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

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Key Points:

- A numerical model was used to estimate annual and seasonal mean travel times ~~and flow pathways~~ across ~~the Krycklan~~ 14 long-term monitored sub-catchment in the boreal region of northern Sweden.
- ~~The estimated annual mean travel times of 14 partly nested sub-catchments ranged between 0.8 and 2.7 years~~
- The estimated travel times and young water fractions were consistent with observed ~~stream chemistry (~~variations of base cation concentration and stable water isotopes, δ[18]O~~)~~.
- ~~Hydraulic conductivity of~~ The soil type was the most important factor regulating the variation in mean travel times between different sub-catchments, while the areal coverage of mires ~~mainly~~ affected the ~~youngest fraction of stream water.~~
- ~~Although all sub-catchments showed seasonal changes in mean travel times and~~ young water ~~fractions,~~ fraction.
- The greatest seasonality in mean travel times was found in sub-catchments dominated by silty soils contributing with ~~a substantial~~old water during winter baseflow, while mires contributed the largest fraction ~~of mires~~young water during spring snowmelt.

Abstract

Understanding travel times and hydrological pathways of rain and snowmelt water transported through the 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 investigate the travel time, and pathway of water in 14 partly nested, long-term monitored boreal sub-catchments. These sub-catchments are characterized by long and snow-rich winters with little groundwater recharge and highly dynamic ~~hydrology~~runoff during ~~the following~~ spring snowmelt. The ~~calculated annual~~geometric mean of the travel ~~times~~time distribution (MTT$_{geo}$) for these sub-catchments varied from 0.8-2.7 years. ~~The seasonality caused considerable variation in travel times between~~ The variations were found to be related to the distribution of different ~~seasons and~~ landscape types~~, with winter mean travel times ranging~~ and their different response to seasonal changes. Winter MTT$_{geo}$ ranged from 1.2-7.7 years~~ and~~, while spring ~~mean travel times ranging from~~MTT$_{geo}$ varied between 0.5-1.9 years. The modelled variation in annual and seasonal ~~travel times~~MTT$_{geo}$ and the fraction of young water ~~(less than three~~(<3 months) was supported by extensive observations of both $\delta^{18}O$ and base cation concentrations in the ~~stream water. The age of~~different streams. The groundwater age was positively correlated to the ~~abundance~~areal coverage of low conductive ~~soils~~silty sediments (r=0.90, P<0.0001). As a result of lacking synchronicity and contrasting hydrological responses between different soil types (e.g., ~~peat~~mires and ~~low-conductive soils~~silty sediments), mixed catchments typically displayed larger ~~differences~~variability in ~~travel times between winter baseflow and spring flood. Mires were~~seasonal MTT$_{geo}$. The areal coverage of mires was found to ~~affect~~ be especially important for the ~~young water fractions of the stream~~ contribution of young water in spring (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~~was attributed to extensive soil frost in mires, causing considerable overland flow ~~in spring. The~~during the snowmelt period. However, this lower groundwater recharge during ~~these periods~~snowmelt caused mire-dominated catchments to have ~~older~~longer stream ~~water contribution~~runoff MTT$_{geo}$ than comparable forest catchments ~~during other parts of the year.~~in winter. Boreal landscapes are sensitive to climate change, and our results ~~show~~suggest that changes in seasonality are likely to ~~affect different landscape types~~cause contrasting responses in different ~~ways.~~

catchments depending on dominating landscape type.

1 Introduction

The age and ~~pathways~~pathway of water through the terrestrial landscape to ~~streams~~stream networks is a widely discussed topic in contemporary hydrology. This interest has emerged because of the ~~important~~significant role residence time, and routing of water through various subsurface environments have on hydrological and biogeochemical processes (McDonnell et al., 2010; Sprenger et al., 2018). ~~These include~~This includes fundamental implications for weathering rates (Burns et al., 2003), transport and dispersal of contaminants (Bosson et al., 2013; Kralik, 2015), and accumulation and mobilization of organic carbon and associated solutes (Tiwari et al., 2017). The travel time, 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~~, and to the 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). Therefore, estimating travel times for contrasting landscape elements is a challenging task, but ~~may~~when successful, will enhance our ability to understand and predict catchment functioning more adequately.

Stream water consists of a blend of ~~groundwater and~~ overland flow and groundwater 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~~. Instead, it is the runoff ~~mix~~ that generally has travelled the ~~farthest~~furthest and is the oldest (Klaus et al., 2013; Hrachowitz et al., 2016). In contrast, young ~~waters are~~stream water is typically connected to overland flow or fast and shallow groundwater, which mainly can be seen at times ~~when new precipitation input and/~~with large rain or snowmelt ~~arrives~~inputs (Peters et al., 2014; Hrachowitz et al., 2016). Travel times are complex to quantify, especially ~~at~~on intra-annual time scales, as they ~~can~~ vary in time and space depending on numerous scale-dependent and independent processes (Botter et al., 2010). A better understanding of the seasonal variability in the ~~connection between~~ fraction of young and old waters ~~in various catchment systems~~ can help provide insights into the fundamental role catchment characteristics play ~~for the regulation of~~in regulating 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 water sources ~~of water~~ and follow ~~its~~their pathways through the landscape (Maulé and Stein, 1990; Rodhe et al., 1996; Goller et al., 2005; Tetzlaff and Soulsby, 2008). Isotopic tracer dampening can provide an estimate of ~~mean travel times~~MTT (Uhlenbrook et al., 2002; McGuire et al., 2005; Peralta-Tapia et al., 2016), and more elaborate time-series analysis can ~~provide~~offer quantitative assessments of water age (Harman, 2015; Danesh-Yazdi et al., 2016). Theoretical transfer functions, such as the gamma distribution model, can also be ~~related to~~used by relating input~~-~~ and output signals ~~(for example,~~of isotopes, such as precipitation~~ to~~ -discharge~~) of isotopes~~ relationships (McGuire et al., 2005; Hrachowitz et al., 2010; Heidbüchel et al., 2013; Birkel et al., 2016). However, the isotope amplitude signal used to estimate ~~mean travel times~~MTT in many transfer functions is~~, however,~~ lost after approximately four to five years because of effective mixing (Kirchner~~.~~, 2016), ~~which limits~~limiting the use of isotopes ~~for~~in catchments with long travel times. The ~~fraction of~~ young water fraction, 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). The main advantage of water isotopes ~~mainly~~ is that they are relatively conservative and fractionate ~~due to~~primarily because of evaporation~~ and are hence not~~. Hence, once in the subsurface environment, the signal is only affected by ~~their subsurface pathways.~~the mixing of different water sources. In contrast, many biogeochemical tracers ~~may~~ react and transform on their route to ~~a stream~~streams

(Lidman et al., 2017; Ledesma et al., 2018). Such ~~transformations~~transformation and reactions depend on the specific solute and soil environment that water encounters and can ~~hence~~therefore give qualitative information about groundwater flow pathways ~~in mineral soils~~ (Wolock et al., 1997; Frisbee et al., 2011; Zimmer et al., 2012). ~~Therefore,~~ Combined information from conservative and reactive tracers can hence provide an enhanced understanding of hydrological ~~and biogeochemical~~ processes ~~(~~as their concentrations and dynamics can tell complementary stories about the specific pathways water take from the source to the recipient stream (Laudon et al~~.~~., 2011).

A complementary approach to field experiments is numerical modelling, which can ~~be useful for achieving~~help achieve a more complete system understanding~~ of catchment hydrology~~. Lumped hydrological models often describe catchments as single integrated entities. In contrast, distributed numerical models can include spatial heterogeneity in input parameters and therefore have the potential to represent catchment processes more ~~realistically.~~mechanistically. 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~~Two common ~~method~~methods 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~~empirical observations to build confidence in their ~~outputs.~~result output. Stream discharge, groundwater levels, and tracer data are examples of such validation data that can provide ~~important~~vital information ~~to understand a catchment hydrological functioning~~ (McGuire et al., 2007; Hrachowitz et al., 2015; Wang et al., 2017). ~~Such empirical~~ Collection of such field data ~~are~~is 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~~types in data-sparse catchments is currently a topic ~~for some debate~~of increasing interest (Bjerklie et al., 2003; Jian et al., 2017; Li et al., 2018).

Snow-dominated landscapes have ~~gained~~received increasing attention in the last decades due to their importance as water resources (Barnett et al., 2005) and their vulnerability to climate change ~~the 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 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~~Landscapes with long-lasting snow cover that often melts rapidly in the spring ~~create~~creates both opportunities and challenges ~~in the context of the determination of~~for determining stream water 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~~long, snow-rich winters do not only cause protracted periods of winter ~~conditions, whereby the source of water to the streams is originating from~~ baseflow ~~(Peralta-Tapia et al. 2016). 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~~with little or no recharge (Spence et al., 2011; Spence and Phillips, 2015; Lyon et al., 2018~~).~~), they also cause considerable amounts of water during the often short and intensive snowmelt in the spring. Although attempts to assess travel times generally have ~~shown good fits for a~~provided useful results using gamma distribution ~~function~~functions in snow-dominated catchments, ~~particularly~~ the winter season has proven to be especially challenging~~, which suggests~~ suggesting that other methods to assess travel times may be required (Heidbüchel et al., 2012; Peralta-Tapia et al., ~~2016). To account~~2016).

The boreal region also consists of numerous patches of lakes and mires, interspersed in a landscape dominated by coniferous forests on different soil types making this task even more challenging. Hence, accounting for the unique circumstances of both baseflow with long travel times and those of the intensive spring snowmelt with potential large overland flow components in heterogeneous landscapes requires ~~the need of~~ models that can handle the complexity and separation of various flow components across scales, soil types and landscape patches.

~~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), ~~with the purpose of enhancing~~to enhance our understanding of stream water contribution in boreal landscapes across seasons and landscape configurations. The water ~~moment~~movement model in Mike SHE calculates saturated (3D) groundwater flow and unsaturated (1D) flow and is fully integrated with the surface water ~~as well as~~and evapotranspiration. The water ~~movement~~flow model setup and results previously presented by Jutebring Sterte et al. (2018) were used as ~~a~~the study platform ~~in the present study.~~for this work. The model has been calibrated and validated to 14 sub-catchment ~~of~~using daily stream-discharge observations and ~~15 groundwater wells of periodically~~periodical measured groundwater levels in 15 wells throughout the Krycklan catchment in the boreal region of northern Sweden (Laudon et al~~.~~., 2013; Jutebring Sterte et al., 2018). The model 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.

The main objective of this study was to quantify yearly and seasonal age distributions and calculate ~~mean travel times~~MTT of ~~stream~~ water ~~contribution~~runoff to streams of the ~~sub-catchment in~~ Krycklan ~~in order~~sub-catchments to disentangle how these are related to physical landscape characteristics and seasonality. Firstly, the credibility of the model results was tested by comparing calculated travel times for the 14 ~~long-term monitored~~ sub-catchments to ten-year seasonal isotope signatures and base cation concentrations record from the Krycklan network. The usefulness of stream isotopic composition and chemistry record has previously been demonstrated for understanding the connection of hydrological flow pathways and travel times 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~~was previously ~~been~~ done by identifying the connection between travel times and different catchment characteristics and test how this varies between ~~different hydrological~~variable hydrologically 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.~~ is still partly frozen, and summer when evapotranspiration (ET) becomes a significant process. We focused especially on the catchment characteristics that have been suggested to be important factors for regulating stream chemistry of the Krycklan sub-catchments, including the areal coverage of mires, catchment size, soil properties, and seasonal changes in groundwater recharge (Karlsen et al., 2016; Klaminder et al., 2011; Laudon et al., 2007; Peralta-Tapia et al., 2015; Tiwari et al., 2017).

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~~spans 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~~elevations to the northwest, which are located above the highest postglacial coastline, the till soils can reach up to 15-20 m in thickness. Here, the soil primarily consists of ~~sandy-silty~~glacial till, and the landscape is intertwined with lakes and peatlands. In this study, we refer to ~~soils~~soil as all unconsolidated material above the 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 $5 \cdot 10^{-5}$ m/s at the ground surface and exponentially decreasing downwards (Nyberg, 1995). At lower ~~altitudes~~elevations, the soils ~~mainly~~ consist of fluvial deposits of primarily sandy and silty ~~clay and sand.~~sediments. Compared to the ~~soils~~soil at higher ~~altitudes~~elevations in the catchment, these deposits can reach thicknesses up to approximately 40~~-~~ to 50 m and ~~are~~have hydrological conductivity that is more ~~homogeneous~~constant with depth.

~~The catchment is divided into 14 nested sub-catchments and has been included~~For more than 30 years, multi-disciplinary biogeochemical and hydrological ~~research for more than 30 years (~~studies have been conducted in Krycklan (e.g., Laudon and Sponseller, 2018). ~~The~~Streamflow is monitored in 14 nested sub-catchments~~ are~~, called C1 to C20~~.~~, with the longest continuously monitored time-series ~~of streamflow began~~starting at the beginning of the 1980s. Connected by a network of streams, the different sub-catchments ~~have distinct characteristics, which allow for~~allow an evaluation of the effects of catchment characteristics on hydrologic transport, including soil type, vegetation, and differences in topography~~.~~ (Table 1).

(Edited) **Table 1: Sub-catchment characteristics.** The list includes all 14 monitored sub-catchments in Krycklan, called C1 to C20~~.~~, including the entire Krycklan catchment, C16. Different branches of the stream network are illustrated in ~~different~~distinct colours in Fig. 1. The table includes the sub-catchment area, average elevation, and average slope. Further ~~description~~descriptions of these characteristics can be found in Karlsen et al. (2016). The table also includes soil proportion based on the soil~~.~~ map (1:100,000) from the Swedish Geological Survey (2014).

	Catchment size (km^2)	Average elevation (m.a.s.l.)	Slope (°)	Till (%)	Mire (%)	Sandy sediments (%)	Silty sediments (%)	Lake (%)
C1	0.48	279	4.87	91	0	0	0	0.0
C2	0.12	273	4.75	79	0	0	0	0.0
C4	0.18	287	4.24	29	42	0	0	0.0
C5	0.65	292	2.91	47	46	0	0	6.4
C6	1.10	283	4.53	51	29	0	0	3.8
C7	0.47	275	4.98	68	16	0	0	0.0
C9	2.88	251	4.25	64	14	7	4	1.5
C10	3.36	296	5.11	64	28	1	0	0.0
C12	5.44	277	4.90	70	18	6	0	0.0
C13	7.00	251	4.52	60	10	9	9	0.7
C14	14.10	228	6.35	46	6	24	15	0.7
C15	19.13	277	6.38	64	15	8	2	2.4
C16	67.90	239	6.35	51	9	21	10	1.0

C20	1.45	214	5.96	55	9	0	28	0.0

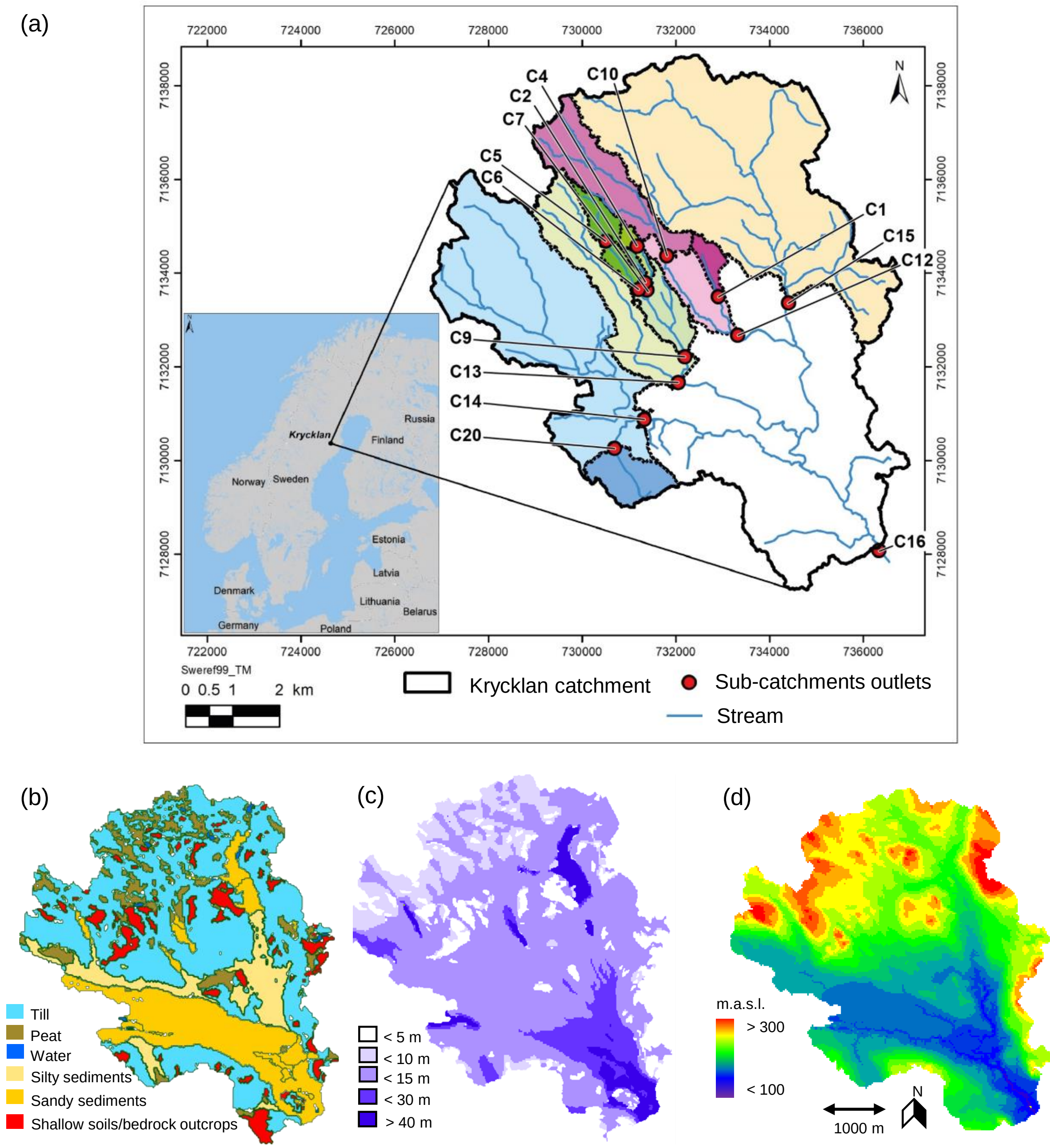


Figure 1: The Krycklan catchment. (a) Location of sub-catchment and their outlets. The areas are color-coded based on their stream network connections, e.g., all sub-catchments of one colour connect before reaching the white area. For further details of the catchment characteristics, see Table 1. (b) The soil map used in the ~~hydrology~~Mike SHE flow model and is based on the 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~~meters below the ground surface. (ed) Catchment topography, shown as ~~meter~~meters above sea level (m.a.s.l.).

~~2.2 Water flow model setup – Mike SHE~~

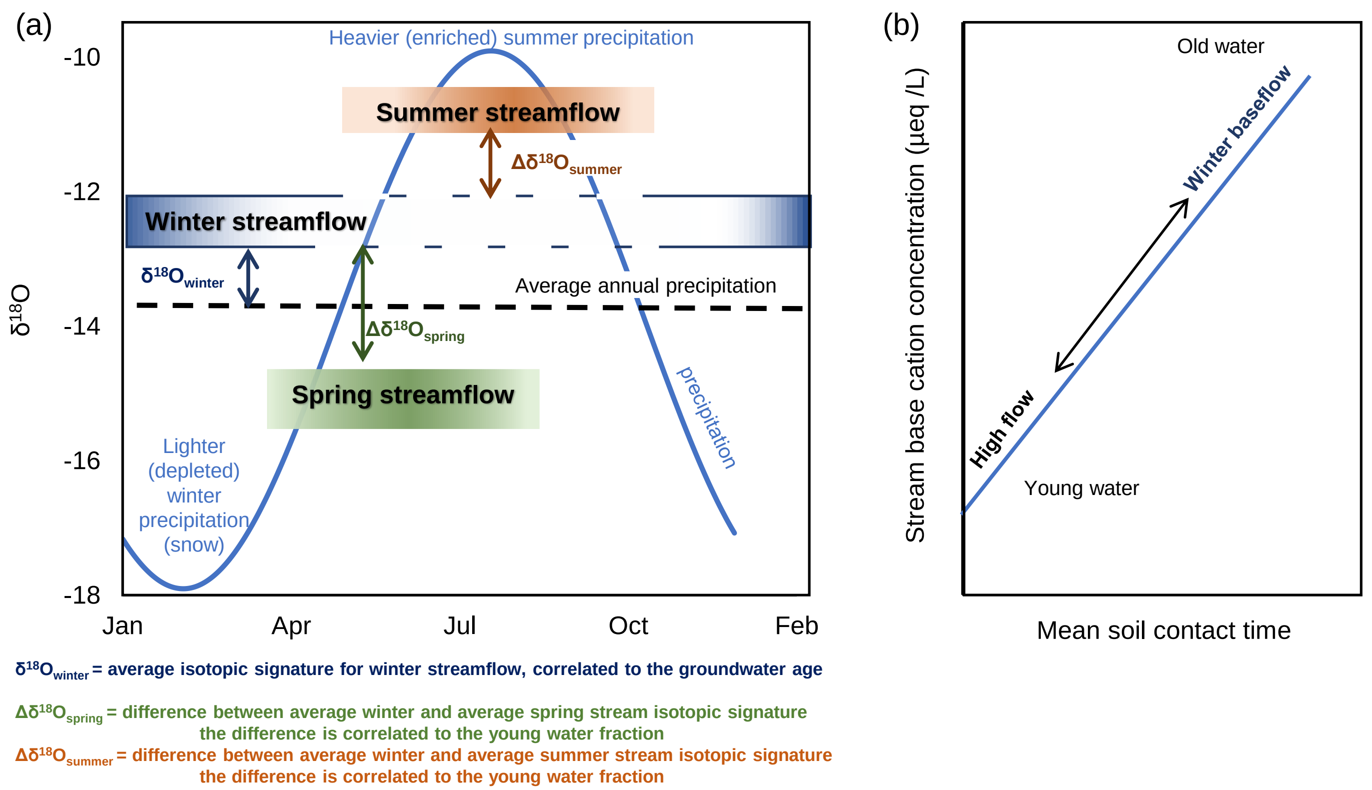
~~The Mike SHE model setup used as a platform in~~ 2.2 Linking seasonal base cation concentration and isotopic signature to stream water age

This study was ~~a slightly modified version of the previously established and validated surface and~~ focused on three seasons in Krycklan, winter, spring, and summer. We defined the winter to occur from late November to late March because it is characterized by an extensive and permanent snow cover with little groundwater recharge. We assumed that the winter stream composition reflects the chemistry of deeper groundwater (Fig. 2). Similarly, we defined spring by the rapid transition in hydrology and biogeochemistry in April-May. During snowmelt, ca 50 % of the annual precipitation leaves the system in a short period of time, diluting baseflow with new input of water. Finally, we defined the summer season as the period between July and September when the hydrology is characterised by rain, high ET and relatively little runoff. June and October were excluded because, hydrologically, they are transition months between the three distinct seasons. This is because snowmelt can still influence runoff in June, and winter conditions (snowfall, soil frost, etc.) can sometimes begin to establish in October.

In this study, stable water isotopes ($\delta^{18}O$) were used to track pathways of precipitation inputs to stream networks (see Appendix for the $\delta^{18}O$ definition). Ten years of $\delta^{18}O$ results for 13 of the 14 sub-catchments were used. Some of the sub-catchments are affected by evaporation from lake surfaces that result in isotopic fractionation that, consequently, affected the signal (Leach and Laudon, 2019). This fractionation was corrected for the percentage of lakes in each sub-catchment (Table 2), using the same principle as Peralta-Tapia et al. (2015) but adjusted to newly acquired $\delta^{18}O$ observations.

The comparison of the modelling results to observations of $\delta^{18}O$ was based on a conceptual model of the seasonal variability and differences between precipitation and runoff (Fig. 2a). Because there is no groundwater recharge during winter, the stream isotopic signature originates from groundwater only (Laudon et al., 2007). Hence, we assumed that $\delta^{18}O$ during winter baseflow should be statistically related to the average age of the groundwater (up until the point where full mixing is reached). The closer the signature is to the long-term precipitation average (which is equal to the deep groundwater measurements in Krycklan (Laudon et al., 2007)), the more well-mixed and, consequently, the older the average groundwater has become. In spring, previous studies have shown that the young water fraction can be distinguished by comparing the change in the isotopic signature to the preceding winter because the snow is much lighter (depleted in ^{18}O) (Laudon et al., 2007; Tetzlaff et al., 2015). We refer to the difference between the average winter and average spring signature as the $\Delta\delta^{18}O_{spring}$, which we assumed to be negatively correlated to the young water fraction (Fig. 2a). Similarly, we refer to the difference between the average winter and average summer signature as $\Delta\delta^{18}O_{summer}$, which similarly should be related to the young water fraction during the summer. However, in summer, precipitation is heavier (more enriched in ^{18}O) compared to winter, which hence should give the young water a heavier signal. Therefore, we assumed a positive relationship between the young water fraction and the $\Delta\delta^{18}O_{summer}$ (Fig. 2a).

Another indicator of stream water age we used was the sum of base cations (BC) concentration (Fig. 2b) (Abbott et al., 2016). Previous attempts to follow the chemical development of groundwater in the Krycklan catchment and other streams have shown that the BC concentration increases along the groundwater flow pathway (Klaminder et al., 2011). Therefore, a correlation between the stream concentration of BCs on the one hand and modelled soil contact time on the other were assumed in this study. The BCs are mainly derived from the weathering of local soils in the Krycklan catchment, with only a minor contribution from atmospheric deposition (Lidman et al., 2014). Our assumption is further based on modelling studies of weathering rates in a soil transect in the Krycklan catchment, which indicates that there is kinetic control of the release of BCs in the soils (Erlandsson et al., 2016). Since all BCs behave relatively conservatively in these environments (Ledesma et al., 2013; Lidman et al., 2014), we used their combined concentration as a proxy for soil contact time. However, the assumption is only valid when the water is in contact with mineral soils, not with peat in mires, which are abundant in some of the investigated sub-catchments. There are no minerals present in the peat and, therefore, the BC concentration cannot be expected to increase during the time the water spends there. Therefore, the BC concentrations were adjusted for the influence of mire, using the sub-catchment mire proportion as a scaling factor to allow a fair comparison to water soil contact time (Lidman et al., 2014) (Table 2).



(Edited) Figure 2: Conceptual figure of stream water travel time vs. stream isotopic signature (a) and stream base cation concentration (b). (a) The connection between $\delta^{18}O$ and stream water travel time, where the sine curve shows the annual variations of $\delta^{18}O$ in precipitation, and approximate seasonal winter, spring, and summer stream composition are marked. In winter, the travel times are related to the deviation in the isotopic signature between the winter baseflow and the long-term precipitation. In spring, the fraction of young water is correlated to the difference between the spring stream signature and the winter baseflow. In summer, the fraction of young water is correlated to the difference between the stream signature and winter baseflow. (b) The connection between base cation (BC) concentration and soil contact time. The longer water spent in the mineral soil, the higher the stream concentrations of BCs due to soil weathering.

All stream chemistry data comes from the online open Krycklan database at www.slu.se/Krycklan (Table 2). The isotopic signatures contain approximately ten years of field observations (2008 to mid-2018), approximately 25 samples per year for each site. Parts of the dataset have been published by Peralta-Tapia et al. (2016), where sampling and analyses are described in detail. It has since been expanded using the same methodology. We used the average winter isotope signatures from these years as a representation of baseflow. These averages were also compared to the volume-weighted average of the long-term precipitation, calculated using 1160 precipitation measurements of $\delta^{18}O$ between 2007 and 2016. The precipitation was measured throughout the year, both as rain and as snow. 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 2: Seasonal stream chemistry.

	$\delta^{18}O$[a]						Base cations (BC)[b]					
	Winter		Spring		Summer		Winter concentration		Spring concentration		Summer concentration	
	‰	SD/SEM[c]	$\Delta\delta^{18}O$	SD/SEM	$\Delta\delta^{18}O$	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]					

~~model presented in Jutebring Sterte et al. (2018). The model has a horizontal grid resolution of 50*50 m. Vertically~~ [a] $\delta^{18}O$ Signature (2008-2018), data have been adjusted according to the lake proportion

[b] Base cation concentration (2008-2016), data has been adjusted according to the mire proportion

[c] SD = standard deviation, SEM = standard error of the mean

[d] Measured precipitation average for isotopes (2007-2016) and measured BC concentration (the year 1997 to 2018)

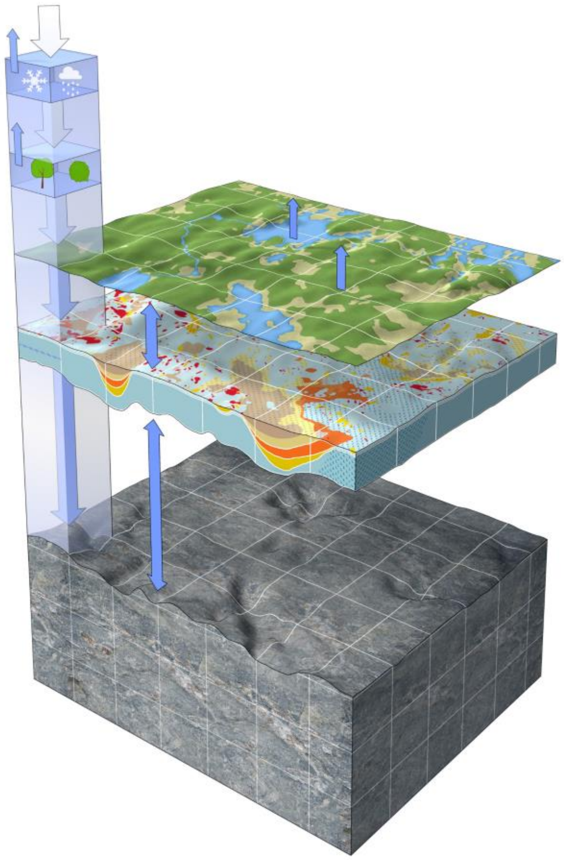
2.3 Water flow model setup

We applied the Mike SHE/Mike-11 hydrological modelling tools to quantify travel times in a pre-calculated 3D transient flow field. The simulated terrestrial hydrological system for the Krycklan catchment includes: the saturated and unsaturated flow, ET, snowmelt, overland flow, and streamflow processes. The fully distributed 3D modelling tool use topography, soil properties, and time-varying climate inputs to calculate the water fluxes throughout a catchment (Rahim et al., 2012; Sishodia et al., 2017; Wang et al., 2012; Wijesekara et al., 2014). The ET processes include canopy interception, open surface evaporation, root uptake, sublimation, and soil evaporation from the unsaturated zone based on a methodology developed by Kristensen and Jensen (1975). Flow in the saturated zone (SZ) is calculated in 3D by the Darcy equation. The flow in the unsaturated zone (UZ) is calculated in vertical 1D using the Richards equation, and overland flow (OL) is calculated using a horizontal 2D diffusive wave approximation in the Saint-Venant equations (Fig. 3). Streams are modelled in 1D using a high-order dynamic wave formulation of the Saint-Venant equations. The river model (Mike 11) is not restricted to the grid size of Mike SHE and allows for a more precise calculation of stream water levels and flow rates. The different model compartments OL, UZ, SZ, and rivers are fully integrated, and water fluxes between and within the compartments are calculated in each time step of the simulation. More in-depth documentation and manuals of Mike SHE and Mike 11 is provided by Mike powered by DHI (www.mikepoweredbydhi.com).

For the Krycklan model, the horizontal grid was set to 50*50 m. Vertically, the model is divided into ten calculation layers (CL) and ~~reaches~~extends to a depth of 100 m below ground. ~~The calculation layers follow the~~ The CLs of the SZ vary with depth and are thinner closer to the soil surface; the first CLs extend to 2.5, 3, 4, and 5 meters below the ground surface, with the soil properties and depth extension following the 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~~ (Table 3). The ET and UZ processes are only fully active in the uppermost SZ-CL, and here the ET and UZ are calculated at a finer resolution, leading to a detailed calculation ~~layer. Therefore, the uppermost layer must be deep enough to cover the part of the soils influenced by evapotranspiration processes and~~ of the groundwater table level. If the groundwater table falls below the first SZ-CL, a more simplistic method, not taking capillary rise ~~of groundwater. This depth averages several soils types~~and all ET-processes into account, was applied. The depth of the first SZ-CL was set to 2.5 m and was calibrated using the influence of the CL thickness on groundwater table level, UZ, and ET dynamics.

The thickness of the first SZ-CL in the Krycklan model results in all soils shallower than 2.5 meters being averaged 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 groundwater velocity in the uppermost part of the soil (Bosson et al., 2008). For more information regarding the model-setup, see Jutebring Sterte et al. (2018).~~soil type. In Mike SHE, horizontal hydraulic conductivity (Kh) is averaged using the thickness of each soil layer. Vertical flows are more dependent on the lowest vertical hydraulic conductivity (Kv). Therefore, the harmonic weighted mean value is used to calculate the new Kv instead (Table 3). In the Krycklan model, and several previous studies (Bosson et al. 2012, 2013, Johansson et al. 2015 and Jutebring et al. 2018), a drain function was used to account for higher hydraulic conductivity in the uppermost part of the first CL. In the Krycklan model, the function was activated whenever the groundwater reached 0.5 m below the ground surface, above which higher K-values have been observed (Table 3) (Bishop et al., 2011; Nyberg, 1995; Seibert et al., 2009). The model also accounted for soil freezing processes, which in Krycklan has been shown to have a strong influence on the water turnover in mires (Laudon et al., 2011). Based on a methodology presented in Johansson et al. (2015), soil freeze and thawing processes were described using time-varying K and infiltration capacity.



(New) **Figure 3: Schematic Mike SHE model set up.** Precipitation falls on the ground as rain or snow. Evapotranspiration (ET) processes include canopy interception, open surface evaporation, root uptake, and soil evaporation from the unsaturated zone (UZ). The overland flow (OL), saturated zone (SZ), and UZ interact depending on the saturation level. The SZ is divided into ten calculation layers (CL), while the UZ has a much finer description. Streamflow is modelled through Mike 11 and is not restricted to the Mike SHE resolution. The figure is used on the courtesy of SKB. Figure illustrator: LAJ.

The Krycklan flow model was able to reproduce observed stream accumulated discharge, groundwater levels, and timing of precipitation events (Jutebring Sterte et al., 2018). These include daily discharge observations (14 streams) and weekly to monthly observed groundwater levels (15 wells) for 2009-2014. The accumulated error in stream discharge was on average 11%, and highest for sub-catchments with few observation points (<25%). For this study, a few changes were made to the ~~original~~ Krycklan ~~Mike SHE~~ model~~.~~ (Jutebring Sterte et al., 2018). Most importantly, new field data from the Krycklan database gave a more precise location~~, and the threshold level~~ of the ~~lake outlet~~observation station at C5 (red circle in Fig. 1). The Kh of ~~C5 and the horizontal conductivity of the silty sand~~silt was also increased from $1*10^{-8}$ m/s to $1*10^{-7}$ m/s~~.~~ due to new soil property samples, which gave a slightly better flow representation of the sites affected silty sediments. However, the corrections and additions did not influence the model results in any substantial way. The improvements were small but were made to better represent the site according to all available data.

(Edited) **Table ~~2~~3: 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~~1b. A drain constant was used to account for coarser material of the upper half meter of the soil.

Soil type surface	Depth below ground (m)[a]	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	4	Silt/Sand	2*10^{-5}	2*10^{-5}
	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}
Drain constant				
Peat	1*10^{-6}			
Till	4*10^{-7}			
Silty sediments	1*10^{-7}			

[a] The table shows the depth to which the same description extends to. For example, the first description of Peat extends down to five meters, while the first calculation layer is 2.5 meters.

2.4 Establishing travel times - Particle tracking

Particle tracking in Mike SHE enables investigations of groundwater travel time ~~investigations~~from recharge into the SZ until reaching the streams, as described in detail 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~~calculates the location and age of separate 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. The particle tracking calculations in Mike SHE are applied to a pre-calculated flow field. Hence, in the first step, the~~ added with infiltrating water ~~movement calculation is performed, while in the second step, the tracing of~~ along their flow lines. The particles~~,~~ move by advection governed by the pre-calculated groundwater flow field from ~~a source point to a sink, is executed.~~the Mike SHE model (Jutebring et al., 2018). This method allows for long-term transport calculations where ~~the~~ particle tracking can be run for several annual cycles based on the same~~,~~ transient or steady-state~~,~~ flow field. The advection-dispersion equation governs the transportation of particles for a porous medium. The Darcy velocity is divided by the porosity to calculate the groundwater velocity. Therefore, the only complementary input data needed to run the particle ~~tracking is~~was porosity values (Table ~~3), which~~4).

Particle tracking was ~~added~~used to ~~the Mike SHE model set up.~~assess groundwater travel times from groundwater recharge to stream runoff for each sub-catchment. The model was run for 1000 years to capture the travel times of all discharging groundwater for each sub-catchment. One year of simulated flow results from Jutebring Sterte et al. (2018) was cycled 1000 times to extend the particle tracking simulation. The year 2010 was selected, as the water balance was close to the long-term annual averages observed for the Krycklan catchment. All particles were released at the top of the transient groundwater table the first year. Numerical constraints restricted the number of particles released to 0.5 particles/10 mm modelled groundwater recharge, which corresponds to a total of approximately 0.6 million particles. This number of particles was assumed to be enough to capture the timing of recharge patterns (Fig. 4).

Table ~~3~~4: 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~~[a] Average of Morris and Johnson. (1967). [b]Joyce et al. (2010). [c]~~Average~~[c] Average value between sand and clay. [d]~~Average~~[d] Average value between silt and clay

2.~~3 Testing model results against stream chemistry~~

~~This study was focused on three distinct seasons in Krycklan: winter, spring, and summer. For calculations~~5 Analysis 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, time series of stable isotopes, $\delta^{18}O$ (see Appendix for the $\delta^{18}O$ definition), in stream water were used to compare to~~ 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). Some of the sub-catchments are affected by evaporation from lake surfaces that result in isotopic fractionation (Leach and Laudon, 2019). These fractionations must be accounted for to use the signature as a representation of the groundwater. The isotopic composition was corrected to the percentage of lakes in each sub-catchment, and a regression equation for $\delta^{18}O$ 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. The long-term regression equation for $\delta^{18}O$ lake adjustment for sub-catchments are as follows:~~

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

~~$\Delta\delta O^{18}_{spring/summer} = \text{average } (Wn - Sn)$ 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.to stream chemistry and landscape characteristics

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 the BC concentration increases along the groundwater flow pathway because of weathering (Klaminder et al., 2011). Therefore, a general agreement between the concentration of BC on the one hand and modelled travel times should be possible to distinguish. The 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). Modelling of weathering rates in a soil transect in the Krycklan catchment has indicated that there is kinetic control of the release of BC in the soils (Erlandsson et al., 2016). The release of BC suggests that the longer the groundwater is in contact with the mineral soils, the higher BC concentrations can be expected, similarly to what was observed by Klaminder et al. (2011). Since BC are expected to behave relatively conservatively in these environments (Ledesma et al., 2013; Lidman et al., 2014), their combined concentration was used as a proxy for water age. Sub-catchments with longer travel times would exhibit higher BC concentrations. It has been observed, however, that mires have a 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 to the same 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)

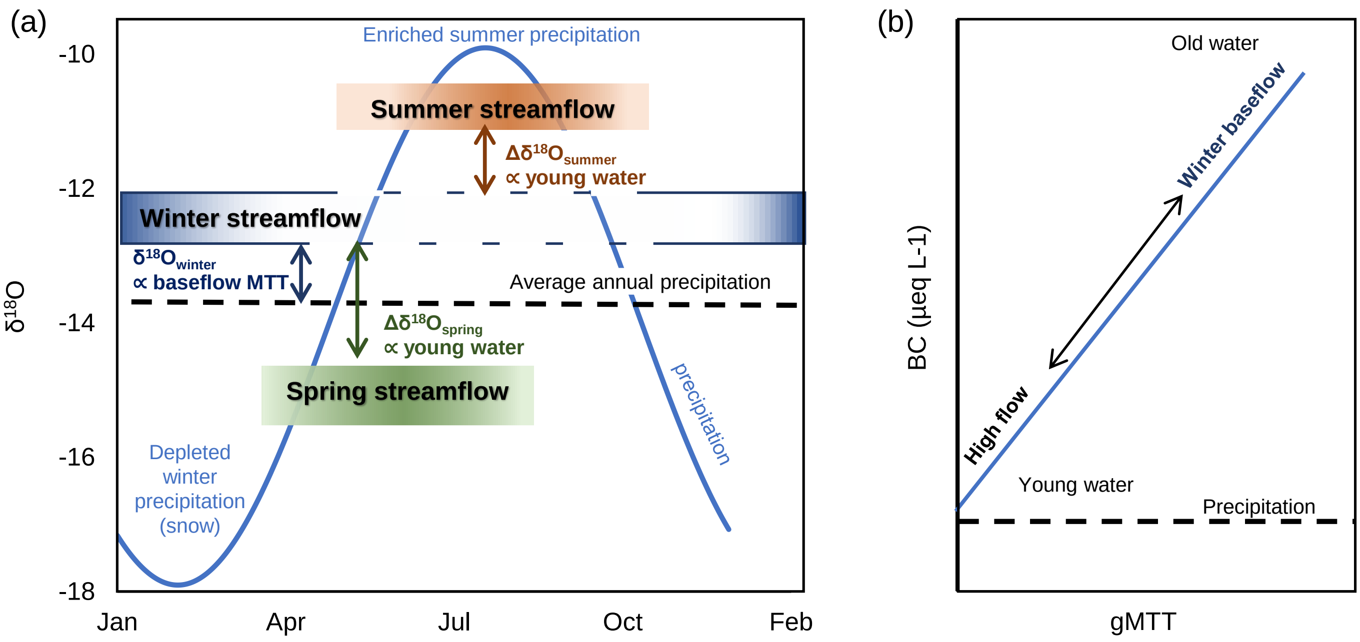


Figure 2: Conceptual figure of connection between water travel time and stream chemistry. (a) The connection between $\delta^{18}O$ and stream water travel time. The sine curve shows the annual variations of $\delta^{18}O$ precipitation composition, and approximate seasonal winter, spring, and summer 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. 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.**~~

~~[a] δ^{18}O Signature (2008-2018), data have been lake adjusted according to equation 2, and delta was calculated using Eq. (1).~~
~~[b] Base cation concentration (2008-2016), data have been mire adjusted according to Eq. (3)~~
~~[c]SD = standard deviation, SEM = standard error of the mean~~
~~[d] Measured precipitation average for isotopes (2007-2016) and measured BC concentration (the year 1997 to 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~~.~~ both annually and for each season. The calculated travel time distributions were analysed using ~~five~~four statistical measurement tools, the arithmetic mean, the geometric mean, median, and the standard deviation~~, the standard error of the mean, and the skew (Appendix). If the standard deviation is higher than half of the~~ (SD). The arithmetic mean, the geometric mean ~~is a better measure of~~and the median are common choices to describe the central tendency of ~~the data set~~a distribution (Destouni et al., 2001; Kaandorp et al., 2018; Massoudieh et al., 2012, 2017; Unlu et al., 2004), which all have their strengths and weaknesses. If the distribution is significantly skewed, the SD is larger than half of the average (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. 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~~In the case of the observed δ ^{18}O and BC concentrations (Table 2), the SD is much smaller than half of the average. Therefore, the arithmetic mean was used to describe the central tendency of the data set. However, if the travel time distribution becomes skewed, the arithmetic mean becomes highly sensitive to the tail of the distribution and produces considerable uncertainty. In these cases, the median and the geometric mean are often better as a measure of the central tendency, of mean travel time (MTT), than the average. However, to compare the MTT of discharged water of different streams, we still wanted the metric to account for the length of the tail. Therefore, we used the geometric mean because the median only states the middle value of a distribution regardless of the tail length (Taagepera, 2008; Unlu et al., 2004; Zhang et al., 1996). However, we provide all metrics, including the arithmetic mean, the geometric mean, median, and SD in the Appendix, Table A1.

~~During~~The MTT was compared to stream chemistry, which is a mix of both groundwater and surface water. In winter, all ~~simulated~~ streamflow ~~contribution comes~~contributions originate from ~~the~~ groundwater. Here the results from the particle tracking ~~reflects~~reflect the actual travel time to the streams. However, ~~during~~in summer and especially ~~during~~in spring, some water will reach the streams ~~as~~via overland flow~~, and therefore~~ (OL), which has not spent ~~zero days~~any time in the ground. Since the particle tracking does not take surface flow into account, ~~two travel times were calculated~~OL was accounted for ~~each site. The first is~~ by reducing 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).~~

~~Overall gMTT=groundwater gMTT*(1-~~MTT by using the OL fraction ~~overland flow) Eq. (4)~~

as a scaling factor. The young water, young water fraction ~~metric~~ was also used as an evaluation criterion. ~~Similar to~~Like previous studies (Kirchner., 2016; von Freyberg et al., 2018; Lutz et al., 2018; Stockinger et al., 2019), we assumed young water fraction to be the sum of all water less than three month old. In our case, this includes all water reaching streams as overland flow and ~~groundwater with age less than three months.~~as young groundwater (<three months). The modelled MTT and young water fraction were also used to identify the main factors determining the age of stream water. The catchment characteristics tested included important terrain factors such as catchment size, slope, and main soil types (Table 1).

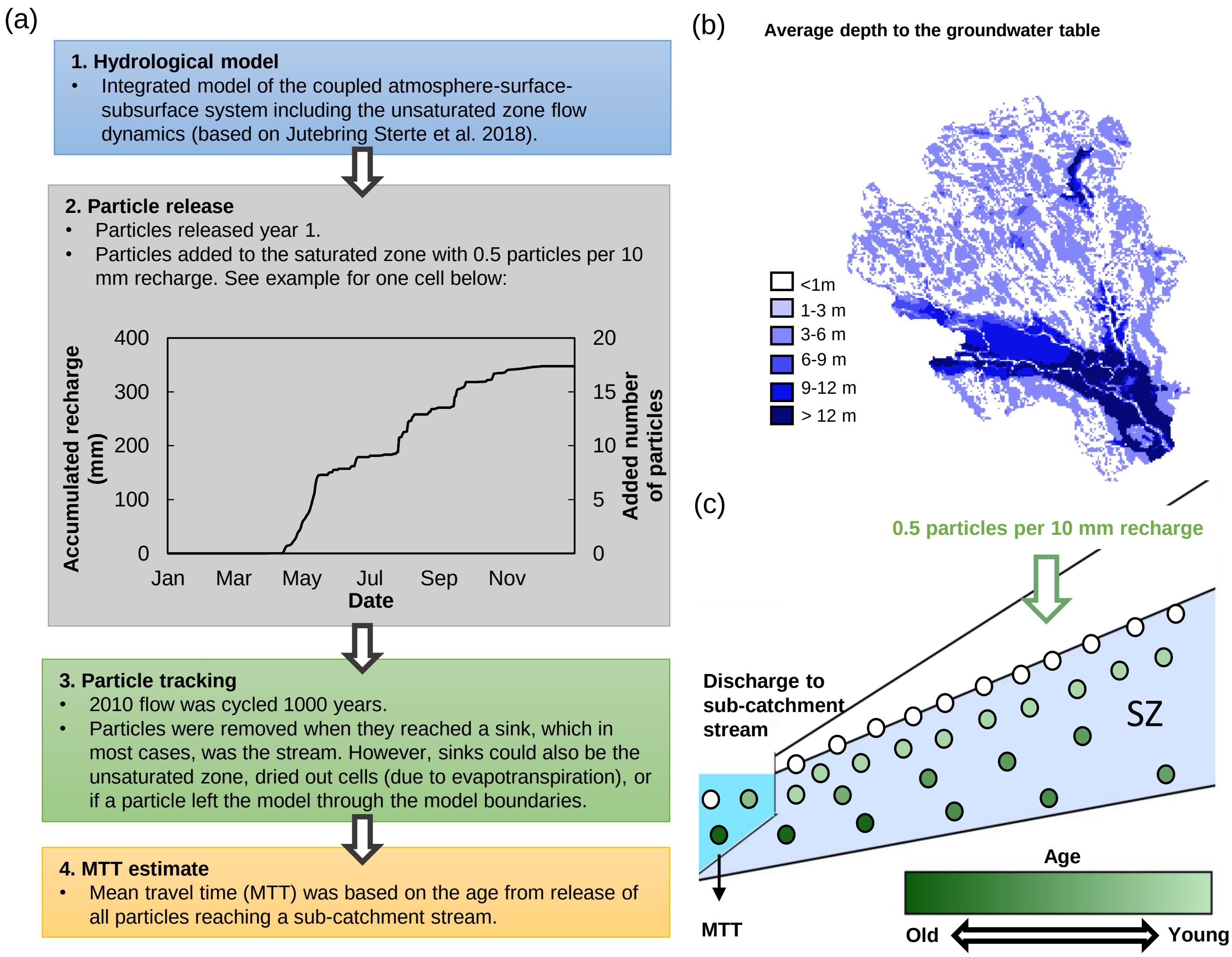


Figure ~~3~~4: Particle model setup. (a) ~~Step by step~~Steps of ~~the~~ particle tracking ~~procedure.~~ (b) Average depth to the groundwater table. The main part of the model area has a calculated depth to the groundwater table between 0-3 m and varied ~~on a daily basis. Note that the top vertical 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.~~daily. (c) Schematic illustration of particle tracking set up. Particles were added to ~~each cell~~groundwater recharge 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~~resulting in increasing ~~in~~ age~~. All particles that reach~~ until reaching 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~~.

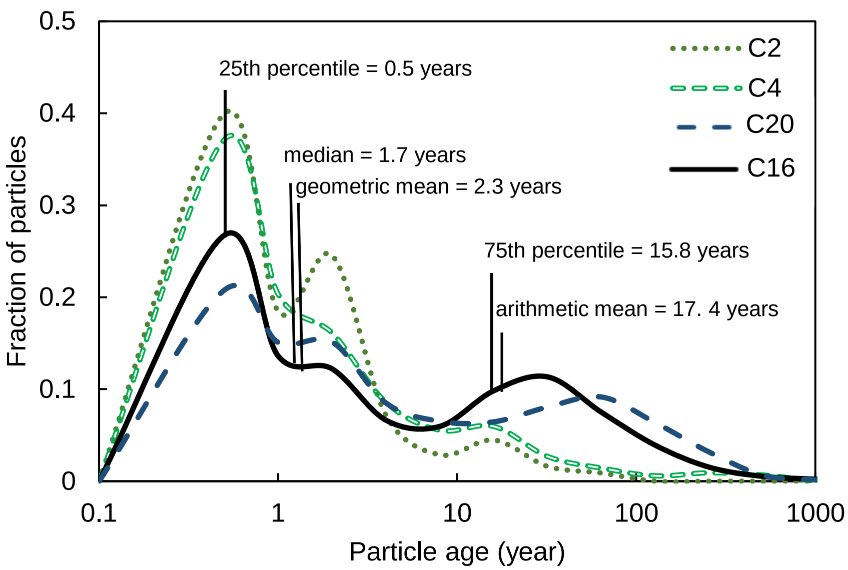
2.5 Catchment characteristic investigation

Correlations between the calculated seasonal gMTT and different catchment characteristics were 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, the 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.

3. Result

3.1 Travel time results

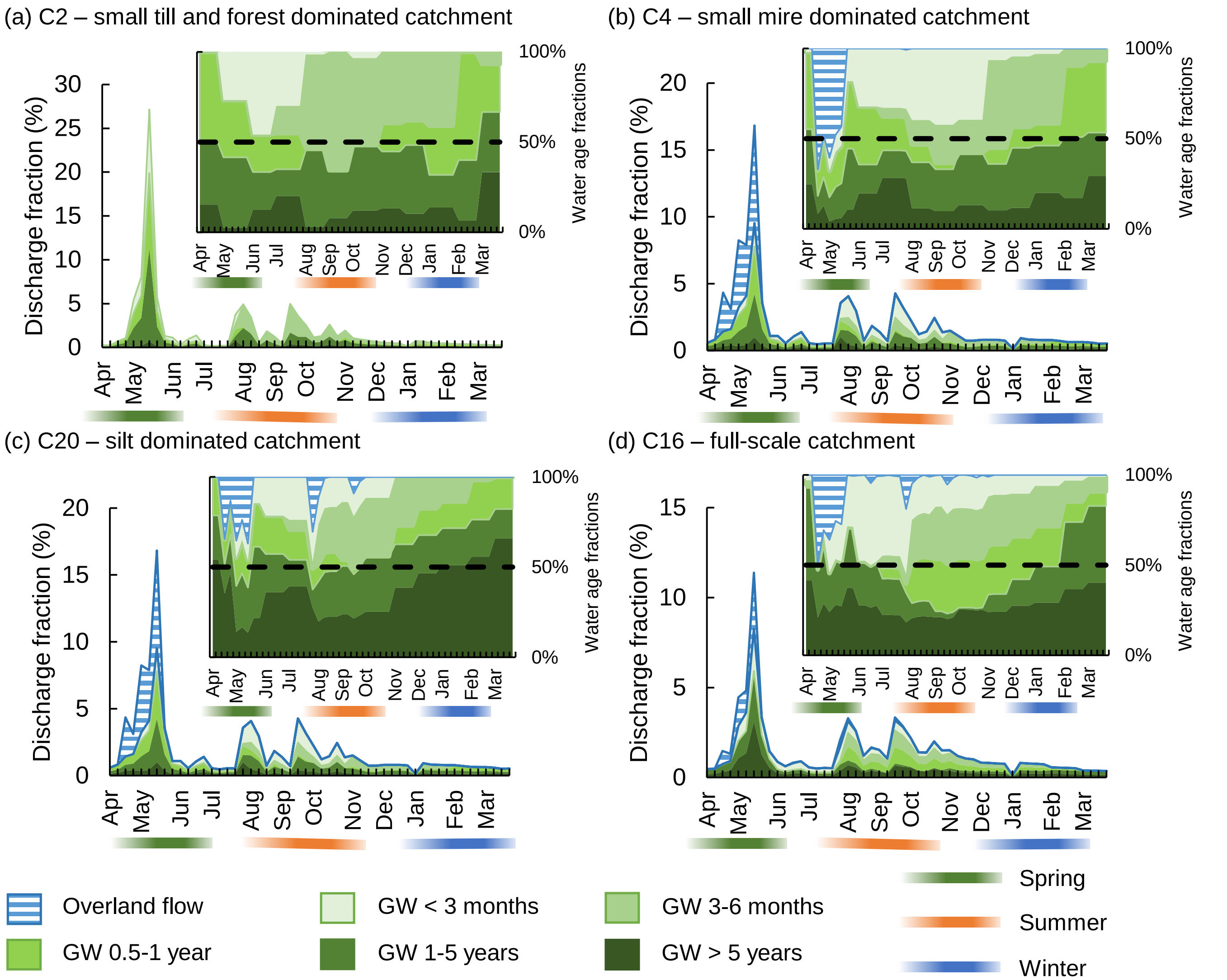
The particle tracking ~~model in Mike SHE was~~ results were used to establish ~~mean~~ travel time ~~in the~~ distributions and MTT of stream water runoff of the 14 sub-catchments~~. The~~ in Krycklan. Since the travel time ~~from groundwater recharge until the groundwater reached a stream was used as an estimation of groundwater~~ distributions were significantly skewed, we assumed that the geometric mean of the travel time~~. The geometric mean (gMTT) was used to describe the central tendency of travel times because of the skewed distribution (~~ distributions provided the best representation of MTT (Table 5, Fig. ~~4). From the particle results, the calculated~~ 5). However, all metrics are stated in the Appendix, Table A1. The annual ~~groundwater and overall gMTT~~MTT_{geo} for all sub-catchments ranged ~~between~~from 0.8 to 3.1 years ~~and 0.8-2.7 years, respectively~~ (Table 5). Most ~~particles~~groundwater discharging to a stream had a travel time of less than one year (34% to 54%). The ~~older groundwater was~~longest stream MTTs were connected to ~~the larger sub-~~silt dominated catchments ~~and~~ such as C16 and C20. We used some sub-catchments ~~with fluvial sediments of C13, C14, C15, C16, and C20. Particles with old ages were generally connected to deep~~ for result representation, but all results are provided in Table 5 and Appendix A1. The displayed sub-catchments were: C2 (small till and forest dominated catchment), C4 (small mire dominated catchment), C20 (small silt dominated catchment), and ~~long flow pathways.~~ C16 (the full-scale Krycklan catchment).



(Edited) **Figure ~~4:~~ 5: Examples of particle tracking results.** The figure shows the ~~**timing**~~age of particles reaching ~~**the sub-catchment outlet.**~~sub-catchment outlets. The solid line showcases the statistics for C16, including the 25th percentile, the median, the geometric mean, the arithmetic mean, and the 75th percentile (Appendix, A1). Moreover, 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**~~three other example ~~**sub-catchments are shown in Fig. 5.**~~distributions, including C2 (small forest and till dominated catchment), C4 (small mire dominated catchment), C20 (small silt dominated catchment).

On an annual basis, a fraction of ~~the~~ water reached ~~a stream~~the streams as overland flow~~, which may enhance or dilute various stream solutes in different ways. The~~. A major part of the overland flow ~~occurs~~occurred during the snowmelt in ~~April to May~~spring, especially in sub-catchments with mires such as C4 (Fig. ~~5). Each site has the oldest age~~ 6). Both the fraction of young water reaching the streams and the MTT_{geo} displayed strong seasonal trends. The longest seasonal MTT_{geo}, 1.2-7.7 years, and the smallest young water fraction were found during the winter season (1.2-7.7 years) and the youngest age in spring and . In winter, the fraction of older water successively increased until the spring snowmelt began in early April. Conversely, the smallest fraction of old discharging water and short MTT_{geo}, 0.5-1.9 years, were connected to events of larger groundwater recharge, such as the spring snowmelt and heavy 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 summer period with little groundwater recharge (Jun-Jul). The oldest groundwater travel times occur during the winter, before the beginning of snowmelt in late March or early Aprilrains.

In spring, mire sub-catchments have the youngest mean travel time during spring snowmelt.had the shortest MTT_{geo}. However, as exemplified by the similar-sized C2 and C4 sub-catchments of C2 (forest) and C4 (mire),, groundwater iswas not renewed to the same extent in mires as in forested sub-catchment (Table 5). The groundwater gMTT of C2 wasmire dominated systems due to a larger fraction surface runoff (Fig. 6). Mire dominated sub-catchments (like C4) displayed stronger seasonal variations in MTT_{geo}, with shorter MTT_{geo} than till dominated sub-catchments (like C2) in spring and longer MTT_{geo} than C2 in winter (Table 5). In C4, the MTT_{geo} reduced from 1.2 years5 to 0.7 years from winter to spring. In C4, groundwater gMTT, while the corresponding change in C2 was reduced from 1.52 to 0.7 years to 1.2 years, despite a larger young water fraction.. The overall gMTTseasonality of C4 decreasedMTT$_{geo}$ was 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 proportionareal coverage of mires combined with low conductive soils (LCS).a larger areal coverage of silt. For example, C20 had an overall gMTTMTT$_{geo}$ 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.compared (Table 5).



(Edited) Figure ~~5~~6: Seasonal fraction of ~~runoff~~discharge to streams. The figure shows the proportion of annual stream ~~water~~discharge arriving as groundwater ~~flow~~ and ~~as direct~~ overland flow. Four sub-catchments are exemplified, including (a) the small ~~forested~~till and forest dominated C2, (b) the small mire ~~-~~dominated C4, (c) the ~~entire Krycklan catchment C16,~~ silt dominated C20, and (d) ~~and the silt-rich C20~~the full-scale Krycklan catchment C16 with mixed mires and forests (extended version in Appendix Fig. ~~A1).~~ A1). The figure showcases the water age fraction discharging to the streams. The fractions are both shown as part of the total annual discharge as well as the water composition. The bands below the months highlight the three investigated seasons, spring, summer, and winter.

(Edited) Table 5: Annual and seasonal (winter, spring, and summer) travel ~~time results.~~times
~~**aMTT** = arithmetic mean (year), **SD** = Standard deviation, **SEM** = standard error of the mean, **OL** = fraction overland flow (%), **gw gMTT**=~~The geometric mean of the ~~particle tracking (groundwater gMTT) (year), **gMTT** = geometric mean of the particle tracking~~travel time distribution (MTT_{geo}) is adjusted for the overland flow. The young water fraction (YWF) includes overland flow ~~according to Eq. (4) (overall gMTT) (year), **Yf** = fraction of surface flow~~ and groundwater that is less than three months ~~(%) (Supporting information)~~(%). An extended version of the results, including arithmetic mean, median, and SD, is included in the Appendix, Table A1.

	Annual		Season - Winter		Season - Spring		Season - Summer	
	MTT_{geo}	YWF	MTT_{geo}	YWF	MTT_{geo}	YWF	MTT_{geo}	YWF
unit	**year**	**%**	**year**	**%**	**year**	**%**	**year**	**%**
C1	1.3	20	3.0	6	1.0	25	0.9	19
C2	0.8	16	1.2	0	0.7	26	0.7	6
C4	0.8	40	1.5	2	0.7	53	0.7	39
C5	0.8	49	2.9	1	0.5	66	0.8	38
C6	0.9	42	2.8	2	0.6	58	0.8	34
C7	1.1	28	2.2	4	0.9	37	0.9	27
C9	1.4	28	3.4	3	1.0	41	1.1	24
C10	1.1	33	2.5	3	0.8	47	0.9	31
C12	1.3	28	2.8	5	0.9	39	1.1	26
C13	1.4	26	3.3	3	1.0	37	1.2	23
C14	2.4	20	5.6	2	1.6	32	1.6	21
C15	1.5	28	3.8	4	0.9	41	1.1	27
C16	2.3	23	5.3	4	1.4	35	1.6	23
C20	2.7	23	7.7	0	1.9	36	1.5	24

3.2 Testing model results to stream isotopic composition and chemistry

In addition to investigating the annual MTT_{geo}, three distinct seasons were evaluated ~~with regards to~~regarding 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~~ sites. In winter, the ~~modelling results, sub-catchments receive older water when the average~~ modelled MTT_{geo} was correlated to isotopic composition ~~is~~(r=-0.80, P<0.01), with older stream water being closer to the long-term precipitation ~~under winter conditions~~average (Fig. ~~6a~~7a). Some of the larger sub-catchments ~~have a~~ had an isotopic signature close to the ~~long-term~~ precipitation average, suggesting ~~that they have reached~~almost complete mixing (~~C15, C14, and~~e.g., 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 ~~$\Delta\delta O^{18}_{spring}$~~$\Delta\delta^{18}O_{spring}$ and the young water fraction was also significant (r=-0.90, P<0.0001, Fig. ~~6c~~7c), 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~~ The same was also true for the summer season, but with the opposite sign of the slope since the summer precipitation was ~~enriched~~heavier compared to the baseflow. The positive correlation was weaker ~~compared to~~than during the spring season~~,~~ but still significant (r=0.80, P<0.001, Fig. ~~6e).~~

7e). The ~~overall gMTT always~~MTT_{geo} had a ~~strong statistical significance~~significant correlation to the BC concentration during all seasons (Fig. ~~6~~7 b, d, and e), ~~generally~~again agreeing with our conceptual model (Fig. 2b). The correlation between the BC concentration and ~~gMTT~~MTT_{geo} was strongest ~~during~~in winter (r=0.88 P<0.0001) and weakest ~~during~~in summer (r=0.79, P<0.001). The sub-catchments with the oldest age and highest BC concentration ~~include some of~~ included the ~~largest~~ sub-catchments ~~of C14~~with larger areal coverage of silt, for example, C16 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.

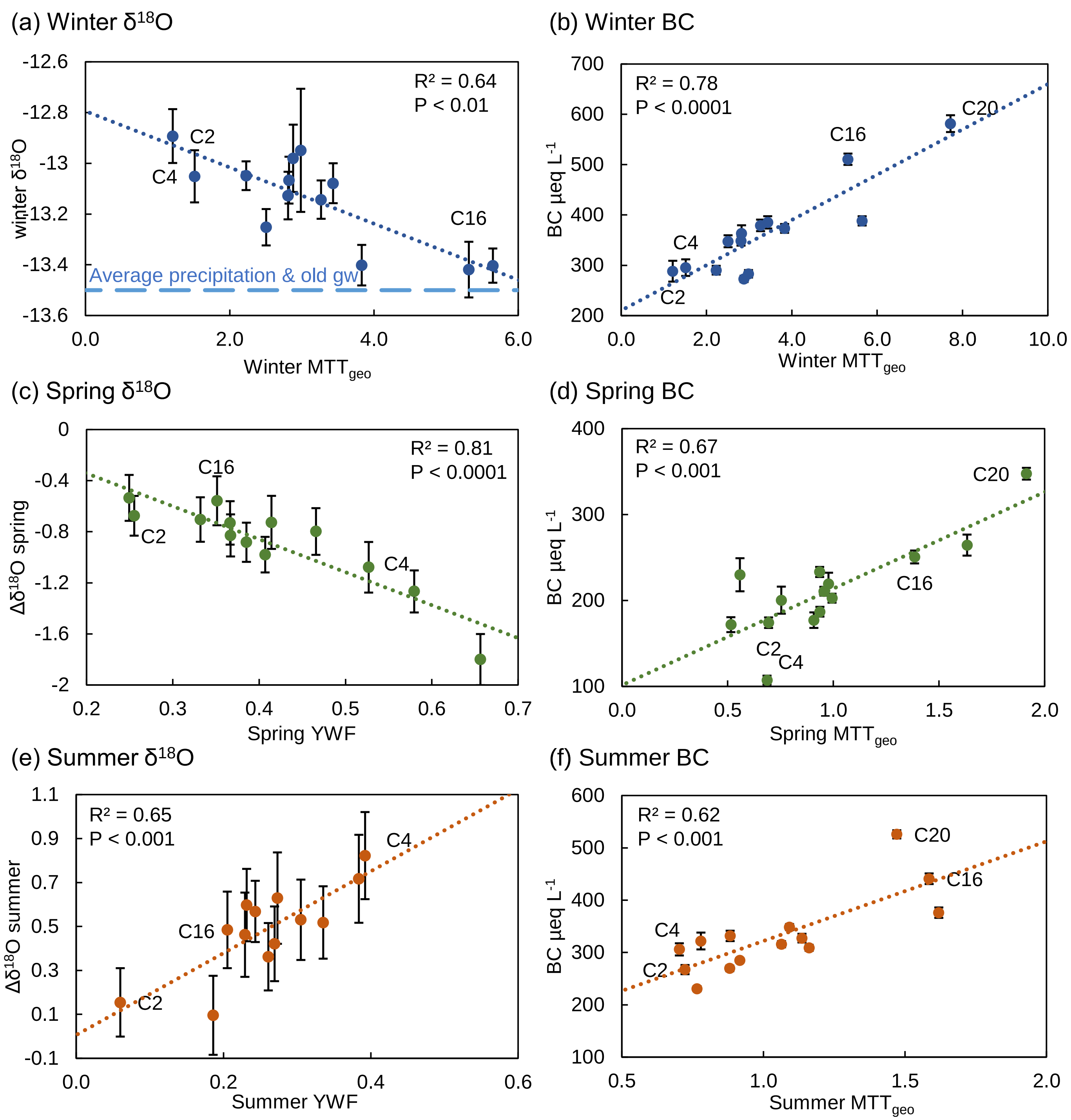
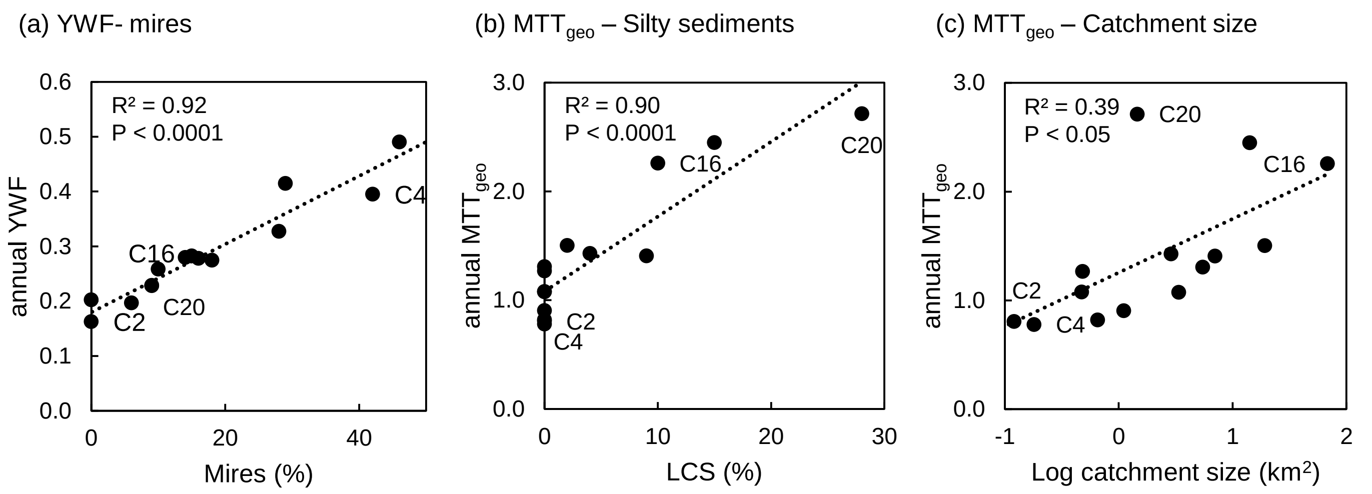


Figure ~~6~~7: Results of seasonal young water fraction (YWF) and MTT_{geo} compared to stream ~~chemistry, $\delta^{18}O$,~~isotopic composition and base cation ~~(BC)~~ concentration. Note that $\delta^{18}O$ results are for 13 sites, while the BC record comprises all 14~~ sites~~. The sub-plots (a) to (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~~MTT$_{geo}$ in winter, spring, and summer, respectively. The standard error of the mean (SEM) ~~is~~ shown ~~for the~~as whiskers denotes variations in field observations.

3.3 Model results compared to catchment characteristics

The main catchment characteristics ~~found to be~~ correlated to ~~gMTT~~ MTT$_{geo}$ and young water fraction were catchment size, the ~~fraction~~ areal coverage of low conductive ~~soils (LCS),~~silty sediments, and the ~~fraction~~areal coverage of mires. The strongest positive correlation was found between the young water fraction and the ~~proportion of sub-catchment mire~~areal coverage of mires (r=0.96, P<0.0001~~), as well as gMTT and low conductive soils (LCS)~~). There was also a strong positive correlation between MTT$_{geo}$ and the areal coverage of silt (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.~~Fig. 8). A positive correlation between catchment size and ~~gMTT was also found. The correlation~~MTT$_{geo}$ was ~~relatively~~also found, albeit weak due ~~mainly~~ to one ~~outlier~~catchment, C20, yet significant (r=0.63, P<0.05) (Fig. ~~7~~8). However, the catchment size ~~is~~was also correlated to the ~~fraction~~areal coverage of ~~LCS~~silt, 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~~.~~ with sorted sediments. The annual and seasonal patterns ~~are generally~~were similar (Table 6). However, the positive correlation between mires and the young water fraction was lost ~~during~~in 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~~MTT$_{geo}$ and the young water fraction was found ~~in~~for the annual ~~average~~ and ~~during~~ spring seasonal results but ~~was~~were lost ~~during~~for the summer and winter.



(Edited) **Figure ~~7: Travel time important~~ 8: Catchment characteristics~~.~~ are important for travel times.** The figure shows the annual averages~~.~~: (a) the areal coverage of mires and the young water fraction~~,~~ (YWF), (b) ~~mires~~areal coverage of silt and ~~gMTT (year), (c) low conductive soils (LCS)~~MTT$_{geo}$, and ~~gMTT (year), and (d)~~ (c) catchment size and ~~gMTT (year). The gMTT has been adjusted for the overland flow for each season, according to Eq. (4).~~MTT$_{geo}$.

(Edited) **Table 6: Correlation matrix – young water ~~fractions, gMTT, and~~ fraction (YWF), geometric mean travel time (MTT_{geo}), and catchment characteristics.** The catchment characteristics include the log catchment size, km^2 (Log C.-size), the areal coverage of mires, and the areal coverage of silt. The table includes ~~yearly calculations (white~~annual (grey), winter ~~calculations~~ (blue), spring ~~calculations~~ (green), and summer ~~calculations~~ (orange~~).~~) results. Darker colours show ~~when the absolute value of.~~ |r| > 0.~~50~~5 with the connected p-value according to [a] p~~>~~<0.05~~, [b] a p<0.05,~~ and ~~c~~[b] p~~<~~>0.~~01~~05.

	Winter season					Summer season				
	Log C.-size	Mire (%)	Silt (%)	MTT_{geo}	YWF	Log C.-size	Mire (%)	Silt (%)	MTT_{geo}	YWF
Log C.-size	1					1			0.91 [a]	
Mire (%)		1	0.92 [a]		0.64 [a]		1	0.80 [a]	-0.50 [b]	0.68 [a]
Silt (%)	0.58 [a]		1		0.58 [a]	0.58 [a]		1		0.58 [a]
MTT_{geo}	0.63 [b]	-0.51 [b]	0.90 [a]	1		0.55 [a]	-0.55 [a]	0.92 [a]	1	
YWF (%)		0.96 [a]		-0.53 [b]	1		0.95 [a]		-0.52 [b]	1
	Annual					Spring season				

4 Discussion

Particle tracking in the Mike SHE model showed promising results in its ability to capture provided valuable insights into the annual and seasonal mean travel times (MTT$_{geo}$) across the 14 Krycklan sub-catchments. Travel times of stream water contribution The modelled MTT$_{geo}$ and the young water fractions were relatedstrongly correlated to observed stream winter $\delta^{18}O\delta^{18}O_{winter}$ signatures, $\delta^{18}O$ seasonal shiftsvariation in $\delta^{18}O$, and base cation (BC) concentrations. This model validation suggests that particle tracking could therefore be a is a useful complementary tool to tracer-based studies of travel time, especially at least in snow-dominated catchments, areas with pronounced seasonality, and streams dominated by old groundwater (older than 4-5 groundwater (> four years). In this study, Overall, we found the hydrologic conductivity of the soil to be that soil type was the most important parameter for the water age and mires to be an important factor regulating the young water fraction.

4.1 Model testing variable explaining MTT$_{geo}$ and **uncertainties** that mires are an important landscape feature regulating the young water fraction in spring (Fig.8).

Particle tracking in Mike SHE is associated with some uncertainties

4.1 Model assumptions and limitations. A comparison of estimated travel times

Comparing the results from this modelling study to previous studies of mean travel times (MTT) for one of the Krycklan sub-catchments (C7) shows, however, that investigations of MTT conducted in the C7 sub-catchment demonstrates that different model approaches gave have provided similar results. While our study estimated a MTT time to suggested a MTT$_{geo}$ of 1.1 years, and a median of 0.8 years (Appendix, Table A1), Peralta-Tapia et al. (2016) calculated a ten-year average travel time MTT of 1.8 (minimum 0.8 and maximum 3.3) years using long-term isotopic data and a gamma transformation method. In a another recent study using the same data in the Spatially distributed Tracer-Aided Rainfall--Runoff (STARR) model for the same stream, the median of the travel time distributionage was approximatedestimated to be 0.9 years for the same sub-catchment (Ala-aho et al., 2017). The close agreement withbetween the previous model runsdifferent studies 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. reliability of the results. However, like all modeling techniques, particle tracking in Mike SHE is associated with some uncertainties and limitations.

In contrast to the Mike SHE flow model, which estimates both the groundwater and overland flow pathways, the particle tracking model is restricted to the subsurface hydrological component. This is a limitation in the modelling approach as water reach streams as a mix of groundwater and overland flow. Therefore, to allow for actual MTT$_{geo}$ estimates, we corrected the results by reducing the estimated MTT_{geo} using the overland flow from the flow model as a scaling factor. This uncertainty primarily affects the mire dominated sub-catchments that have a large fraction of overland flow, especially during the spring.

Another uncertainty related to the particle tracking model in Mike -SHE is related to the travel time from the point of infiltration ~~under~~through the unsaturated ~~condition~~ soil horizons to the saturated groundwater ~~recharge, which,~~. Due to technical limitations, ~~was not~~ this travel time cannot be accounted for in the particle tracking calculations. Particles are placed ~~in~~ at the groundwater table proportionally to the groundwater recharge (Fig. ~~3).~~ 4). Therefore, the main ~~portion~~ fraction of particles ~~is~~ introduced to the model occurs at high ~~recharge~~ infiltration rates when the groundwater level is ~~shallow across the catchment.~~ close to the soil surface. Under these conditions, the water has, in most cases, spent a relatively short time in the unsaturated zone. However, some particles are also introduced when the groundwater level is lower, such as early snowmelt or ~~after~~ following 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~~ Under such conditions, the model uncertainty increase. In this context, the smallest potential uncertainty occurs in mires that seldom experience a 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.~~ below one meter below the soil surface. The uncertainty becomes somewhat larger in the till areas where the unsaturated zone on average is above 1 m but can extend down to 3 m below the ground during low flow. C14 and the lower part of C16 are exceptions to these relatively shallow saturated conditions as a deep esker traverses the sub-catchments resulting in a groundwater level up to 10 m below the soil surface (Fig. 1). Accounting for the travel time from infiltration to recharge could impact the results and ~~give~~ provide, especially for C14 ~~older~~ and C16, longer MTT than if the groundwater level ~~was~~ were at the same level throughout the whole catchment. This limitation primarily affects catchments with ~~long~~ the longest MTTs and, therefore, does not seriously question the general ~~patterns that were observed.~~

~~We used the~~pattern observed. The distance from the ground surface to the groundwater table is for most model cells much shorter than the distance to the nearest stream ~~winter isotopic composition and BC concentration to test Mike SHE's ability to capture the variability of travel times in the 14 Krycklan sub-catchments. Based on our results, we found significant and robust correlations between the winter isotopic signature $\delta^{18}O$ as well as the stream chemistry, on the one hand, and the calculated travel times on the other (Fig. 6). Theoretically, infinitely long travel time would result in a stream water isotopic signature approaching the long-term average precipitation input (Fig 2). In contrast, the 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.~~so most of the transit time should be related to the groundwater flow rather than to percolation. Although water, especially during dry conditions, no doubt can spend considerable time in the unsaturated zone, it must also be acknowledged that this water volume is small compared to the groundwater inventory in the saturated zone so its impact on the average MTTs should be relatively small.

4.~~1.1 Testing model results against~~2 Seasonality of isotopic composition

~~According to~~Following the conceptual model (Fig. ~~2), older baseflow water should result~~2), patterns in stream isotopic signatures can be explained by seasonal changes in ~~an isotopic signature closer to the precipitation average. There was~~ travel times. The modelling results show that all sub-catchments discharged the oldest water in winter, somewhat younger water in summer, and water of the youngest age in spring. When winter arrived, the main precipitation was snow resulting in that groundwater recharge effectively ceased, which caused an increasing proportion of old groundwater discharging into the streams (Fig. 6). In agreement with our conceptual model (Fig. 2), a strong negative correlation between ~~groundwater age~~winter MTT_{geo} and the ~~streams~~ isotopic ~~signature~~stream signatures during winter baseflow was observed (Fig. ~~6a), suggesting that the model produces credible~~7a). At an average 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~~four years, it can be expected that the groundwater has reached full mixing. Hence, older water can no longer be accurately ~~quantifiable~~quantified using water isotopes only due to amplitude loss (Kirchner., 2016). These theoretical considerations strengthen ~~our~~the results of a winter ~~MTT of 4-6~~MTT_{geo} between four and six years for the larger sub-catchments ~~and provided new insights into travel times for these systems~~as their stream isotopic signatures were close to the long-term precipitation average and, therefore, should have reached complete mixing.

~~In spring, the~~ When snowmelt began in late April or early May, the MTTs consistently decreased in all sub-catchments. The fraction of young groundwater in different sub-catchments was well reflected in the change in the isotope signal (Fig. 7). For snowmelt in spring, the calculated young water fraction was used to evaluate the proportion of water reaching the stream ~~as~~ through rapid pathways, including overland flow. It is well established that the difference in stream isotopic signature between ~~the previous~~ winter baseflow and ~~stream isotopic signature~~ spring peak flow at snowmelt (~~$\Delta\delta O^{18}_{spring}$~~$\Delta\delta^{18}O_{spring}$) is mechanistically ~~related~~linked to the amount of ~~young~~new water reaching the stream (~~Laudon et al., 2007;~~ Tetzlaff et al., 2009). In agreement with this, we found a strong ~~connection~~statistical relationship between ~~$\Delta\delta O^{18}_{spring}$~~$\Delta\delta^{18}O_{spring}$ and ~~our~~the 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).~~fraction (Fig. 7c). 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~~.~~., 2004, 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~~

Similar to the conditions in spring, the conceptual model predicted that ~~$\Delta\delta O^{18}_{summer}$~~the difference in stream isotopic signature between winter baseflow and summer flow, $\Delta\delta^{18}O_{summer}$, should ~~also~~ be correlated to the young water fraction in summer, but with the opposite sign, due to ~~the enriched~~isotopically heavier summer ~~rain~~rains (Fig. 2). A larger inter-annual variation in precipitation and ~~evapotranspiration variability~~high ET 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~~evident than compared to the ~~spring $\Delta\delta O^{18}$~~ $\Delta\delta^{18}O_{spring}$, there was still a ~~strong connection~~significant correlation between the average ~~summer $\Delta\delta O^{18}$~~$\Delta\delta^{18}O_{summer}$ and the modelled young water fraction (Fig. ~~6e).~~7e).

4.~~1.2 Testing model results against~~3 Controls of travel times on base cation ~~concentration~~concentrations

~~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.~~The annual and seasonal average BC concentrations were positively correlated with the MTT_{geo} (Fig. 7b, 7d, and 7f). Since the weathering rates generally arewere assumed to be kinetically controlled, i.e., and hence related to the travelexposure time, such stream chemistry variables of water to minerals, spatial and temporal variability in BCs can be used as a relative indicator for stream water age as long astransit time if the mineralogy remains comparatively homogenous (Erlandsson Lampa et al., 2020). This study showed that the modelled travel times were significantly correlated to the BC concentrations. However, reducing weathering to a matter of travel times may be an oversimplification as the rate is also affected by, for example, chemical conditions, differences in mineralogy, and particle size distributions. and the chemical conditions in the groundwater. However, previous research in the Krycklan catchment has indicatedsuggested that the chemical composition of the local mineral 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). Therefore, we dodid not expect mineralogical differences between soil types to have a significantsignificantly impact on the release of cations. The One exception is, however, are peat deposits, which strongly affect the cation concentrations, and that on a landscape scale. The effect of the peat 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 soils will have less surface area per volume unit, therefore allowing for less weathering. However, such 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 travel times.

Despite arguments that can be made against the use of cationsBCs as tracers, they still offer a complementary possibility to test the model performance of the model.(Abbott et al., 2016). As emphasized by McDonnell and Beven (2014),) emphasized, the inclusion of tracers in hydrological models is necessary to ensure that a model reproduces the speed of flow, which is an important parameter when assessing travel time distributions. InFor 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; Hrachowitz et al., 2013). 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 our model results and the system understanding of catchment-scale travel times and their connection to biogeochemistry. More specifically, the results increase the likelihood that the model is producing credible results for the right reasons.provides additional support that our modelling of these processes – and thus the entire system – was reasonable and consistent with the empirical data.

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

The main catchment characteristics found that affect the ageAll sub-catchments showed similar synchronicity in the seasonal patterns in MTT_{geo} and young water fraction were low conductive soils (LCS) and, but catchment characteristics influenced the fraction of mires (Fig. 7, Table 6). The most significant factor for the mean travel times was related to the proportion of low conductive soils (LCS),magnitude of the seasonal patterns across the landscape. On a landscape level, the main causal mechanism determining the annual MTT_{geo} was the areal coverage of silt, which overshadowed the importance of other catchment size.characteristics (Fig. 8, Table 6). This finding stands in contrast to earlier studies in Krycklan by Peralta-Tapia et al. (2015) and Tiwari et al. (2017) havethat suggested that the MTT of groundwater istravel times are nonlinearly linked to the catchment size. However, We found that the silt-rich but relatively small silt dominated C20 to becatchment was a distinct outlier to thissuch a scale-dependent pattern, indicating that catchment size may not be the underlyingprimary factor causing high MTTsdetermining the variability (Fig. 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~~ most likely that C20 is the only relatively small sub-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.~~in the silt-dominated areas. Hence, the long travel times in ~~relation to the relatively small catchment size in~~ C20 ~~means~~suggest that the groundwater flow velocity ~~generally~~ is ~~lower~~slower than elsewhere~~. Nevertheless, the~~ in Krycklan, despite the fact that the average catchment slope ~~of C20~~ is steeper than ~~in~~ comparably sized sub-catchments in till areas~~, so the topographical possibilities to build up hydraulic gradients that can drive the water transport should be large~~ (Table 1 and Fig. 1). ~~The fluvial sediment deposit~~Similarly, silt may also explain the ~~relatively~~ long travel times ~~of~~at C14 and C16. Although C14 is smaller than C15, which mostly lacks ~~LCS, it~~silt, C14 still has a longer ~~MTT.~~

MTT_{geo}. In contrast, MTT_{geo} in C15 is much closer to C12 and C13 ~~in MTT~~, even though the C15 catchment is ~~almost~~ twice ~~as large~~the size (Table 5). The results suggest that the critical difference between these sub-catchments and other sub-catchments is related to the soil hydraulic conductivity ~~of the soils~~ rather than the catchment size. ~~Without the contribution of water from headwater catchments with fine soils (such as C20), the MTT of sub-catchments like C14 and C16 would probably be much closer to that of the other smaller till-dominated sub-catchments.~~ The results~~, therefore,~~ further emphasize that one cannot ~~generally~~ assume that the travel time would increase with catchment size unless the distribution of different soils ~~is~~are 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.~~

~~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 silt fraction effect is especially prominent in winter when the range in MTT_{geo} is between one and almost eight years. The seasonal MTT_{geo} change from winter to spring is also largest for the silt dominated catchments, with, for example, six years difference for C20 compared to two years for the similar-sized till dominated sub-catchment C6. These intra-annual variations can also be linked to another landscape feature, namely the areal coverage of mires. Mires affected the young water fraction but only when new precipitation or snowmelt input into the system occurred in spring and summer. The lack of synchronicity between the response of mire and silt areas caused greater annual MTT_{geo} variation for sub-catchments with both features. For example, the MTT_{geo} for C4, dominated by mires, decreased from 1.5 years to 0.7 years from winter to spring. In contrast, winter MTT_{geo} for the C20 catchment dominated by silt was 7.7 years, which decreased to 1.5 years in spring. The results also show that groundwater recharge is affected by the soil frost in mires. For example, C4 showed more variations in its seasonal MTT_{geo}, although C2 (dominated by forest and till) and C4 (dominated by mires) had almost an equal annual MTT_{geo} (Table 5). In spring, the MTT_{geo} was shorter in C4 than in C2 due to surface runoff on the frozen mire, and in winter, the MTT_{geo} in C4 was longer than in C2 due to the lower recharge and displacement of older water during the spring. Besides the slightly higher specific discharge from mires (Karlsen et al. 2016), empirical-based studies suggest that the soil frost on mires causes a large fraction of overland flow (Laudon et al. 2007; 2011).

Earlier studies have demonstrated that fluxes of old groundwater are more stable throughout the year than younger groundwater showing a more variable temporal pattern (Rinaldo et al., 2011; van der Velde et al., 2015; Kaandrop et al., 2018). In our system, such a pattern can mechanistically be linked to till soils dominating most sub-catchments, where the groundwater response to precipitation events can be described by transmissivity feedback processes (Bishop, 1991), caused by the fact that the hydraulic conductivity increases exponentially towards the soil surface. When water infiltrates the ground, the water table rises and activates more conductive soil layers, resulting in rapid increases in the lateral flow. This implies that much of the water transport in till soil occurs relatively close to the surface, while the groundwater in deeper layers is more stagnant, which further explains the relatively short and consistent MTTs of till soils. Measurements of chlorofluorocarbons (CFCs) further support that deeper groundwater water transport in till soils in Krycklan is slow. Not far below the groundwater table CFCs have indicated that the groundwater can be several decades old, suggesting that most of the groundwater transport occurs close to the surface (Kolbe et al., 2020). Consistent with this explanation, silt dominated areas, that have more consistent hydrological conductive with soil depth, had much longer MTTs than comparatively sized sub-catchments underlain by till soils (Fig 6, Fig. 8 and Appendix Fig. A1).

5 Conclusions

The combination of stable water isotopes, stream water chemistry, and particle tracking provided a consistent picture of ~~how~~ the hydrological functioning of a boreal ~~landscape functions hydrologically~~catchment and what processes and factors are ~~of~~

importance. Hence, this system approach not only strengthensmost important for regulating travel times and pathways. We identified specific landscape characteristics that impact the credibilityseasonal distribution of these specific modelling results but also more broadly confirms the applicability of process-based numerical modelling and particle tracking under the complex hydrological conditionstravel times by combining a distributed hydrological model with, for example, long dry winters, temporary soil frost, and intensive spring floods that prevail in the boreal region. empirical observations from 14 nested sub-catchments. In the wake of a changing climate and intensified pressure from forestry and other formstypes of land use, this study provides a useful foundationbaseline for assessing the often intricate connections and feedbacks between hydrological and biogeochemical processes throughout the boreal landscape. Our results showed that water travel times could vary considerably on annual and seasonal scales between different types of catchments. This was mainly related to soil properties, with low conductivity silty sediments leading to the longest travel times on annual and seasonal timescales. In contrast, mires lead to increased fractions of young water, and hence shorter travel times, but mainly in spring when the soil was frozen. As a result of the lower groundwater recharge during the snowmelt, however, the MTTs in mires were, in turn, longer than in forests during the winter. In a warmer climate with reduced soil frost and decreased snowmelt input, we would expect the effect of mires to be reduced while the impact of till and silty sediment soils likely will remain relatively unaffected.

6 Data availability

The Data from the Hydrological Research at Krycklan Catchment Study is available in Svartbergets open database (www.slu.se/Krycklan). The software (Mike SHE/Mike 11) is available at https://www.mikepoweredbydhi. com/

7 Author contribution

Elin Jutebring Sterte and Hjalmar Laudon were responsible for the design, conceptual ideadesigning, conceptualizing, and evaluation ofevaluating results in collaboration with the other co-authors. Elin Jutebring Sterte and Emma Lindborg were responsible for numerical modelling. Elin Jutebring Sterte prepared the manuscript and figures. 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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Appendix

In the Appendix, ~~the~~ statistical measurements for ~~the~~ particle tracking and isotope data are further explained, used in e.g., table 5 and table A1. The statistics used in this study include arithmetic mean (Eq. A1), geometric mean (Eq. A2), standard deviation (Eq. A3), standard error of the mean (Eq. A4). The isotopic signature of $\delta^{18}O$ has been calculated as Eq. A5.

~~aMTT =~~ $\text{Arithmetic mean of the travel time distribution} = \left(\frac{1}{n}\right)\sum_{i=1}^{n} ai$ (Eq. A1)

~~gMTT =~~ $\text{Geometric mean of the travel time distribution} = 10^{\left(\frac{1}{n}\right)\sum_{i=1}^{n}\log(ai)}$ (Eq. A2)

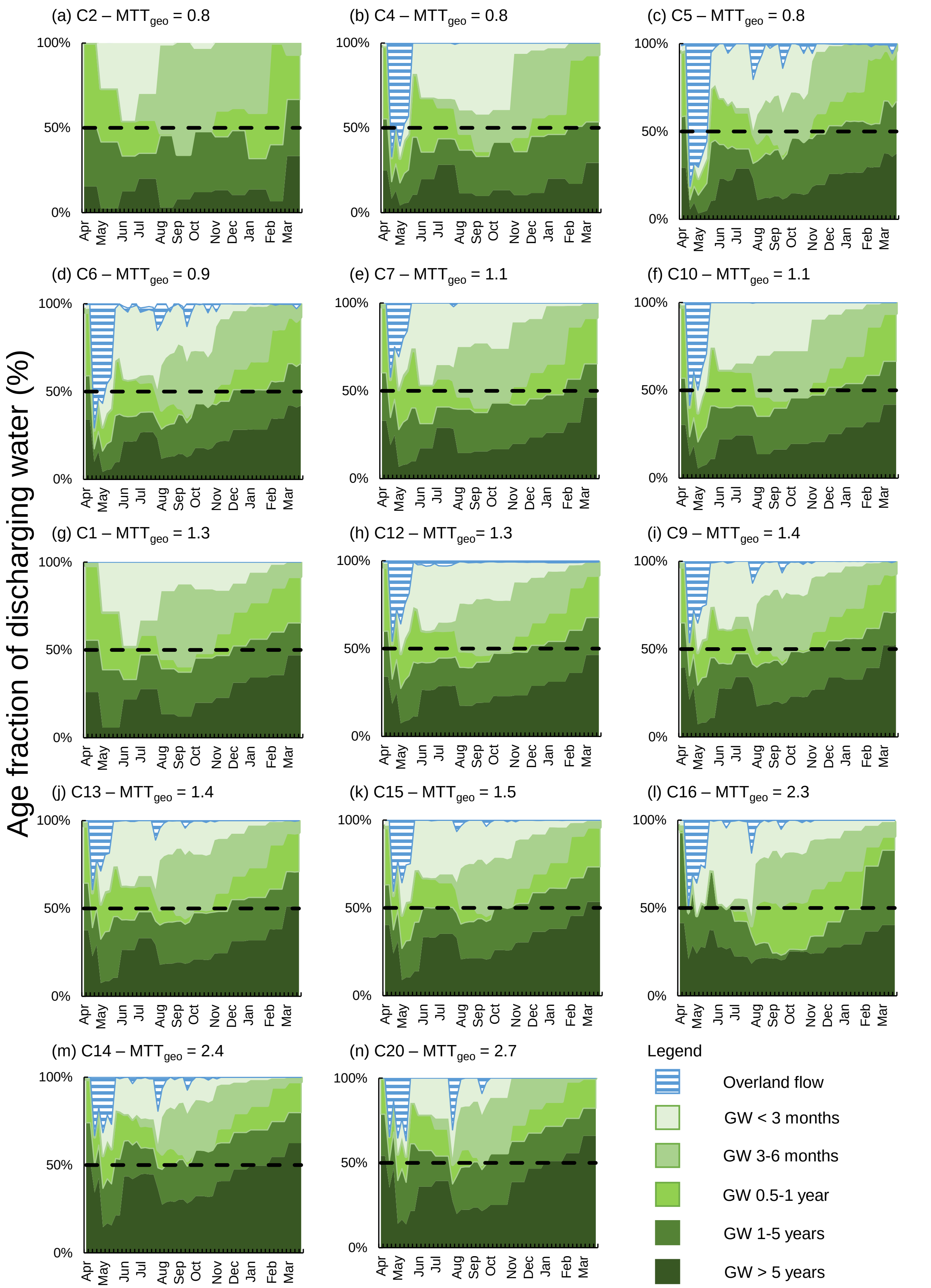
$\text{SD} = \text{standard deviation} = \sqrt{\frac{\sum(ai-aMTT)^2}{N}}$ (Eq. A3)

$\text{SEM} = \text{standard error of the mean} = \frac{SD}{\sqrt{n}}$ (Eq. A4)

$\delta = \left(\frac{R_{sample}}{R_{standard}} - 1\right)$ ‰, whereas $R = \frac{18O}{16O}$ (Eq. A5)

whereas: ai= data set values, n=number of values.

The Appendix also includes an extended version of table 5 and Fig. ~~5~~6, including all sub-catchments (Table A1 and Fig. A1). The figure shows the fraction of different age groups to the streams of the sub-catchment ~~is~~in Krycklan ~~on a yearly basis.~~annually. The figure shows both the groundwater fraction (age fraction calculated using the ~~results from the~~ particle tracking results) and the fraction of simulated direct runoff. The table shows more statistical information regarding the travel time distribution, including the Skew, SD, and SEM.



(Edited) Figure A1: Age fraction of ~~wate4r~~water discharging to the streams of Krycklan~~.~~, in increasing annual geometric mean travel time, MTT$_{geo}$ (up left to down right). The ~~figure~~black vertical line shows the ~~proportion~~50 % mark for visual aid. All charts begin in spring (late March/early April) and end in winter (early March).

(Edited) Table A1: Extended version of ~~water that comes to the stream as groundwater flow and as direct surface flow.~~ **Table 5 - Annual and seasonal (winter, spring, and summer) travel time results.**

The ~~direct surface flow has no calculated age since~~table includes the travel time results based on particle tracking ~~only includes the groundwater.~~ ***before*** taking the overland flow into account. The results include arithmetic mean (A), median (M), geometric mean (Geo), skew, standard deviation (SD), and standard error of the mean (SEM).

	Annual						Season - Winter					
	A	**M**	**Geo**	**Skew**	**SD**	**SEM**	**A**	**M**	**Geo**	**Skew**	**SD**	**SEM**
unit	**year**	**year**	**year**	**-**	**-**	**-**	**year**	**year**	**year**	**-**	**-**	**-**
C1	10.1	1.0	1.3	4.1	27	0.4	18.8	1.4	3.0	2.7	36	1.2
C2	2.2	0.9	0.8	5.0	5	0.2	2.7	0.7	1.2	2.4	4	0.5
C4	7.7	0.8	1.0	7.5	34	1.0	10.5	1.1	1.5	6.6	42	2.7
C5	15.2	1.0	1.3	6.4	61	1.0	30.4	1.4	2.9	4.1	84	3.2
C6	13.7	0.8	1.2	6.8	51	0.6	25.9	1.4	2.8	4.6	69	1.8
C7	8.0	0.8	1.2	7.4	25	0.4	13.2	1.3	2.2	5.6	32	1.1
C9	13.2	1.0	1.6	6.7	38	0.3	21.6	1.6	3.4	5.0	47	0.7
C10	10.9	0.9	1.2	6.4	35	0.2	16.5	1.4	2.5	4.5	40	0.6
C12	11.9	1.0	1.4	5.5	33	0.2	17.6	1.0	2.8	4.0	37	0.4
C13	13.3	1.0	1.5	8.0	43	0.2	21.6	1.6	3.3	6.4	53	0.5
C14	18.3	1.9	2.7	7.8	54	0.2	26.4	6.6	5.6	6.8	60	0.4
C15	14.3	1.0	1.7	8.7	43	0.1	21.9	2.4	3.8	6.7	49	0.3
C16	17.4	1.7	2.5	8.5	50	0.2	25.3	6.7	5.3	7.3	57	0.2
C20	21.9	1.8	3.1	6.0	52	0.6	32.9	9.7	7.7	5.8	55	1.1

	Season - Spring						Season - Summer					
	A	**M**	**Geo**	**Skew**	**SD**	**SEM**	**A**	**M**	**Geo**	**Skew**	**SD**	**SEM**
unit	**year**	**year**	**year**	**-**	**-**	**-**	**year**	**year**	**year**	**-**	**-**	**-**
C1	5.2	1.0	1.0	6.8	19	0.5	8.4	0.4	0.9	4.5	25	0.8
C2	1.6	1.0	0.7	6.1	3	0.2	2.7	0.4	0.7	4.2	8	0.6
C4	5.7	0.3	1.2	10.8	27	1.5	9.1	0.4	0.7	6.1	39	2.0
C5	9.9	1.0	1.2	9.3	50	1.5	11.3	0.4	0.8	7.6	52	1.7
C6	9.1	0.9	1.0	9.5	45	1.0	9.9	0.4	0.8	8.2	42	1.0
C7	5.5	1.0	1.1	11.2	19	0.6	7.5	0.4	0.9	7.5	27	0.8
C9	8.2	1.0	1.3	10.8	31	0.4	11.2	0.8	1.2	7.0	34	0.5
C10	8.0	1.0	1.1	8.2	32	0.4	9.0	0.4	0.9	6.3	31	0.4
C12	8.2	1.0	1.2	7.6	29	0.3	10.2	0.8	1.1	5.3	29	0.3
C13	7.8	1.0	1.2	10.8	30	0.3	12.5	0.8	1.2	8.8	45	0.4
C14	12.2	1.6	2.1	10.6	45	0.3	16.3	1.1	1.8	7.9	54	0.4
C15	9.2	1.0	1.2	11.4	34	0.2	12.4	0.9	1.2	9.2	41	0.2
C16	11.4	1.1	1.7	11.0	40	0.1	15.6	1.0	1.8	8.9	48	0.2
C20	12.2	1.9	2.6	10.5	39	0.8	20.4	1.0	1.6	5.4	59	1.3