow for long-term particle tracking. This has been clarified this throughout the manuscript.

- Line 154: at what time frequency were particles injected into the model? Just at the start of each year?

Reply: This has been clarified in section 2.4 and the new Figure 3. The particles are only introduced in the first year. The Mike SHE flow results of that year are then cycled 1000 times to let the particles travel for a longer period (1000 years).

- Line 159: I think the more standard reference for this would be heavy tailed rather than long tailed.

Reply: A distribution with a long tail means that there is a large proportion of the distributions is far away from the main central tendency. A long-tailed distribution can also be heavy tailed, meaning that a distribution goes towards zero slower than an exponential distribution. In the section in question, we are talking about the use of the geometric mean instead of the arithmetic mean for long-tailed distributions because the geometric mean is less bias to the long tail and describes the central tendency more effectively.

- The term simulation is used to refer to both the hydrology model and the particle tracking model and this can make the methods confusing when you are talking about run times for example.

Reply: We looked through the manuscript and try to be more distinct when using the term simulation so that it becomes clearer when we talk about the flow model or the particle tracking. We also made a distinction in the method section were we first talk about the site (section 2.1), then the hydrology model (section 2.2) and thereafter the particle tracking (section 2.4)

- At the beginning of section 2.3 you say that you used several years of simulation but actually it looks like you use only one year of the hydrologic simulation but repeated it 1000 times (i.e. more than several). This description is confusing.

Reply: We have taken one year and repeated it 1000 times. We tried to be more distinct and not use the word several for better clarity in the revised manuscript.

- I think some of the tables could be converted to figures to better present the information. For example Table 4 could be presented as a series of maps.

Reply: We do not understand exactly what the Reviewer is asking for. Most of the information in the old manuscript Table 4 was shown in Figure 2. We attempted to make maps at the onset of this study, but the information was not clearly visible. However, we added travel time distribution per month for each sub-catchment to visualize the impact of various fractions of travel times. See new figure 5 and Appendix.

Linking ~~groundwater~~ travel times and flow pathways to stream chemistry, isotopic composition, and catchment characteristics in a snow-dominated landscape

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Key Points:

- A numerical model was used to ~~simulate groundwater~~estimate annual and seasonal mean travel times and flow pathways across the Krycklan catchment in the boreal region of northern Sweden.
- The ~~modeled~~estimated annual mean travel times of ~~groundwater in~~ 14 partly nested sub-catchments ranged ~~from~~between 0.~~5-3.6~~8 and 2.7 years
- The ~~modeled~~estimated travel times and young water fractions were consistent with ~~both~~ observed stream chemistry (base cation concentration and ~~pH) and~~ stable water isotopes ~~(~~, $\delta^{18}O$ ~~and δ^2H)~~).
- Hydraulic conductivity of the ~~regolith~~soil was the most important factor regulating the variation in ~~groundwater~~mean travel times ~~between different~~, while mires mainly affected the youngest fraction of stream water.
- Although all sub-catchments showed seasonal changes in mean travel times and young water fractions, the greatest seasonality was found in sub-catchments with a substantial fraction of mires.

Abstract

Understanding travel times and hydrological pathways of rain and snowmelt ~~inputs~~water transported through the ~~subsurface environment~~ landscape to recipient surface waters is critical in many hydrological and biogeochemical investigations. In this study, a particle tracking model approach in Mike SHE was used to ~~investigating~~investigate the travel time, and pathway of ~~stream groundwater input to~~water in 14 partly nested, long-term monitored boreal sub-catchments~~. Based on previous studies in the area, we hypothesized that the main factor controlling groundwater travel times was catchment size. The modeled mean travel time (MTT) in the different sub-catchments ranged between 0.5 years and 3.6 years. Estimated MTTs were tested against the observed~~ characterized by long~~-term winter isotopic signature ($\delta^{2}H$, $\delta^{18}O$) and chemistry (base cation concentration~~ and ~~pH) of the stream water. The underlying assumption was that older water would have an isotopic signature that resembles the long-term average precipitation input, while seasonal variations would be more apparent in catchments~~ snow rich winters with ~~younger water. Similarly, it was assumed that older water would be more affected by weathering, resulting in higher concentrations of base cations~~little groundwater recharge and ~~higher pH. 10-year average winter values~~ highly dynamic hydrology during the following snowmelt. The calculated annual mean travel times for ~~stream chemistry were used for each sub-catchment. We found significant correlations between the estimated~~these sub-catchments varied from 0.8-2.7 years. The seasonality caused considerable variation in travel times between different seasons and landscape types, with winter mean travel times ~~and average water isotope signature ($r=0.80$, $p<0.001$ for $\delta^{18}O$; $r=0.81$, $p<0.001$ for $\delta^{2}H$). We also found a strong correlation between MTT~~ranging from 1.2-7.7 years and ~~base cation concentration ($r=0.77$, $p<0.001$)~~spring mean travel times ranging from 0.5-1.9 years. The modelled variation in annual and ~~pH~~seasonal travel times and the fraction of young water (less than three months) was supported by extensive observations of both $\delta^{18}O$ and base cation concentrations in the stream water. The age of the ~~streams ($r=0.54$, $p<0.01$), which strengthened the credibility of the model. There was no statistical correlation between catchment size and MTT of groundwater, hence refuting our hypothesis. Instead, one landscape characteristic,~~ groundwater was positively correlated to the abundance of low conductive ~~sediments,~~soils ($r=0.90$, $P<0.0001$). As a result of lacking synchronicity and contrasting hydrological responses between different soil types (e.g., peat and low-conductive soils), mixed catchments typically displayed larger differences in travel times between winter baseflow and spring flood. Mires were found to ~~be most influential. Its areal proportion was found to positively~~ affect ~~MTT.~~

the young water fractions of the stream contribution ($r=0.96$, $P<0.0001$) by introducing larger differences in the mean travel times between the seasons compared to forest dominated sub-catchments. The main factor for this difference is likely related to the soil frost in mires, causing considerable overland flow in spring. The lower recharge during these periods caused mire-dominated catchments to have older stream water contribution than comparable forest catchments during other parts of the year. Boreal landscapes are sensitive to climate change, and our results show that changes in seasonality are likely to affect different landscape types in different ways.

1 Introduction

The age and ~~origin~~pathways of ~~stream~~ water through the terrestrial landscape to streams is a widely discussed ~~research area~~topic in contemporary hydrology. This interest has emerged ~~due to~~because of the important role ~~water age~~residence time, and ~~flow pathways~~ routing of water through various subsurface environments have ~~for~~on hydrological and biogeochemical processes (McDonnell et al., 2010; Sprenger et al., 2018). ~~Such processes~~These include fundamental implications for weathering rates (Burns et al., 2003), transport and dispersal of contaminants (Bosson et al., 2013; Kralik, 2015~~), weathering (Burns et al., 2003~~), and accumulation and mobilization of organic carbon and associated solutes (Tiwari et al., 2017). ~~These processes have received increasing attention also in snow-dominated landscapes due to their importance as water resources (Barnett et al., 2005) and their susceptibility to climate change (Aubin et al., 2018; Tremblay et al., 2018; Price~~The ~~et al., 2013). In the vast boreal region, the landscape often consists of heterogeneous patches of lakes, mires, and coniferous forests regulated by sometimes contrasting hydrologic mechanisms. This heterogeneity emphasizes the need for an enhanced understanding of hydrological and biogeochemical processes and their inter-linkage (Winnick et al., 2017; Waddington et al., 2015; Demers et al., 2010).~~ travel time ~~to~~, from precipitation input to the outflow into streams, provides valuable information about catchment sensitivity to changes in land use and climate as well as for the ~~input~~fate of long-range transport of contaminants and nutrients deposited with precipitation (van der Velde et al., 2012). The travel time distribution can vary substantially in time and space, depending on catchment characteristics and hydrological conditions, including, for example, slope, catchment size, soil heterogeneity, and seasonality (Botter et al., 2010; Lin., 2010; Heidbüchel et al., 2012; Hrachowitz et al., 2013). ~~numerous catchment characteristics and hydrological conditions (McGuire and McDonnell, 2006; Scanlon et al., 2001). Therefore, estimating travel times for various contrasting landscape elements may enhance our process understanding and ability to more correctly quantify the contribution of water and various solutes derived from catchments. Groundwater is an especially important component of the hydrology and a regulator of many biogeochemical processes. From a surface water perspective, groundwater is a source of recharge for streams, lakes and, wetlands. Groundwater is the part of stream water contribution which is not linked to a specific hydrological episode and can, therefore, be used as a reference point to study solute transport, water quality and other event-activated processes (Bergknut et al., 2010; Doyle et al., 2005; Olson & Hawkins, 2012; Pinder & Jones, 1969). Northern landscapes with long-lasting snow cover and prolonged frozen conditions without the input of new surface water can give a unique opportunity to investigate stream-groundwater input conditions (Peralta-Tapia et al., 2015).~~Therefore, estimating travel times for contrasting landscape elements is a challenging task but may enhance our ability to understand catchment functioning more adequately.

Stream water consists of a blend of groundwater and overland flow of different ages. The mean travel time (MTT) to streams is calculated as the average age of this mix (McGuire et al., 2006). The baseflow is the part of stream groundwater contribution that is not linked to a specific hydrological episode and instead part of the runoff mix that generally has travelled the farthest and is the oldest (Klaus et al., 2013; Hrachowitz et al., 2016). In contrast, young waters are connected to overland flow or fast and shallow groundwater, which mainly can be seen at times when new precipitation input and/or snowmelt arrives (Peters et al., 2014; Hrachowitz et al., 2016). Travel times are complex to quantify, especially at intra-annual time scales, as they can vary in time and space (Botter et al., 2010). A better understanding of the seasonal variability in the connection between young and old waters in various catchment systems can help provide insights into the fundamental role catchment characteristics play for the regulation of the hydrology and biogeochemistry of streams and rivers.

Stable water isotopes and biogeochemical tracers are some of the most common tools applied in field investigations to locate sources of water and follow its pathways through the landscape (~~Barthet al., 2007; Goller et al., 2005;~~ Maulé and Stein, 1990; Rodhe et al., 1996; Goller et al., 2005; Tetzlaff and Soulsby, 2008). Isotopic tracer dampening can provide an estimate of ~~water~~mean travel times (Uhlenbrook et al., 2002; McGuire et al., 2005; Peralta-Tapia et al., 2016~~; Uhlenbrook et al., 2002; van Geldern et al., 2014~~), and~~,~~ more elaborate time-series analysis can provide quantitative assessments of water age (Harman, 2015; Danesh-Yazdi et al., ~~2016; Harman, 2015). Many solutes will~~2016). Theoretical transfer functions, such as the gamma distribution model, can be related to input-output signals (for example, precipitation to discharge) of isotopes (McGuire et al., 2005; Hrachowitz et al., 2010; Heidbüchel et al., 2013; Birkel et al., 2016). The isotope amplitude signal used to estimate mean travel times in many transfer functions is, however, lost after approximately four to five years (Kirchner., 2016), which limits the use of isotopes for catchments with long travel times. The fraction of young water, often defined as water younger than two to three months, can, however, still be quantifiable in such catchments (von Freyberg et al., 2018; Lutz et al., 2018; Stockinger et al., 2019). Water isotopes mainly fractionate due to evaporation and are hence not affected by their subsurface pathways. In contrast, biogeochemical tracers may react and transform ~~differentially during~~on their route to ~~the~~a stream (~~Ledesma et al., 2018;~~ Lidman et al., 2017~~).~~; Ledesma et al., 2018). Such transformations and reactions depend on the specific solute and soil environment that water encounters and can hence give qualitative information about ~~travel time through the~~ groundwater flow pathways in mineral soils ~~and groundwater flow pathways (~~(Wolock et al., 1997; Frisbee et al., 2011; ~~Wolock et al., 1997;~~ Zimmer et al., 2012). Therefore, combined information from conservative and reactive tracers can provide an enhanced understanding of hydrological and biogeochemical processes ~~in the catchment~~ (Laudon et al. 2011). ~~However, field investigations often require tracer inputs and outputs to be adequately controlled and can hence be impractical at larger temporal and spatial scales.~~

A complementary approach to field experiments is numerical ~~modeling~~modelling, which can be useful for achieving a system understanding of catchment hydrology. Lumped hydrological models ~~can~~often describe catchments as single integrated entities. In contrast, distributed numerical models can ~~consider~~include spatial heterogeneity in input parameters~~. Therefore, they~~ and therefore have the potential to represent catchment processes ~~in a~~ more ~~realistic manner, which~~realistically. In turn, this can lead to a more process-based understanding of hydrology and biogeochemistry at the catchment-scale (Brirhet and Benaabidate, 2016; Soltani, 2017). A common method to calculate travel times using numerical methods includes isotope models and particle tracking (Hrachowitz et al., 2013; Ameli et al., 2016; Kaandorp et al., 2018, Yang et al., 2018). Models, however, need – as far as possible – proper tests against real observations to build confidence in their outputs~~, and as a rule, this requires large amounts of empirical data.~~. Stream discharge, groundwater, and tracer data are examples of such validation data that can provide important information to understand a catchment hydrological functioning (McGuire et al., 2007; Hrachowitz et al., 2015; Wang et al., 2017). Such empirical data are costly and time-consuming to collect. Therefore, data for calibration and validation is often limited, and the minimum length of data sets and methods needed in data-sparse catchments is currently a topic for some debate (Bjerklie et al., 2003; Jian et al., 2017; Li et al., 2018).

~~In this study, advective travel times groundwater input~~

Snow-dominated landscapes have gained increasing attention due to ~~streams were investigated in~~their importance as water resources (Barnett et al., 2005) and their vulnerability to climate change the ~~well-studied Krycklan~~ last decades (Tremblay et al., 2011; Aubin et al., 2018). One snow-dominated catchment with long continuous data sets for multiple monitoring stations in the boreal ~~region~~ catchment Krycklan in northern Sweden (Laudon et al~~.~~., 2013). These data sets give a unique opportunity for investigation of the hydrological functioning of the heterogeneous boreal landscape. Boreal catchments with long-lasting snow cover that melts rapidly in the spring create both opportunities and challenges in the context of the determination of age and pathways of stream water. The boreal region consists of heterogeneous patches of lakes, mires, and mostly coniferous forests regulated by sometimes contrasting hydrologic mechanisms. This heterogeneity emphasizes the need for an enhanced understanding of hydrological and biogeochemical processes and their inter-linkage in these systems (Temnerud and Bishop, 2005). In high latitude snow-dominated catchments, little to no new input of water occurs to the soil during the several months of winter conditions, whereby the source of water to the streams is originating from baseflow (Peralta-Tapia et al. 2016).~~) using a~~ These specific conditions provide unique opportunities to study the source of water that have spent the longest time in the sub-subsurface environment.

In contrast to the conditions of winter baseflow, significant amounts of water are added to the system during the often short and intensive spring snowmelt period (Spence et al., 2011; Spence and Phillips, 2015; Lyon et al., 2018). Although attempts to assess travel times generally have shown good fits for a gamma distribution function in snow-dominated catchments, particularly the winter season has proven to be challenging, which suggests that other methods to assess travel times may be required (Heidbüchel et al., 2012; Peralta-Tapia et al., 2016). To account for the unique circumstances of both baseflow with long transit times and those of the intensive spring snowmelt with potential large overland flow components requires the need of models that can handle the complexity and separation of various flow components.

In order to overcome previous model limitations, this study used particle tracking in the physically based distributed numerical model, Mike SHE (Graham and Butts, 2005~~). The model~~), with the purpose of enhancing our understanding of stream water contribution in boreal landscapes across seasons and landscape configurations. The water moment model in Mike SHE calculates saturated (3D) groundwater flow and unsaturated (1D) ~~groundwater~~ flow and is fully integrated with the surface water as well as evapotranspiration. The water movement model setup and results previously presented by Jutebring Sterte et al. (2018) ~~was~~were used ~~and~~as a platform in the present study. The model has been calibrated and validated to ~~observed~~ 14 sub-catchment of daily stream-discharge ~~and~~observations and 15 groundwater wells of periodically measured groundwater levels throughout the Krycklan catchment~~.~~ (Laudon et al. 2013; Jutebring Sterte et al., 2018). The complexity of the model allows for an in-depth investigation of advective travel times by non-reactive particle tracking simulations in a transient flow field.

~~Based on previous work in Krycklan (Peralta-Tapia et al., 2015), we hypothesize that~~

The main objective of this study was to quantify seasonal age distributions and calculate mean travel times of stream water contribution of the sub-catchment ~~size is the primary factor determining groundwater travel times since the study site has a relatively uniform glacial history, geology, and climatic conditions. Firstly, we tested~~ in Krycklan in order to disentangle how these are related to physical landscape characteristics and seasonality. Firstly, the credibility of the model results was tested by comparing ~~particle~~calculated travel times ~~from~~for the 14 long-term monitored sub-catchments to ten-year ~~winter $\delta^{18}O$ and $\delta^{2}H$~~seasonal isotope signatures~~, as well as ten-year winter~~ and base cation ~~and pH records~~concentrations record from the Krycklan network. ~~Thereafter, we tested our hypothesis by linking~~The usefulness of stream isotopic composition and chemistry has previously been demonstrated for understanding the ~~estimated particle~~connection of hydrological flow pathways and travel times ~~to various~~for this site (Laudon et al., 2007; Peralta-Tapia et al., 2015), but with the limitation of studies on only short periods or single catchments. Secondly, the purpose was to go beyond what has previously been done by identifying the connection between travel times and different catchment characteristics ~~to evaluate and generalize the regulating landscape factors.~~and test how this varies between different hydrological conditions and seasons. This was accomplished by capturing contrasting seasons such as the low flow conditions in winter with limited input of new precipitation, high flow in spring when the system still is partly frozen, and summer when evaporation becomes a significant process.

2 Method

2.1 Site description

The Krycklan study catchment, located in the boreal region at the transition of the temperate/subarctic climate zone of northern Sweden, is spanning elevations from 114 to 405 m.a.s.l (Fig. 1, Table 1).~~.~~ The characteristic features of this boreal landscape are the dominance of Scots pine (*Pinus sylvestris*) and Norway spruce (*Picea abies*) covering most of the catchment (Laudon et al., 2013). Krycklan has a landscape distinctively formed by the last ice age (Ivarsson and Johnsson, 1988; Lidman et al., 2016). At the higher altitudes to the northwest, which are located above the highest postglacial coastline, the ~~regolith~~till soils can reach up to 15-20 m in thickness~~ (Figure .1, Table 1).~~ Here, the ~~regolith~~soil primarily consists of sandy-silty till, and the landscape is intertwined with lakes and ~~mires. Previous studies have indicated that~~peatlands. In this study, we refer to soils as all unconsolidated material above the ~~hydraulic conductivity of the till decreases with depth with the significant part of the flow occurring in the upper half meter of the regolith (Ågren et al., 2014; Bishop et al., 2004).~~bedrock. The decreasing hydraulic conductivity with depth is characteristic for glacial tills in northern Sweden (Bishop et al., 2011; Seibert et al., 2009) with conductivities close to $5\cdot10^{-5}$ m/s~~$^{-1}$~~ at the ~~soil~~ground surface and exponentially decreasing ~~with depth~~downwards (Nyberg, 1995). ~~The high conductivity near the surface causes the main lateral groundwater transport to occur in the shallower parts of the till.~~ At lower altitudes, the ~~regolith~~soils mainly ~~consists~~consist of fluvial deposits of silty clay and sand. Compared to the ~~regolith~~soils at higher altitudes, these ~~sandy~~ deposits can reach thicknesses up to approximately ~~60~~40-50 m~~ or more~~ and are more homogeneous with depth.

The catchment~~, which is used for~~ is divided into 14 nested sub-catchments and has been included multi-disciplinary biogeochemical and hydrological research for more than 30 years (Laudon ~~&~~and Sponseller, 2018~~), is divided into 14 nested~~). The sub-catchments~~,~~ are called C1 to C20~~,~~. The longest continuously monitored time-series of ~~which 14 were used in this study.~~ streamflow began at the beginning of the 1980s. Connected by a network of streams, the different sub-catchments have distinct characteristics, which allow for an evaluation of the effects of catchment characteristics on hydrologic transport, including soil type~~ regolith~~, vegetation, and differences in topography.

Table 1: Sub-catchment characteristics. The ~~table~~list includes all 14 monitored sub-catchments in Krycklan, called C1 to C20~~,~~. Different branches of the stream network are illustrated in ~~order of catchment size (see~~different colours in Fig. 1~~)~~. The table includes sub-catchment area, average elevation, and average slope. Further description of these characteristics can be found in Karlsen et al. (2016). ~~The table also includes soil type based on the regolith map (1:100,000) from the Swedish Geological Survey. The area proportions were calculated from the 50*50 m map created in Mike SHE. The numbers in brackets represent the proportion of the sub-catchments that are assumed to be more silty sediments, i.e., with low conductive soils (LCS).~~The table also includes soil proportion based on the soil.

	Catchment size (km^2)	Average elevation (m.a.s.l.)	Slope (°)	Till (%)	Mire (%)	Sorted sediments (%)	Lakes (%)
C1	0.48	279	4.87	91	0	0	0.0
C2	0.12	273	4.75	79	0	0	0.0
C4	0.18	287	4.24	29	42	0	0.0
C5	0.65	292	2.91	47	46	0	6.4
C6	1.10	283	4.53	51	29	0	3.8

C7	0.47	275	4.98	68	16	0	0.0
C9	2.88	251	4.25	64	14	11 (4)	1.5
C10	3.36	296	5.11	64	28	1	0.0
C12	5.44	277	4.90	70	18	6	0.0
C13	7.00	251	4.52	60	10	18 (9)	0.7
C14	14.10	228	6.35	46	6	39 (15)	0.7
C15	19.13	277	6.38	64	15	10 (2)	2.4
C16	67.90	239	6.35	51	9	31 (10)	1.0
C20	1.45	214	5.96	55	9	28 (28)	0.0

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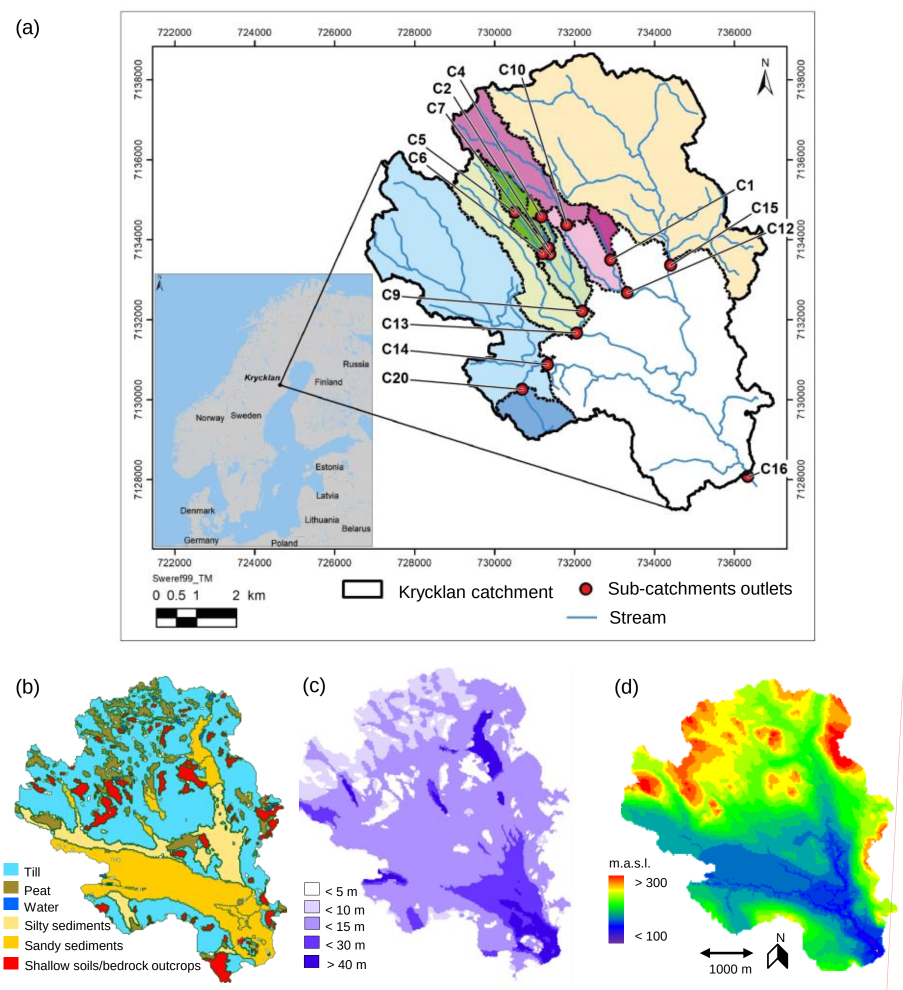

Figure 1: **The Krycklan catchment.** (a) Location of sub-catchment and ~~**sub-catchment**~~ their outlets ~~**(red circles).**~~. The areas are color-coded based on their stream network connections, e.g., all ~~green~~ sub-catchments of one colour connect before reaching the ~~main outlet, C16.~~white area. For further details of the catchment characteristics, see Table 1. ~~The scale and coordinate system refer to~~ (b) The soil map used in the ~~local figure over~~hydrology model is based on the ~~catchment.~~ soil map (1:100,000) from the Swedish Geological Survey (2014), combined with field investigations. (c) Depth to bedrock from the Swedish Geological Survey (2014) is shown in meter below the ground surface. (e) Catchment topography, shown as meter above sea level (m.a.s.l.).

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2.2 Water flow model setup

~~The~~ – Mike SHE

The Mike SHE model setup used as a platform in this study was a slightly ~~updated~~modified version of the previously established and validated surface and groundwater model presented ~~by~~in Jutebring Sterte et al. (2018). The model has a horizontal grid resolution of 50*50 m. Vertically the model is divided into ten calculation layers and reaches a depth of 100 m below ground. The calculation layers follow the ~~regolith~~ stratigraphy of the soil with one exception: the uppermost layer thickness was set to 2.5 m~~.~~ (Fig. 1, Table 2). This exception was due to the numerical implementation of the unsaturated zone and the evapotranspiration processes in Mike SHE, which only are fully active in the uppermost calculation layer. Therefore, the uppermost layer ~~has to~~must be deep enough to cover the part of the ~~regolith~~soils influenced by evapotranspiration processes and capillary rise of groundwater. This depth averages several ~~regolith~~soils types into one calculation layer, which may underestimate the observed high horizontal hydrological conductivity in the shallowest parts of the till (Peralta-Tapia et al., 2015). Numerically this is accounted for by implementing a depth-dependent drainage function, which increases the ~~flow~~groundwater velocity in the ~~shallowest~~uppermost part of the ~~till~~soil (Bosson et al., 2008). For more information regarding the model~~-~~ setup, see Jutebring Sterte et al. (2018). For this study, a few changes were made to the original Krycklan Mike SHE model. Most importantly, new field data from the Krycklan database gave a more precise location, and the threshold level of the lake outlet of C5 and the horizontal conductivity of the silty sand was increased from 1*10-8 m/s to 1*10-7 m/s. The corrections and additions did not influence the model results in any substantial way.

Table 2: Flow model setup. Flow model setup from the calibrated and validated Mike SHE model presented in Jutebring Sterte et al. (2018). The "soil type surface" corresponds to the soil type shown in Fig. 1b

Soil type surface	Depth below ground (m)	Soil type	Horizontal hydraulic conductivity (m/s)	Vertical hydraulic conductivity (m/s)
Till	2.5	Till	$2*10^{-5}$	$2*10^{-6}$
	To bedrock	Fine till	$1*10^{-6}$	$1*10^{-7}$
	Bedrock		$1*10^{-9}$	$1*10^{-9}$
Peat	5	Peat	$1*10^{-5}$	$5*10^{-5}$
	7	Clay	$1*10^{-9}$	$1*10^{-9}$
	To bedrock	Fine till	$1*10^{-6}$	$1*10^{-7}$
	Bedrock		$1*10^{-9}$	$1*10^{-9}$
Silty sediments	3	Silt/clay	$1*10^{-7}$	$1*10^{-7}$
	To bedrock	Fine till	$1*10^{-6}$	$1*10^{-7}$
	Bedrock		$1*10^{-9}$	$1*10^{-9}$
Sandy Sediments	0.8	Silt/Sand	$1*10^{-7}$	$1*10^{-7}$
	2.8	Silt/clay	$1*10^{-8}$	$1*10^{-7}$
	0.9*max depth	Sand	$3*10^{-4}$	$3*10^{-5}$
	To bedrock	Gravel	$1*10^{-4}$	$1*10^{-4}$
	Bedrock		$1*10^{-9}$	$1*10^{-9}$
Soil frost				
Peat	Reduced vertical and horizontal flow during winter			

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Particle tracking in Mike SHE enables groundwater travel time investigations, as described in detail ~~by~~in Bosson et al. (2010, 2013). Particles in the model will only follow the saturated groundwater flow by advection. In Mike SHE, it is possible to release particles, with unique identification numbers, at any depth and location. During the particle tracking, the particle locations (x-, y- and z- coordinates) from the release point to the sink where it leaves the saturated zone are stored~~, for example, through the unsaturated zone, through the river network or the model boundaries.~~. The particle tracking calculations in Mike SHE are applied to a pre-calculated flow field. Hence, in the first step, the water movement calculation is performed, while in the second step, the tracing of particles, from a source point to a sink, is executed. This method allows for long-term transport calculations where the particle tracking ~~simulations~~ can be run for several annual cycles based on the same, transient or steady-state, flow field. ~~The porosity of the regolith and the bedrock were added to drive the~~The only complementary input data needed to run particle tracking ~~calculations~~is porosity values (Table ~~2).~~3), which was added to the Mike SHE model set up.

Table ~~2~~

3: Porosity values for different soil types used in the Mike SHE model.

Soil type	Porosity
Gravel[a]	0.32
Sand[b]	0.35
Silt[c]	0.45
Clay[b]	*0.55
Silt-clay[d]	0.50
Till[b]	0.30
Peat[b]	0.50
Bedrock[b]	0.0001
Bedrock fractures/deformation zones[b]	0.001

[a]Average of Morris and Johnson. (1967). [b]Joyce et al. (2010). [c]Average value between sand and clay. [d]Average value between silt and clay

2.3 ~~Numerical method~~

~~Particle tracking was used to assess travel times for each sub-catchment. The particle tracking was run to simulate several years to capture the travel times of most of the released particles in the area. One year of calculated flow~~Testing model results ~~from Jutebring Sterte et al. (2018) was cycled multiple times to extend the particle tracking simulation for several years. The year 2010 was selected, as the precipitation and evapotranspiration data for this year were close to the long-term annual averages observed for the Krycklan catchment (approximately 600 mm and 300 mm, respectively (Laudon et al., 2013)).~~

~~The number of particles had to be restricted due to numerical constraints. Particles were released at the top of the transient groundwater table during the first year. We released one particle per cell per 5 mm average modeled groundwater recharge to capture the timing of general recharge patterns, i.e., ca 24,000 particles per km² and year. The time it took for particles to reach a~~against stream ~~or lake (onwards called 'travel time') was calculated for each sub-catchment. Simulated travel time distributions were analyzed using five statistical measures, the arithmetic mean, the geometric mean, the median, the standard deviation, and the skew. If the~~

~~**standard deviation is higher than half of the arithmetic mean, the geometric mean may be a better measure of the central tendency of a data set (Taagepera, 2008), and this can often be the case with travel time distributions with long tails. The standard deviation and skew were therefore used to evaluate which measure of central tendency was best for describing the simulated travel times. To identify the minimum simulation time needed for robust travel time estimates, we compared simulated median travel times for varying lengths of simulations. We assumed that the calculation was run for enough time when the median of the travel time was stabilized for all sub-catchments. The median stabilized after 500 years of simulation time, but in the end, we let the simulation run in total 1000 years to ensure that the results were stable for all parts of the catchments. Thereafter, we used the entire simulation (the year 2010 cycled 1000 times) to calculate mean travel times for each sub-catchment.**~~**chemistry**

~~2.3.1 Observations of stable isotopes~~This study was focused on three distinct seasons in Krycklan: winter, spring, and summer. For calculations of seasonal chemistry, the hydrograph for each site was used. Winter occurs from late November to early April and has been distinguished in the hydrograph as the latest date of new unfrozen precipitation input until the first spring snowmelt. The spring typically occurs in late April to early May, and summer has been assumed to occur from July to September. June and October were excluded because, hydrologically, they are considered transition months between the three distinct seasons. The snowmelt can still influence the runoff in June, and winter conditions (snowfall, soil frost, etc.) can sometimes begin to establish in October.

Stable water isotopes are often used to track pathways of precipitation inputs to a stream network. In this study, ~~we used a~~ time series of stable isotopes ~~(,~~ δ^{18}O ~~and δ^{2}H)~~(see Appendix for the δ^{18}O definition), in stream water were used to compare to ~~modeled~~modelled travel times (Peralta-Tapia et al., 2014). Water was collected at 13 of the 14 sub-catchments included in the study. Hydrological patterns emanating from differences in the landscape structure can be seen in the isotopic composition of stream and groundwater (Ala-aho et al., 2017).

~~In this study, we compared the isotopic signature of winter baseflow, defined here as streamflow from December to February, to calculated winter groundwater travel times. During this season, the primary input to stream water comes from the groundwater due to the prolonged freezing temperatures at these times. In Krycklan, the winter is usually much longer than that, but since even minor melting episodes can have an influence on the isotopic composition and the chemistry of the stream water, only December-February were considered. Therefore, the isotopic stream signature of these months was assumed to describe the isotopic signature of the groundwater the best. The average isotopic signatures of approximately ten years of field observations (the year 2008 to the spring of 2018) were calculated, which consists of approximately 35 measurements from each sub-catchment. Parts of the dataset has been published by Peralta-Tapia et al. (2016), where sampling and analyses are described in detail, and it has since been expanded using the same methodology. We used the average of the stable isotope signature from these years as a representation of baseflow. These averages were also compared to the average (weighted average calculated by precipitation amount) of the long-term precipitation, calculated using approximately 1160 precipitation measurements of both δ^{18}O and δ^{2}H between 2007 and 2016.~~

~~The underlying assumption in this approach is that the strong seasonal signature from precipitation will be reduced with travel time due to mixing in the soil. With infinitely long travel times, the stream water signature should equal the long-term average of precipitation inputs, while short travel times should make the stream water signature reflect the input signal from the precipitation. There should, therefore, be a significant relationship between the simulated travel times and the observed winter isotopic stream signature provided that the model performs well.~~ Some of the sub-catchments are~~, however,~~ affected by evaporation from lake surfaces that result in isotopic fractionation (Leach and Laudon, 2019). ~~This fractionation~~These fractionations must be accounted for ~~in order~~ to use the signature as a representation of the groundwater. The isotopic composition was corrected ~~with respect~~ to the percentage of lakes in each sub-catchment, and a regression equation for ~~each isotope~~$\delta^{18}O$ was determined and applied to sub-catchments containing lakes. We used the same principle as in Peralta-Tapia et al. (2015) but adjusted it for newly acquired data ~~with approximately 270 samples from each site (2008-2018).~~. The long-term regression ~~equations~~equation for ~~each isotope~~$\delta^{18}O$ lake adjustment for sub-catchments are as follows:

~~$18O = 0.18(lake\ coverage\ [\%]) - 13.20\ (r^2 = 0.87,\ p < 0.001)$ Eq. (1)~~

~~$2H = 0.81(lake\ coverage\ [\%]) - 96.03\ (r^2 = 0.68,\ p < 0.001)$ Eq. (2)~~

~~2.3.2 Observations of stream chemistry~~

~~Comparisons were also made to the long-term annual and winter averages of stream chemistry.~~

$$\delta^{18}O=0.18(\text{lake coverage } [\%])-13.20\ (p<0.001,\ R^2=0.87) \qquad \text{Eq. (1)}$$

The comparison of the modelling results to observations of $\delta^{18}O$ was based on a conceptual understanding of the seasonal variability of $\delta^{18}O$ in precipitation and runoff (Fig. 2a). In spring, studies have shown that the young water fraction can be distinguished by comparing the dilution of the isotopic signature to the previous winter because the snow has a much more depleted signal (Laudon et al., 2007; Tetzlaff et al., 2009; Tetzlaff et al., 2015). The difference between winter and spring signature is referred to as the $\Delta\delta^{18}O_{spring}$ (Eq. (2)):

$$\Delta\delta O^{18}_{spring/summer} = \text{average } (Wn - Sn) \qquad \text{Eq. (2)}$$

Wn= Winter isotopic composition average of year n

Sn= Spring/summer isotopic composition average of year n

The more negative $\Delta\delta^{18}O_{spring}$ becomes, the larger the young water fraction is. Hypothetically, the same pattern should be distinguishable in summer but reversed. In summer, the precipitation is enriched compared to the winter signal, which in turn gives younger water an enriched isotopic signal. There should, therefore, be a positive relationship between the young water fraction and the $\Delta\delta^{18}O_{summer}$. In wintertime, there is no infiltration, whereby the isotopic signature can be directly related to the age of the groundwater. The closer the signature comes to the long-term precipitation average (which is equal to the measurements from the deep groundwater in Krycklan), the more well-mixed and, consequently, the older the water should be.

Other indicators of stream water age are base cation (BC) concentration (Fig. 2b). Previous attempts to follow the chemical development of groundwater in the Krycklan catchment have shown that ~~pH and~~ the BC concentration ~~of base cations increase~~increases along the groundwater flow pathway because of weathering (Klaminder et al., 2011). Therefore, a general agreement between the concentration of ~~base cation and pH~~BC on the one hand and ~~modeled~~modelled travel times ~~on the other would strengthen the credibility of the model results. pH is generally expected~~should be possible to ~~increase with the groundwater age since protons are consumed in the weathering of silicate minerals. In addition, the decomposition of organic acids over time will also increase the pH.~~distinguish. The ~~base cations are~~ BC is mainly derived from the weathering of local soils in the Krycklan catchment, with only a minor contribution from atmospheric deposition (Lidman et al., 2014).

~~Modeling~~Modelling of weathering rates in a soil transect in the Krycklan catchment has indicated that there is kinetic control of the release of ~~base cations~~BC in the soils (Erlandsson et al., 2016). The release of ~~base cations~~BC suggests that the longer the groundwater is in contact with the mineral soils, the higher ~~base cation~~BC concentrations can be expected, similarly to what was observed by Klaminder et al. (2011). Since ~~base cations have been observed~~BC are expected to behave relatively conservatively in these environments (Ledesma et al., 2013; Lidman et al., 2014), ~~we used~~ their combined concentration was used as a proxy for water age~~, i.e., subareas~~. Sub-catchments with longer travel times would ~~hypothetically~~ exhibit higher ~~base cation~~BC concentrations ~~and higher pH.~~. It has been observed, however, that mires have a ~~negative~~significant impact on the concentration of cations in the streams within the Krycklan catchment. The reason is that the peat does not contain any appreciable amounts of minerals, so groundwater passing through mires will not acquire cations ~~at~~to the same ~~rate~~amounts as when it passes through mineral soils (Lidman et al., 2014). In practice, this will cause cations in specific subareas to be diluted by groundwater from the mires in a manner that is not related to the groundwater travel time. The cation concentrations were therefore adjusted for the influence of mires, according to~~:~~ Eq. (3):

Adjusted cation concentration = Observed cation concentration/ (1-fraction of mire coverage)   Eq. (3)

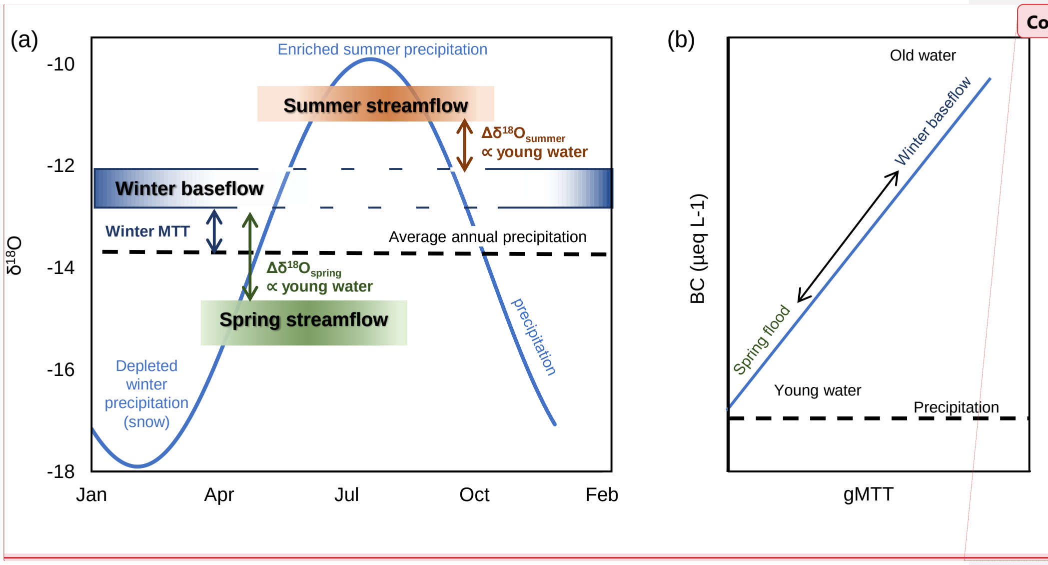

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Figure 2: Conceptual figure of connection between water travel time and stream chemistry. (a) The connection between $\delta^{18}O$ and stream water travel time. The sine curve shows the annual variations of $\delta^{18}O$ precipitation composition, and approximate seasonal winter, spring, and summer stream composition are marked. In winter, older travel times are proportional to winter baseflow isotopic signature closer to the long-term precipitation average. In spring, a greater fraction of young water is proportional to a greater difference between the spring (snowmelt) signature and the winter baseflow signature (negative sign). In summer, a greater fraction of young water is proportional to a larger difference between the summer signature and winter baseflow (positive sign). (b) The connection between base cations (BC) and travel time. The older the mean travel time, the higher concentrations of BC due to weathering.

All stream chemistry data comes from the online open Krycklan database at www.slu.se/Krycklan (Table 4). The isotopic signatures contain approximately ten years of field observations (2008 to mid-2018), approximately 25 samples per year for each site. A small part of the dataset has been published by Peralta-Tapia et al. (2016), where sampling and analyses are described in detail, and it has since been expanded using the same methodology. We used the average of the stable isotope signature from these years as a representation of baseflow.

$$\cancel{Adjusted\ cation\ concentration = Observed\ \ cation\ concentration / (1 - mire\ coverage\ )}$$

~~Eq. (3)~~

These averages were also compared to the volume-weighted average of the long-term precipitation, calculated using approximately 1160 precipitation measurements of $\delta^{18}O$ measured between 2007 and 2016. The long-term precipitation average is -13.5 ‰, which is equal to observations of the isotopic signature at the deep groundwater wells of Krycklan (10 m depth). The BC data collection methodology is reported in Ledesma et al. (2013).

Table 4: Seasonal stream chemistry.

~~All pH and base cation data were taken from the open Krycklan database, and the collection methodology and analysis are reported in Laudon et al., 2013. The base cation and pH data comprise approximately 25 (2008-2017) and 20 (2011-2018) observations for the winter period (December to February) for each stream, respectively (Table 3, an extended table can be found in Supporting information 1). Since pH and base cations are less impacted by precipitation, compared to water isotopes, the annual average was also considered. The annual base cation and pH data comprise approximately 235 (2008-2017) observations and 180 (2011-2018) observations, respectively.~~

~~Table 3: Stream chemistry of Krycklan in order of catchment size~~

	δ^{18}O[a]						Base cations (BC)[b]					
	Winter		Spring		Summer		Winter concentration		Spring concentration		Summer concentration	
	‰	SD/SEM[c]	Δδ^{18}O	SD/SEM	Δδ^{18}O	SD/SEM	µeq/L	SD/SEM	µeq/L	SD/SEM	µeq/L	SD/SEM
C1	-12.9	0.28/0.05	-0.53	0.60/0.18	0.10	0.38/0.19	283	39/7	211	36/5	285	31/4
C2	-12.9	0.46/0.07	-0.68	0.52/0.16	0.15	0.45/0.16	288	104/21	174	41/6	267	58/9
C4	-13.1	0.36/0.06	-1.08	0.66/0.20	0.82	0.48/0.21	295	77/17	107	33/5	306	77/12
C5	-13.0	0.47/0.08	-1.80	0.66/0.20	0.72	0.65/0.21	273	27/6	172	49/9	231	34/5
C6	-13.1	0.35/0.06	-1.27	0.55/0.16	0.52	0.47/0.17	364	80/16	230	133/19	322	120/16
C7	-13.0	0.22/0.04	-0.73	0.56/0.17	0.42	0.37/0.18	290	43/9	177	59/9	270	38/5
C9	-13.1	0.29/0.05	-0.98	0.46/0.14	0.57	0.44/0.15	385	61/12	219	87/13	327	61/8
C10	-13.3	0.28/0.05	-0.80	0.61/0.18	0.53	0.39/0.19	348	58/12	200	104/16	332	72/10
C12	-13.1	0.30/0.05	-0.88	0.48/0.15	0.36	0.43/0.16	349	48/10	187	40/6	316	45/6
C13	-13.1	0.26/0.05	-0.83	0.55/0.16	0.60	0.48/0.17	379	57/12	203	37/5	309	43/6
C14	-13.4	0.23/0.04	-0.70	0.55/0.17	0.48	0.45/0.18	388	46/10	264	85/12	376	74/10
C15	-13.4	0.40/0.07	-0.73	0.69/0.21	0.63	0.44/0.22	373	44/9	233	41/6	349	45/6
C16	-13.4	0.44/0.08	-0.56	0.64/0.64	0.46	0.33/0.20	511	56/11	251	53/8	441	76/10
C20	-	-	-	-	-	-	582	80/17	348	48/7	526	60/8
Long term precipitation average												
	-13.5 ‰ [d]						80 µeq L^{-1} [d]					

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[a] ~~Average measured winter isotope~~δ^{18}O Signature (2008-2018), ~~sub-catchments with lakes~~data have been lake adjusted according to ~~Eqs. 1~~equation 2, and ~~2, respectively.~~delta was calculated using Eq. (1).
[b] ~~Average measured winter and yearly~~Base cation ~~signature (2011-2018), sub-catchments~~concentration (2008-2016), data have been mire adjusted according to ~~equation~~ Eq. (3)
~~[c] Average measured winter and yearly pH signature (2008-2017)~~

[c]SD = standard deviation, SEM = standard error of the mean
[d] Measured precipitation average for isotopes (2007-2016) and measured ~~base cations (~~BC concentration (the year 1997 to ~~2003~~2018)

2.4 Establishing travel times - Particle tracking

Particle tracking was used to assess travel times for each sub-catchment. The model was run 1000 years to capture the travel times from source to sink of most of the released particles in the area. One year of simulated flow results from Jutebring Sterte et al. (2018) was cycled multiple times to extend the particle tracking for 1000 years. The year 2010 was selected, as the water balance for this year was close to the long-term annual averages observed for the Krycklan catchment. The number of particles released had to be restricted due to numerical constraints, and particles were released at the top of the transient groundwater table during the first year. Approximately 0.5 particles/10 mm modelled groundwater recharge was released to capture the timing of recharge patterns (Fig. 3).

The time it took for particles to reach a stream or lake via groundwater (hereafter called 'travel time') was calculated for each sub-catchment. The calculated travel time distributions were analysed using five statistical measurement tools, the arithmetic mean, the geometric mean, the standard deviation, the standard error of the mean, and the skew

(Appendix). If the standard deviation is higher than half of the arithmetic mean, the geometric mean is a better measure of the central tendency of the data set (Taagepera, 2008). The geometric mean is defined as the back-transformed arithmetic mean of the log-transformed data. The standard deviation and skew were therefore used to evaluate which measure of central tendency was best for describing the simulated travel times. To identify the minimum particle tracking time needed for robust travel time estimates, we compared median travel times for varying lengths of particle tracking. We assumed that the calculation was run for enough time when the median of the travel time was stabilized for all sub-catchments. ~~2.3.3~~The median stabilized after 500 years of simulation time, but in the end, we let the particle tracking run in total 1000 years to ensure that the results were stable for all parts of the catchments. Thereafter, we used all particles that reached a stream or lake to calculate mean travel times for each sub-catchment.

During winter, all simulated streamflow contribution comes from the groundwater. Here the particle tracking reflects the actual travel time to the streams. However, during summer and especially during spring, some water will reach the streams as overland flow, and therefore has spent zero days in the ground. Since the particle tracking does not take surface flow into account, two travel times were calculated for each site. The first is the groundwater age directly based on the particle tracking results (groundwater gMTT), and a second version where the surface flow component was assumed to have a very young age (zero days), which can be interpreted as the total time stream water contribution have spent in the ground (overall gMTT). To reduce the travel time according to calculate the overall travel time, we used Eq. (4).

$$\text{Overall gMTT} = \text{groundwater gMTT} * (1 - \text{fraction overland flow}) \qquad \text{Eq. (4)}$$

The young water fraction metric was also used as an evaluation criterion. Similar to previous studies (Kirchner., 2016; von Freyberg et al., 2018; Lutz et al., 2018; Stockinger et al., 2019), we assumed young water to be the sum of water reaching streams as overland flow and groundwater with age less than three months.

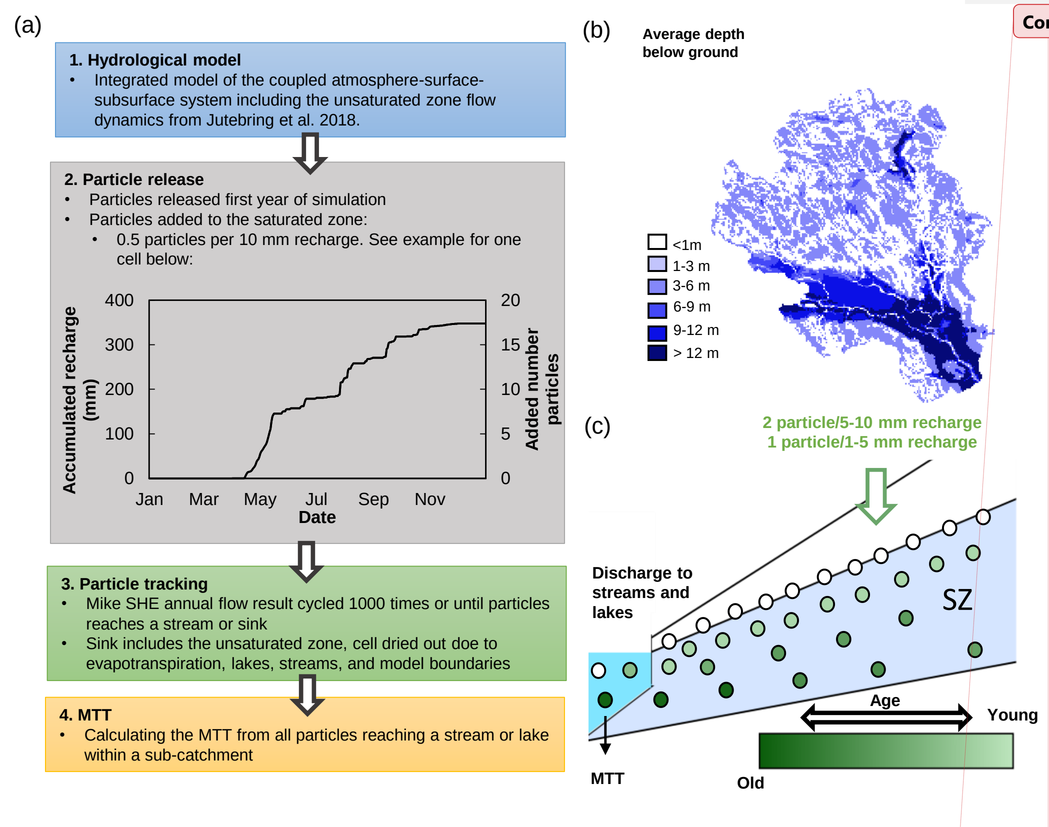

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Figure 3: Particle model setup. (a) Step by step of the particle tracking procedure. (b) Average depth to the groundwater table. The main part of the model area has a calculated depth to the groundwater table between 0-3 m and varied on a daily basis. Note that the top vertical layering of the saturated zone was set to 2.5 m below the ground surface, and the thickness thereafter follows the soil layers (thickness increasing with depth). The horizontal grid-size used was 50*50 m. (c) Schematic illustration of particle tracking set up. Particles were added to each cell at the transient groundwater table. The age of these particles was zero at the time of recharge. The particles then followed the groundwater flow while increasing in age. All particles that reach a stream or lake receives an end age, which is equal to the time from recharge to discharge in the stream. MTT is calculated for each stream using the age of all particles reaching it.

2.5 Catchment characteristic investigation

~~To test the hypothesis that the catchment size is the primary factor affecting the groundwater travel time, the~~ Correlations between the calculated ~~MTT~~seasonal gMTT and different catchment characteristics were ~~investigated.~~established to identify the main factors that affect the travel time of water to streams. The young water fraction was also tested against catchment characteristics. The characteristics tested included important terrain factors such as size and slope as well as soil types. As many factors can affect the hydrology of a catchment, ~~we list~~ the ~~main~~most important descriptive physical landscape characteristics are listed in Table 1~~,~~ (from Karlsen et at. 2016), which together describe much of the landscape variability of Krycklan. ~~Karlsen et al. (2016) also suggested that these factors are some of the most important landscape characteristics affecting the hydrology of the catchment. The simulated specific baseflow to the streams, as well as the mean travel distance (MTD) of the particles, were also calculated to investigate if they could help explain the travel time patterns in the landscape (Table 4).~~

3 ~~**Results**~~. Result

~~**3.1 Estimation of mean travel time**~~

~~**The simulated yearly mean groundwater travel time (MTTy) for all sub-catchments ranged between 0.5 to**~~ **3.**~~**6 years. The geometric mean**~~**1 Travel time results**

The particle tracking model in Mike SHE was used to establish mean travel time in the 14 sub-catchments. The time from groundwater recharge until the groundwater reached a stream was used as an estimation of groundwater travel time. The geometric mean (gMTT) was used to describe the central tendency of travel times~~,~~ because of the skewed distribution (Table ~~**4, extended table in Supporting information 1). The travel time distribution as reflected by the MTTy, with, for**~~ 5, Fig. 4). From the particle results, the calculated annual groundwater and overall gMTT for all sub-catchments ranged between 0.8 to 3.1 years and 0.8-2.7 years, respectively (Table 5). Most particles had a travel time of less than one year (34 to 54 %). The older groundwater was connected to the larger sub-catchments and sub-catchments with fluvial sediments of C13, C14, C15, C16, and C20. Particles with old ages were generally connected to deep and long flow pathways.

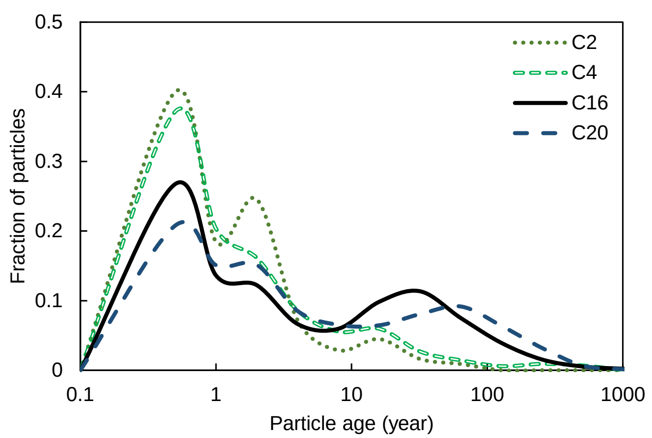

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Figure 4: Particle tracking results. The figure shows the timing of particles reaching the sub-catchment outlet. The figure shows four examples, including C2 (forest dominated sub-catchment), C4 (mire dominated sub-catchment), C16 (Krycklan as a whole), and C20 (silt dominated sub-catchment). The same example~~, C2 having the youngest~~

~~mean age and the largest proportion of young particles. In comparison, C20 had the oldest age and the largest proportion of old particles (Table 4, Fig. 2).~~ sub-catchments are shown in Fig. 5.
~~Over~~
On an annual basis, a ~~year, a small~~ fraction of the water ~~reaches the~~reached a stream as ~~surface~~overland flow, which may enhance or dilute various stream solutes in different ways. ~~Winter baseflow conditions may, therefore, be a better representation~~The major part of the ~~groundwater chemistry. From December to February, there was no input of precipitation due to freezing conditions, resulting in that the only input to the streams came from the groundwater. The mean simulated travel time of winter baseflow (MTTw) was older for all~~ overland flow occurs during the snowmelt in April to May, especially in sub-catchments ~~compared to MTTy. According to the simulation, winter baseflow (Dec-Feb) accounts for approximately~~with mires such as C4 (Fig. 5 ~~15% of the total yearly stream contribution.~~

~~**Table 4: Particle tracking results for all sites in Krycklan.** Statistics of particle tracking results with a simulation time of 1000~~). Each site has the oldest age during the winter season (1.2-7.7 years~~. The table is ordered by increasing sub-catchment size. The statistics are calculated for each sub-catchment~~) and ~~include~~the youngest age in spring and summer (0.5-1.9 years). The input of new water is also reflected in the seasonal groundwater gMTT. The groundwater is youngest in connection with the snowmelt during late spring, then increases during the ~~MTT (MTTy is~~ summer period with little groundwater recharge (Jun-Jul). The oldest groundwater travel times occur during the ~~yearly mean travel time, and MTTw is the winter (Dec-Feb) mean travel time), skew, and standard deviation (SD). Further statistical information can be found in Supporting information 1. MTD is the mean travel distance of the particles. Winter baseflow is the fraction of the total annual runoff generated during Dec-Feb.~~winter, before the beginning of snowmelt in late March or early April.

Mire sub-catchments have the youngest mean travel time during spring snowmelt. However, as exemplified by the similar-sized sub-catchments of C2 (forest) and C4 (mire), groundwater is not renewed to the same extent in mires as in forested sub-catchment (Table 5). The groundwater gMTT of C2 was reduced from 1.2 years to 0.7 years from winter to spring. In C4, groundwater gMTT was reduced from 1.5 years to 1.2 years, despite a larger young water fraction. The overall gMTT of C4 decreased even more, from 1.5 years to 0.7 years. A more pronounced seasonality in mean travel times also occurs for catchments with a larger proportion of mires combined with low conductive soils (LCS). For example, C20 had an overall gMTT that reduced from 7.7 years to 1.9 years from winter to spring, while the overall gMTT of the similar-sized till sub-catchment C6 only changed from 2.8 to 0.6 years.

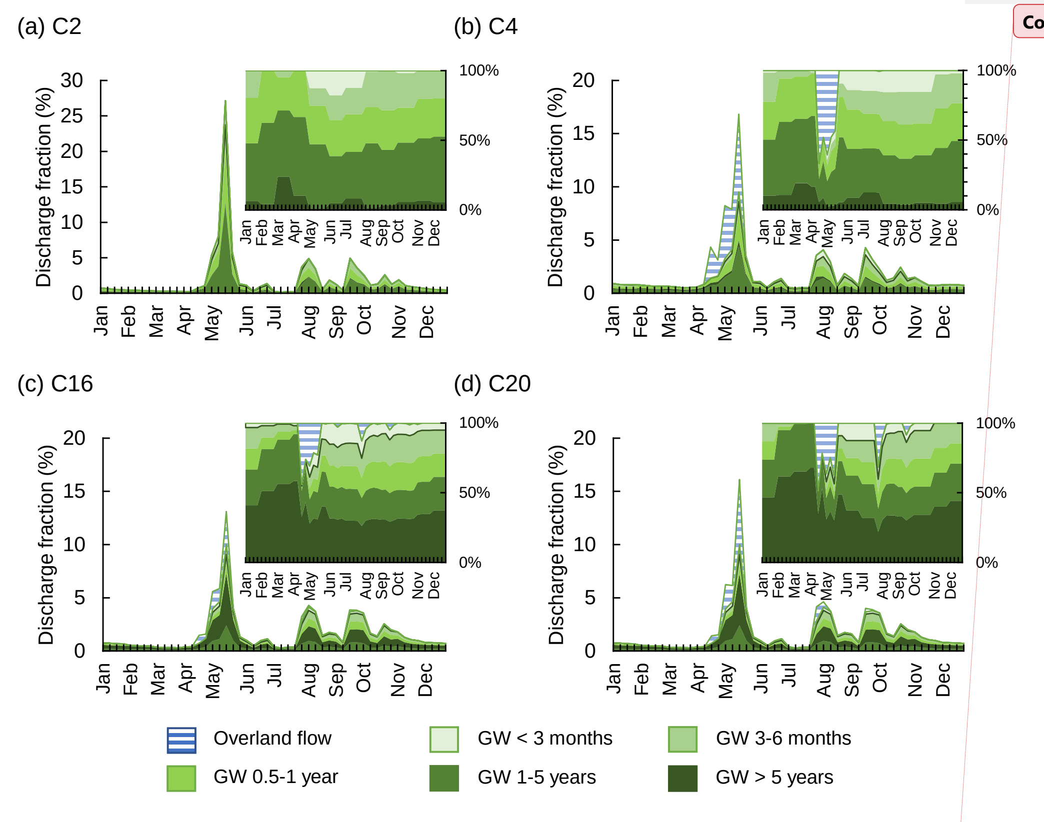

Figure 5: Seasonal fraction of runoff to streams. The figure shows the proportion of stream water arriving as groundwater flow and as direct overland flow. Four sub-catchments are exemplified, including (a) the small forested C2, (b) the mire-dominated C4, (c) the entire Krycklan catchment C16, and (d) and the silt-rich C20 (extended version in Appendix Fig. A1).

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Table 5: Annual and seasonal (winter, spring, and summer) travel time results.

	Annual								Season - Winter							
	aMTT	Skew	SD	SEM	gw gMTT	OL	gMTT	Yf	aMTT	Skew	SD	SEM	gw gMTT	OL	gMTT	Yf
unit	year	-	-	-	year	%	year	%	year	-	-	-	year	%	year	%
C1	10.1	4.1	27	0.4	1.3	0	1.3	20.2	18.8	2.7	36	1.2	3.0	0	3.0	5.6
C2	2.2	5.0	5	0.2	0.8	0	0.8	16.3	2.7	2.4	4	0.5	1.2	0	1.2	0.0
C4	7.7	7.5	34	1.0	1.0	21	0.8	39.5	10.5	6.6	42	2.7	1.5	0	1.5	2.1
C5	15.2	6.4	61	1.0	1.3	35	0.8	49.1	30.4	4.1	84	3.2	2.9	0	2.9	1.1
C6	13.7	6.8	51	0.6	1.2	23	0.9	41.5	25.9	4.6	69	1.8	2.8	0	2.8	1.9
C7	8.0	7.4	25	0.4	1.2	8	1.1	27.8	13.2	5.6	32	1.1	2.2	0	2.2	3.5
C9	13.2	6.7	38	0.3	1.6	12	1.4	28.0	21.6	5.0	47	0.7	3.4	0	3.4	2.9
C10	10.9	6.4	35	0.2	1.2	14	1.1	32.8	16.5	4.5	40	0.6	2.5	0	2.5	3.2
C12	11.9	5.5	33	0.2	1.4	9	1.3	27.5	17.6	4.0	37	0.4	2.8	0	2.8	5.2
C13	13.3	8.0	43	0.2	1.5	9	1.4	25.8	21.6	6.4	53	0.5	3.3	0	3.3	3.1
C14	18.3	7.8	54	0.2	2.7	9	2.4	19.7	26.4	6.8	60	0.4	5.6	0	5.6	1.5
C15	14.3	8.7	43	0.1	1.7	10	1.5	28.2	21.9	6.7	49	0.3	3.8	0	3.8	4.0
C16	17.4	8.5	50	0.2	2.5	8	2.3	22.8	25.3	7.3	57	0.2	5.3	0	5.3	3.5
C20	21.9	6.0	52	0.6	3.1	13	2.7	22.9	32.9	5.8	55	1.1	7.7	0	7.7	0.0

	Season - Spring								Season - Summer							
	aMTT	Skew	SD	SEM	gw gMTT	OL	gMTT	Yf	aMTT	Skew	SD	SEM	gw gMTT	OL	gMTT	Yf
unit	year	-	-	-	year	%	year	%	year	-	-	-	year	%	year	%
C1	5.2	6.8	19	0.5	1.0	0	1.0	24.9	8.4	4.5	25	0.8	0.9	0	0.9	18.6
C2	1.6	6.1	3	0.2	0.7	0	0.7	25.5	2.7	4.2	8	0.6	0.7	0	0.7	6.0
C4	5.7	10.8	27	1.5	1.2	44	0.7	52.7	9.1	6.1	39	2.0	0.7	0	0.7	39.2
C5	9.9	9.3	50	1.5	1.2	57	0.5	65.6	11.3	7.6	52	1.7	0.8	8	0.8	38.4
C6	9.1	9.5	45	1.0	1.0	42	0.6	58.0	9.9	8.2	42	1.0	0.8	7	0.8	33.5
C7	5.5	11.2	19	0.6	1.1	18	0.9	36.6	7.5	7.5	27	0.8	0.9	0	0.9	26.9
C9	8.2	10.8	31	0.4	1.3	24	1.0	40.7	11.2	7.0	34	0.5	1.2	5	1.1	24.3
C10	8.0	8.2	32	0.4	1.1	31	0.8	46.6	9.0	6.3	31	0.4	0.9	0	0.9	30.5
C12	8.2	7.6	29	0.3	1.2	21	0.9	38.5	10.2	5.3	29	0.3	1.1	1	1.1	26.1
C13	7.8	10.8	30	0.3	1.2	18	1.0	36.7	12.5	8.8	45	0.4	1.2	3	1.2	23.1
C14	12.2	10.6	45	0.3	2.1	21	1.6	32.0	16.3	7.9	54	0.4	1.8	11	1.6	20.5
C15	9.2	11.4	34	0.2	1.2	22	0.9	41.4	12.4	9.2	41	0.2	1.2	9	1.1	27.3
C16	11.4	11.0	40	0.1	1.7	20	1.4	35.1	15.6	8.9	48	0.2	1.8	11	1.6	22.9
C20	12.2	10.5	39	0.8	2.6	27	1.9	35.8	20.4	5.4	59	1.3	1.6	9	1.5	24.3

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~~**Figure 2 Simulated travel time distribution of the groundwater in Krycklan.** C16 is used as a visual reference in all panels. The figure includes all 14 investigated sub-catchments, color-coded as Fig. 1 and displayed in the legend in size order from left to right with C2 being the smallest and C16 the largest sub-catchment. The figure is divided into (a) all sub-catchments, (b) the sub-catchments of C13, (c) the sub-catchments of C12 and, (d) the sub-catchments of C14 and C15.~~

~~3.2 Stable isotopes and stream chemistry~~

~~The simulated winter mean travel times (MTTw) were compared to the measured winter isotope signature for each site, as well as to the measured average winter cation concentration and pH, using linear regressions (Fig. 3). There was a significant correlation between the calculated mean winter travel times and both δ^{18}O (r=0.90, p<0.001) and δ^{2}H (r=0.90, p<0.001). Both δ^{18}O and δ^{2}H decreased with travel time, approaching the long-term precipitation average of -13.46 ‰ and -99.88 ‰, respectively. There was also a significant correlation between the measured winter base cation concentration and the simulated travel times (r=0.88, p<0.001; Fig. 3). pH had a similar behavior, but the correlation was somewhat weaker (r=0.73, p<0.001). The main outliers were the mire-dominated sub-catchments C4 and C5, which have high concentrations of organic acids that influence pH negatively. Note that there are isotope data for 13 sub-catchments and chemistry data for 14 sub-catchments (isotope data excludes C20, see Table 3). The yearly mean travel times (MTTy) were also compared to the yearly average of base cations and pH, with significant results for both pH (r=0.83, p<0.001) and base cations (r=0.90, p<0.001).~~

~~**Figure 3: Linear regressions of stream chemistry and calculated geometric mean travel times (MTT).** The black line is the regression line, and the green lines are the 95% prediction bands. The plots also show the SEM (standard error of the mean) of the calculated average travel time and the chemistry observations (see Supporting information 1). (a) and (b): Average winter isotope signature, δ^{18}O, and δ^{2}H (‰), against MTTw. Here the long-term average of precipitation signature is shown as a horizontal blue line in each graph (-13.46 δ^{18}O and -99.88 δ^{2}H for the years 2007-2016). (c): Average winter base cation concentration (µeq/ L, mire adjusted according to table 3 and eq. 3) against MTTw. (d): Average winter pH against MTTw. E: Average yearly base cation concentration (µeq L^{-1}, mire adjusted according to table 3 and eq. 3) against MTTy. F: Average yearly pH against MTTy.~~

~~3.3 Catchment characteristics~~

~~There was no significant correlation between sub-catchment size and calculated mean annual travel time (MTTy) (Fig. 4), C20 being the main outlier. Furthermore, when comparing the MTTy to other catchment characteristics, there was no significant correlation to the proportion of mires, till or lake area. However, there were significant (although weak) correlations between MTTy and mean travel distance (MTD) (r=0.76, p<0.01) and MTTy and catchment slope (r=0.73, p<0.01). A strong significant correlation between the MTTy and the proportion of low conductive sediments (LCS) (r=0.90, p<0.001), was also found. By using multiple regression, two simple relationships could be established between the sub-catchments and three characteristics (further description of this relationship can be found in Supporting information 2). These show that although there is a correlation between the MTTy on one hand and slope (Eq. 4) or MTD (Eq. 5) on the other hand, the most significant parameter is the LCS:~~

$$MTT\ (y) = -0.33 + 0.31 * Slope[°] + 0.09 * LCS[\%]\ (R^2 = 0.90\ , p < 0.001) \qquad \text{Eq. (4)}$$

$$MTT\ (y) = 0.19 + 0.01 * MTD\ [m] + 0.08 * LCS[\%]\ (R^2 = 0.90, p < 0.001) \qquad \text{Eq. (5)}$$

~~**Figure 4: Linear regression of catchment characteristics and calculated mean annual travel times (MTTy).**~~ ~~The black line is the regression line, and the green lines are the 95% prediction bands. The plots also show the SEM (standard error of the mean) of the simulated average travel time (see Supporting information 1). (a): Catchment area against MTTy. (b): Mean travel distance of particles (MTD) against MTTy (c): Catchment slope (°) against MTTy. (d): Low conductive sediments (LCS) (%) against MTTy.~~

aMTT = arithmetic mean (year), **SD** = Standard deviation, **SEM** = standard error of the mean, **OL** = fraction overland flow (%), **gw gMTT**= geometric mean of the particle tracking (groundwater gMTT) (year), **gMTT** = geometric mean of the particle tracking adjusted for overland flow according to Eq. (4) (overall gMTT) (year), **Yf** = fraction of surface flow and groundwater less than three months (%) (Supporting information)

3.2 Testing model results to stream chemistry

Three distinct seasons were evaluated with regards to the stream chemistry: winter, spring, and summer. The isotopic composition was available for 13 out of 14 sub-catchments (C20 excluded because of short time-series), while the base cation (BC) data was available for all sub-catchments. According to the modelling results, sub-catchments receive older water when the average isotopic composition is closer to the long-term precipitation under winter conditions (Fig. 6a). Some of the larger sub-catchments have a signature close to the long-term precipitation average, suggesting that they have reached complete mixing (C15, C14, and C16). However, the negative correlation is significant ($r=-0.80$, $P<0.01$), with older stream water age being closer to the long-term precipitation average. The negative correlation between the $\Delta\delta O^{18}_{spring}$ and the young water fraction was also significant ($r=-0.90$, $P<0.0001$, Fig. 6c), following our conceptual model (Fig. 2a). Sub-catchments with a larger fraction of young water during the spring displayed a greater dynamic in the isotopic composition of the stream water. The opposite was also true for the summer since the precipitation was enriched compared to the baseflow. The positive correlation was weaker compared to the spring season, but still significant ($r=0.80$, $P<0.001$, Fig. 6e).

The overall gMTT always had a strong statistical significance to the BC concentration (Fig. 6 b, d, and e), generally agreeing with our conceptual model (Fig. 2b). The correlation between BC and gMTT was strongest during winter ($r=0.88$ $P<0.0001$) and weakest during summer ($r=0.79$, $P<0.001$). The sub-catchments with the oldest age and highest BC concentration include some of the largest sub-catchments of C14 and C16, but also C20, which is one of the smaller sub-catchments. These three sub-catchments have the largest portions of fluvial sediment deposits (Table 1). The youngest ages and lowest BC concentrations were connected to smaller sub-catchments in the till areas, such as C2 and C4.

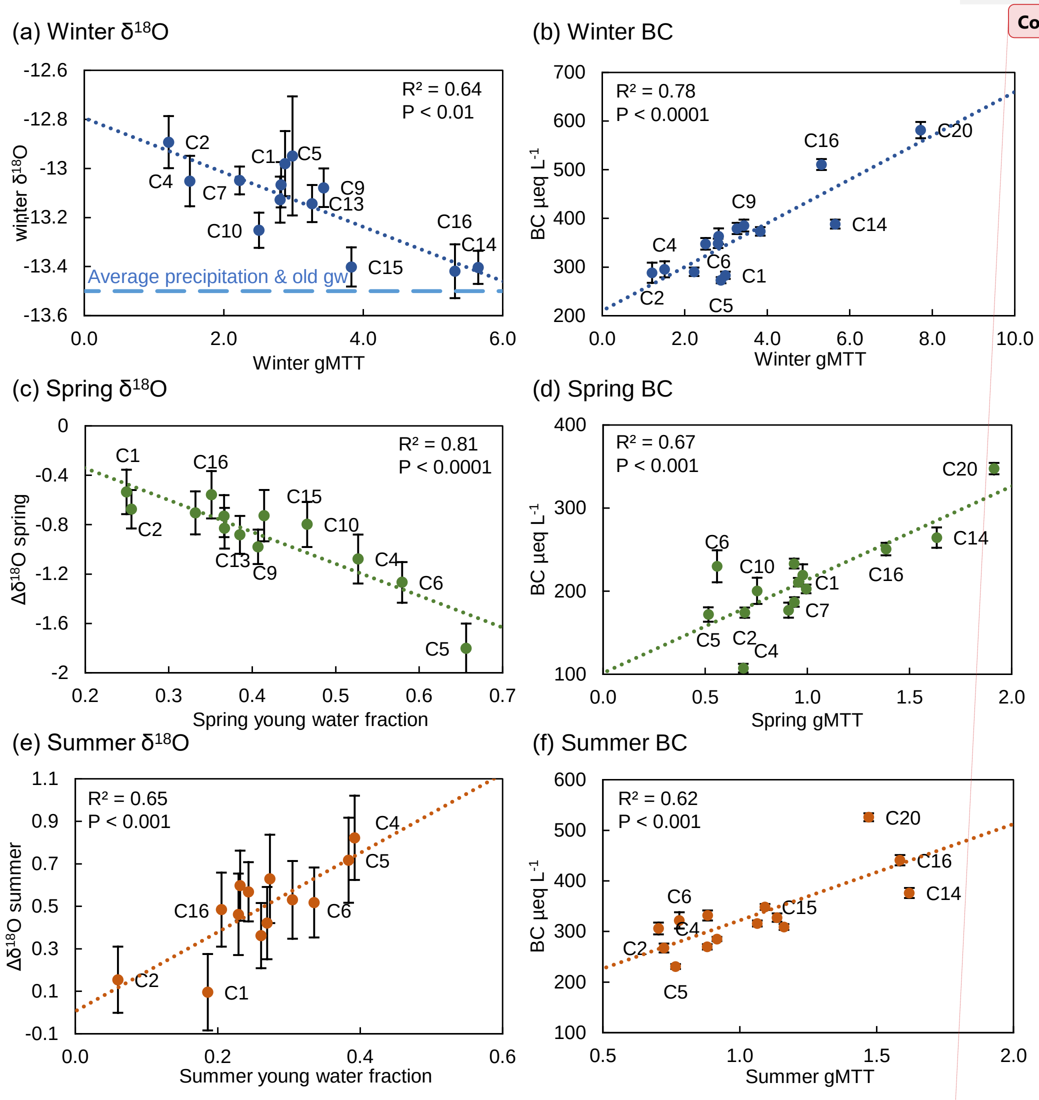

Figure 6: Results of seasonal water fraction compared to stream chemistry, δ[18]O, and base cation (BC) concentration. Note that δ[18]O results are for 13 sites, while the BC record comprises all 14 sites. The sub-plots (a) to

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(f) show the $\delta^{18}O$ (winter) or $\Delta\delta^{18}O_{spring/summer}$ and BC concentrations as a function of the overall gMTT during winter, spring, and summer, respectively. The standard error of the mean (SEM) is shown for the field observations.

3.3 Model results compared to catchment characteristics

The main catchment characteristics found to be correlated to gMTT were catchment size, the fraction of low conductive soils (LCS), and the fraction of mires. The strongest positive correlation was found between the young water fraction and the proportion of sub-catchment mire coverage (r=0.96, P<0.0001), as well as gMTT and low conductive soils (LCS) (r=0.90, P<0.0001) (Figure 7). A larger fraction of mires increases the young water fraction, and a larger fraction of LCS increases gMTT. A positive correlation between catchment size and gMTT was also found. The correlation was relatively weak due mainly to one outlier, C20, yet significant (r=0.63, P<0.05) (Fig. 7). However, the catchment size is also correlated to the fraction of LCS, which may be the underlying reason for this correlation (Table 6) as C20 is the only relatively small monitored sub-catchment located in the sedimentary soil area. The annual and seasonal patterns are generally similar (Table 6). However, the positive correlation between mires and the young water fraction was lost during winter, presumably due to a lack of new precipitation input into the system. The gMTT and the young water fraction correlation followed the pattern of the correlation between gMTT and mires. A weak negative correlation between gMTT and the young water fraction was found in the annual average and during spring but was lost during summer and winter.

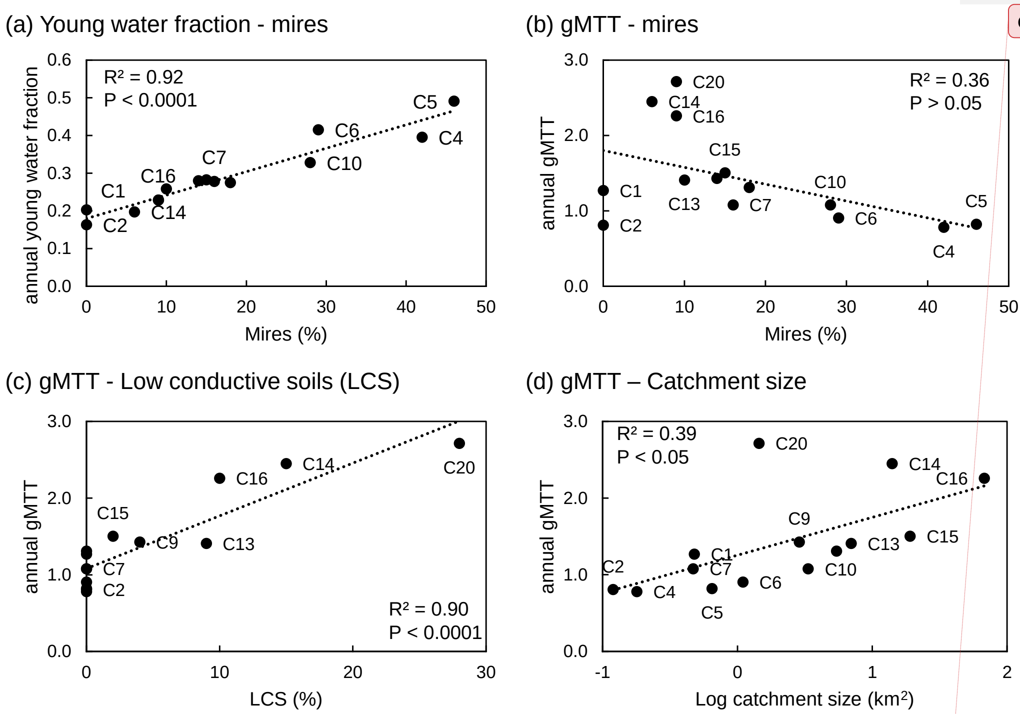

Figure 7: Travel time important catchment characteristics. The figure shows the annual averages. (a) Mires and young water fraction, (b) mires and gMTT (year), (c) low conductive soils (LCS) and gMTT (year), and (d) Catchment size and gMTT (year). The gMTT has been adjusted for the overland flow for each season, according to Eq. (4).

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Table 6: Correlation matrix – young water fractions, gMTT, and catchment characteristics. The table includes yearly calculations (white), winter calculations (blue), spring calculations (green), and summer calculations (orange). Darker colours show when the absolute value of. r > 0.50 with the connected p-value according to [a] p>0.05, [b] a p<0.05, and c p<0.01.

Annual

	Log C.-size	Mire (%)	LCS (%)	gMTT	Yf
Log C.-size	1				
Mire (%)	0.02	1			
LCS (%)	0.58 [b]	-0.37	1		
gMTT (y)	0.63 [a]	-0.51 [a]	0.90 [c]	1	
Yf (%)	-0.02	0.96 [c]	-0.39	-0.53 [a]	1

Season - winter

	Log C.-size	Mire (%)	LCS (%)	gMTT	Yf
Log C.-size	1				
Mire (%)	0.02	1			
LCS (%)	0.58 [b]	-0.37	1		
gMTT (y)	0.64 [b]	-0.34	0.92 [c]	1	
Yf (%)	-0.08	-0.14	-0.43	-0.21	1

Season - spring

	Log C.-size	Mire (%)	LCS (%)	gMTT	Ywf
Log C.-size	1				
Mire (%)	0.02	1			
LCS (%)	0.58 [b]	-0.37	1		
gMTT (y)	0.55 [b]	-0.55 [b]	0.92 [c]	1	
Yf (%)	0.11	0.95 [c]	-0.29	-0.52 [a]	1

Season - summer

	Log C.-size	Mire (%)	LCS (%)	gMTT	Yf
Log C.-size	1				
Mire (%)	0.02	1			
LCS (%)	0.58 [b]	-0.37	1		
gMTT (y)	0.68 [c]	-0.50 [a]	0.80 [c]	1	
Yf (%)	0.20	0.91 [c]	-0.20	-0.28	1

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4 Discussion

~~4.1 Simulated travel times are consistent with the isotopic signal and stream chemistry~~

Particle tracking in the Mike SHE model showed promising results in its ability to capture the travel times across the 14 Krycklan sub-catchments. Travel times of stream water contribution and young water fractions were related to stream winter $\delta^{18}O$ signatures, $\delta^{18}O$ seasonal shifts, and base cation (BC) concentrations. Particle tracking could therefore be a useful complementary tool to tracer-based studies of travel time, especially in snow-dominated catchments, areas with pronounced seasonality, and streams dominated by old groundwater (older than 4-5 years). In this study, we found the hydrologic conductivity of the soil to be the most important parameter for the water age and mires to be an important factor regulating the young water fraction.

4.1 Model testing and uncertainties

Particle tracking in Mike SHE is associated with some uncertainties and limitations. A comparison of the results from this study to previous studies of mean travel times (MTT) for one of the Krycklan sub-catchments (C7) shows, however, that the different approaches gave similar results. While our study estimated a MTT time to 1.1 years, Peralta-Tapia et al. (2016) calculated a ten-year average travel time of 1.8 (minimum 0.8 and maximum 3.3) years using isotopic data and a gamma transformation method. In a study using the same data in the Spatially distributed Tracer-Aided Rainfall–Runoff model, the median of the travel time distribution was approximated to 0.9 years for the same sub-catchment (Ala-aho et al., 2017). The close agreement with the previous model runs strengthen our results.

One limitation of our modelling approach is that particle tracking is restricted to the saturated zone. This restriction is primarily related to the overland flow component, most visible in mire dominated catchments in connection with the spring snowmelt. We accounted for this effect by assuming the age of the overland flow component to be zero days (Eq. (4)). If the age of the water – or its travel time – is the time it spends in the ground, this would be the actual age of the water. Alternatively, one could define the age as the time from when a water unit melted. However, that would add additional uncertainties, and for overland flow, it would most likely still only amount to an additional couple of days in most cases and would likely not influence the overall gMTT to any large extent. Counting the number of days from when the snow fell is not particularly meaningful from a hydrological point of view as the storage of snow in winter can last up to six months.

Another uncertainty in Mike-SHE is related to the travel time from infiltration under the unsaturated condition to groundwater recharge, which, due to technical limitations, was not accounted for in the particle tracking calculations. Particles are placed in the groundwater proportionally to the recharge (Fig. 3). Therefore, the main portion of particles is introduced to the model at high recharge when the groundwater level is shallow across the catchment. However, some particles are introduced when the groundwater level is lower, such as early snowmelt or after extended dry periods. In our simulations, the groundwater table varies between 0-3 m below the ground surface (Figure 3). While mires generally have an average groundwater table above 1 m, till areas range between 2-3 m. C14 is an exception:

here, a deep esker traversing the sub-catchment results in a lower water table than in other Krycklan locations. Accounting for the travel time from infiltration to recharge could impact the results and give especially C14 older MTT than if the groundwater level was at the same level throughout the whole catchment. This limitation primarily affects catchments with long MTTs and, therefore, does not question the general patterns that were observed.

We used the stream winter isotopic composition and ~~chemistry~~BC concentration to test Mike-~~ ~~SHE's ability to capture the variability of ~~groundwater~~ travel times in the 14 Krycklan sub-catchments. Based on ~~this~~our results, we found significant and robust correlations between the winter isotopic signature ~~of both~~ $\delta^{18}O$ ~~and δ^2H~~ as well as the stream chemistry, on the one hand, and the calculated travel times on the other (Fig. ~~3~~6). Theoretically, infinitely long travel time would result in a stream water isotopic signature approaching the long-term average precipitation input~~, while~~ (Fig 2). In contrast, the ~~base cation~~BC concentration of the stream water would increase until it reaches thermodynamic equilibrium with the soil mineral composition (Erlandsson et al., 2016). The strong statistical agreement between both the observed winter isotopic composition and stream chemistry and the particle travel times on the other supports the credibility of the model results.

~~A comparison of these results to previous studies of MTT for one of the Krycklan sub-catchment (C7) shows that the different approaches gave similar results. While this study estimated the long term mean travel time to 1.3 years, Peralta-Tapia et al. (2016) calculated a ten-year average travel time of 1.7 years using isotopic data and a gamma transformation method. In a non-distributed modeling study using the same data, but another travel time distribution technique, the median of the travel time distribution was approximated to 0.9 years in the same catchment (Ala-aho et al., 2017).~~

~~The simulated travel times were compared to stream pH and base cation concentrations.~~

4.1.1 Testing model results against isotopic composition

According to the conceptual model (Fig. 2), older baseflow water should result in an isotopic signature closer to the precipitation average. There was a strong negative correlation between groundwater age and the streams isotopic signature during baseflow (Fig. 6a), suggesting that the model produces credible water age patterns for the winter season. The larger sub-catchments, including C14, C15, and C16, are close to the long-term precipitation average, which limits the ability to estimate the travel times using isotopes. Water older than 4-5 years is argued not to be accurately quantifiable using isotopes due to amplitude loss (Kirchner., 2016). These theoretical considerations strengthen our results of a winter MTT of 4-6 years for the larger sub-catchments and provided new insights into travel times for these systems.

In spring, the young water fraction was used to evaluate the proportion of water reaching the stream as overland flow. The difference between the previous winter baseflow and stream isotopic signature at snowmelt ($\Delta\delta O^{18}_{spring}$) is mechanistically related to the amount of young water reaching the stream (Laudon et al., 2007; Tetzlaff et al., 2009). We found a strong connection between $\Delta\delta O^{18}_{spring}$ and our calculated young water fractions (Fig. 6c). The larger young water fraction was generally found in mire dominated sub-catchments, such as C4 and C5. In contrast, equally sized sub-catchments without mires, such as C1 and C2, had a less $\Delta\delta O^{18}_{spring}$ and hence smaller young water fraction. Notably, these small, entirely forested catchments are the only ones with no overland flow during the spring flood, which again emphasizes the importance of the mires for the hydrology of the boreal landscape (Fig. 5). These results are well in line with previous work in Krycklan using end-member mixing of new and old water in the same streams (Laudon et al. 2007; 2011). Those earlier results showed a large overland flow component in wetland catchments because of frozen conditions during spring flood with biogeochemical consequences during snowmelt.

In summer, the conceptual model predicted that $\Delta\delta O^{18}_{summer}$ should also be correlated to the young water fraction, but with the opposite sign, due to the enriched summer rain (Fig. 2). Inter-annual precipitation and evapotranspiration variability likely caused the relationship to be less evident compared to the spring flood results as the snowmelt conditions are more consistent from year to year. However, although less strong than compared to the spring $\Delta\delta O^{18}$, there was still a strong connection between the average summer $\Delta\delta O^{18}$ and the modelled young water fraction (Fig. 6e).

4.1.2 Testing model results against base cation concentration

The base cation (BC) concentration followed the same pattern throughout the year (Fig. 6b, 6d, and 6f), with increasing concentration strongly correlated to increasing age. Since the weathering rates generally are kinetically controlled, i.e., related to the travel time, such stream chemistry variables can be used as a relative indicator for stream water age~~.~~, as long as the mineralogy remains comparatively homogenous (Erlandsson Lampa et al., 2020). This study showed that the ~~modeled~~modelled travel times were significantly correlated to the ~~observed pH and base cation~~BC concentrations. ~~The use of pH and cations as tracers for groundwater residence times should be done with caution since both are involved in several biogeochemical processes.~~ Reducing weathering to a matter of travel times ~~only~~ may be an oversimplification~~. Weathering~~ as the rate is affected by, for example, chemical conditions, differences in mineralogy, and particle size distributions. However, previous research in the Krycklan catchment has indicated that the chemical composition of the local mineral ~~Quaternary deposits~~soils is surprisingly homogeneous, even when comparing unsorted till and sorted sediments (Klaminder et al., 2011; Peralta-Tapia et al., 2015; Erlandsson et al., 2016; Lidman et al., 2016~~; Peralta-Tapia et al., 2015; Klaminder et al., 2011~~). Therefore, we do not expect mineralogical differences between soil types to have a significant impact on the release of cations. The one exception is peat deposits, which strongly affect the cation concentrations, and that was accounted for by adjusting the concentrations for the influence of the mires following Lidman et al. (2014). Differences in particle size distribution may be important because coarser ~~regolith~~soils will have less surface area per volume unit, therefore allowing for less weathering. However, such ~~Quaternary deposits~~soils can also be expected to have higher hydraulic conductivities, leading to higher flow velocities and, consequently, less time available for weathering. Therefore, differences in area-volume ratios between different soil types would not counteract the effect of travel times on the weathering, rather enhance it. Accordingly, base cation concentrations should still be a useful indicator of transit times. ~~The pH of some sub-catchments has also been shown to be affected by mires, especially C4 and C5 (Buffam et al., 2007), due to high concentrations of organic acids that influence pH, especially when the buffer capacity is low (Hruska et al. 2003). This effect can be observed in the results of the mire dominated sub-catchments, which fall below the 95 % prediction line (Fig. 3D and 3F). Hence, we do not think that these deviations contradict the credibility of the model results.~~

~~It can also be argued that pH is not a mixable quantity and therefore unsuitable as a tracer. Still, the purpose of the comparisons to stream water chemistry was not to mechanistically explain the evolution of stream water chemistry, but instead to compare the modeling results to some parameters that could be expected to reflect the groundwater travel times. Such tests are crucial for the credibility of the model results. Because pH increases as a result of silicate weathering, it is likely that pH would increase with the groundwater travel time, albeit not necessarily in a linear manner. Complementing isotopic tracers with transported solutes for testing simulated travel times provide more insight into catchment processes. Despite the arguments that can be made against the use of pH and~~

Despite arguments that can be made against the use of cations as tracers, they still offer a complementary possibility to test the performance of the model. As emphasized by McDonnell and Beven (2014), the inclusion of tracers in hydrological models is necessary to ensure that a model ~~does reproduce~~reproduces the speed of flow, which is ~~a crucial~~an important parameter when assessing travel time distributions. In catchment-scale models, this could be an isotopic tracer or a solute that is transported with the water (Hooper et al., 1988; Seibert et al., 2003; Fenicia et al., 2010; ~~Hooper et al., 1988;~~ Hrachowitz et al., 2013~~; Seibert et al., 2003~~). Although neither the travel time distribution nor the kinetics of weathering is fully understood, the strong agreement between the calculated travel times and the observed stream water chemistry strengthens ~~the credibility of~~ our model results and~~,~~ the system understanding of catchment-scale travel times and their connection to biogeochemistry. More specifically, the ~~chances~~results increase the likelihood that the model is producing credible results for the right reasons.

~~Strengthening the credibility of particle tracking in Mike SHE to produce travel time distributions enables the use of particle tracking as a useful complement to other similar studies in the future. For example, stable water isotopes and biogeochemical tracer tests can be affected by dilution or chemical reactions, and here particle tracking could be a useful complement. Further extensions in the Mike SHE family (Graham & Butts, 2005) also allow the incorporation of solutes or isotopes with more complex biogeochemical behaviour. These extensions could be used for further calibration and validation of the hydrological model (McDonnell and Beven, 2014) as well as investigation of biogeochemical processes in the catchment. A more mechanistic investigation of the relationship between groundwater travel times and stream water chemistry would require such extensions.~~

~~4.2 Catchment slope and hydraulic conductivity control travel times~~

~~Contrary to our hypothesis,~~

4.2 Mean travel times, young water fractions, and catchment characteristics

The main ~~factor controlling the groundwater travel times was the hydraulic conductivity and slope of the~~ catchment ~~rather than the catchment size itself. In agreement with previous studies by Capell et al. (2012), Muñoz-Villers et al. (2016), and Tetzlaff et al. (2009), there were significant correlations between catchment slope and travel time~~characteristics found that affect the age and young water fraction were low conductive soils (LCS) and the fraction of mires (Fig. ~~4). However,~~7, Table 6). The most significant factor for the mean travel times was ~~found~~related to ~~be~~ the proportion of low conductive ~~sediments~~soils (LCS), which overshadowed ~~both~~ the ~~slope~~importance of ~~the~~ catchment ~~and the travel distance of particles~~size. Earlier studies in Krycklan by Peralta-Tapia et al. (2015) and Tiwari et al. (2017) have suggested that the MTT of groundwater is ~~linked~~ nonlinearly linked to the catchment size~~, i.e., the~~

~~travel times increases with the catchment size.~~. However, we found the silt-rich but relatively small C20 to be a distinct outlier to this pattern, indicating that catchment size may not be the underlying factor causing high MTTs (Fig. ~~4).~~6). As shown in Table 6, the catchment size is correlated to the fraction of LCS. In other words, there are few small catchments in the silt-areas with low conductivity. The reason is partly the setup of the Krycklan catchment study, which initially focused on the till areas, and partly the fact that the LCS are located in the lower parts of the Krycklan catchment so that all large catchments by necessity must contain at least some LCS. The long travel times in relation to the relatively small catchment size in C20 means that the groundwater flow velocity generally is lower than elsewhere. Nevertheless, the average catchment slope of C20 is steeper than in comparably sized sub-catchments in ~~the~~ till areas, so the topographical possibilities to build up ~~high~~ hydraulic gradients that can drive the water transport should be ~~larger~~large (Table 1~~;~~ and Fig. ~~4). This is probably related to the fact that C20 is the only relatively small sub-catchment (~~1~~.45 km^2) in the area largely covered by LCS. However,~~). The fluvial sediment deposit ~~fraction~~ may also explain the relatively long travel times of C14 and C16. ~~For example,~~ Although C14 is smaller than C15, which mostly lacks LCS, it still has longer MTT.

In contrast, C15 is much closer to C12 and C13 in MTT, even though the C15 catchment is almost twice as large~~. This suggests~~ (Table 5). The results suggest that the critical difference between these sub-catchments and other sub-catchments is related to the hydraulic conductivity of the ~~regolith,~~soils rather than the catchment size. Without the contribution of water from headwater catchments with fine ~~regolith~~soils (such as C20), the MTT of sub-catchments like C14 and C16 would probably ~~have a MTT~~be much closer to that of the other smaller till dominated sub-catchments. The results, therefore, emphasize that one cannot generally assume that the travel time would increase with catchment size unless the distribution of ~~regolith is comparable throughout the landscape~~soils is comparable throughout the landscape. The effect of LCS is more prominent in winter than during the other seasons. For example, the difference between the winter and spring mean travel time is almost six years for C20 compared to two years for the similar-sized sub-catchment C6 and the mean travel time of C14 is four years compared to three years for the similar-sized sub-catchment C15.

~~5 Summary remarks~~

~~The Mike SHE model showed promising results in its ability to capture groundwater travel times, which was firmly related to winter stream isotope signatures. The simulated travel times were, in turn, well correlated to the base cation concentration and pH of the streams. In contrast to our hypothesis, we found that the catchment size itself is not the main factor determining groundwater travel times. Instead, we found the hydrologic conductivity of regolith to be the most important parameter, but also that the catchment slope and travel distance of the groundwater could have an impact. This essentially points back to Darcy's law, which states that the groundwater flow is governed by the pressure gradient (approximated by the catchment slope in this case) and the hydraulic conductivity of the medium (approximated by soil types). In that sense, the results are in line with theory. However, it is far from evident that precisely these catchment parameters would stand out as most important in the complex landscape of the Krycklan~~

~~catchment or, for that matter, the boreal landscape at large. It is also important to note that catchment size may not be as significant as previously thought.~~

Sub-catchments with mires receive the highest young water fraction during the spring snowmelt; however, the annual age of water is not as strongly connected to that landscape feature (Table 6). The main factor that controls this is the soil frost on the mires (Peralta-Tapia, 2014), which reduces the renewal of the groundwater at spring because a larger fraction of water flows directly to the stream as overland flow. For example, C2 and C4 have a similar catchment size and soil properties, with the main difference that C4 has a significant fraction of mires and a greater seasonality in travel times. Even though C2 and C4 have this landscape difference, they still have a similar annual age (Table 5). Besides the somewhat higher specific discharge from mires compared to forests (Karlsen et al. 2016), the main hydrological effect of mires consequently appears to be a redistribution of water between the seasons, causing younger runoff during the spring and older water during dry and cold seasons. In forest till soils, on the contrary, most of the snowmelt infiltrates the ground and instead displace older, pre-event water during the spring flood. The infiltration of snowmelt water leads to a replacement of older water by younger in the forest soils. This process called transmissivity feedback explains the younger water age during the rest of the year and the smaller seasonal variation of forested till-soil catchments (Bishop, 1991; Laudon et al. 2004). The process is a consequence of exponentially increasing hydraulic conductivity toward the soil surface in till soils.

5 Summary remarks and implications

Northern landscapes are sensitive to climate change (Tetzlaff et al., 2013; Sprenger et al., 2018). Climate predictions suggest that warming will affect higher latitudes to a disproportionally large extent, and hence soon begin to affect the annual snowpack, shorten the longevity of snow cover, increase the frequency of winter thawing episodes, reduce soil frost, and increase annual precipitation (IPCC., 2014; Jungqvist et al., 2014; Brown et al.,2017; Lyon et al., 2018). To foresee the implications of such changes, it is important to have a good baseline understanding, including both empirical data but also well-calibrated and tested models, upon which we can build future predictions of what such changes will mean to our water resources. In a mosaic landscape, such as the northern boreal landscape, distributed models can be of great value in this context since variable impacts on different landscape characteristics can be distinguished and disentangled.

The present study was based on the integration of a large dataset from a previously well-investigated catchment and an advanced distributed 3D hydrological model. The results showed that the groundwater travel times vary considerably on annual and intra-annual scales in the boreal landscape, both as an effect of physical differences between different types of catchments, most notably the hydrological conductivity of the soils, and the response of different landscape units to the changing of the seasons. Yet, the combination of stable water isotopes, stream water chemistry, and particle tracking provided a consistent picture of how the boreal landscape functions hydrologically and what processes and factors are of importance. Hence, this system approach not only strengthens the credibility of these specific modelling results but also more broadly confirms the applicability of process-based numerical modelling and particle tracking under the complex hydrological conditions with, for example, long dry winters, temporary soil frost, and intensive spring floods that prevail in the boreal region. In the wake of a changing climate and intensified pressure from forestry and other forms of land use, this provides a useful foundation for assessing the often-intricate connections and feedbacks between hydrological and biogeochemical processes throughout the boreal landscape.

6 Data availability

The Data from the Hydrological Research at Krycklan Catchment Study is available in Svartbergets open database ~~(www.slu.se/Krycklan).~~(www.slu.se/Krycklan).

7 Author contribution

Elin Jutebring Sterte ~~was~~and Hjalmar Laudon were responsible for the design, conceptual idea, and evaluation of results in collaboration with ~~all~~ the other co-authors. Elin Jutebring Sterte and Emma Lindborg were responsible for ~~the~~ numerical modelling. Elin Jutebring Sterte prepared the manuscript and figures. ~~All co-authors contributed to~~Elin Jutebring Sterte lead the writing of the paper with contributions from all co-authors.

8 Competing interests

The authors declare that they have no conflict of interest.

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