Editor Comment

As you have seen, the reviewers are generally positive about your study. I also appreciate the vast amount of work that went into this elaborate field experiment that generated very interesting data. Reviewer 1 raised however an important point: you present water fluxes and P concentrations, but failed to calculate the obvious thing: P fluxes. In your answer you refer to another manuscript that you are working on, which specifically deals with P fluxes. For an editor, this is frankly not very satisfying, because how do you guarantee that the present publication is not obsolete, the moment the second will be published? Again, I appreciate the vast amount of work that went into this experiment and I can understand that you want to get the necessary impact out of it. On the other hand I am not a big fan of splitting data to get more publications. So, please reconsider whether it is smart to publish the P fluxes in a seperate paper, or provide good scientific arguments why is is worth published the concentrations and water fluxes separately from the P fluxes. In any case I expect that, independent of your decision, substantial changes will be necessary to your manuscript because at present a lot of the introduction is focussing of P fluxes even though they are not part of the results.

Best regards,

Edzo Veldkamp

Response to the Editor:

Dear Prof. Veldkamp,

Thank you for handling the review process of our paper manuscript. In the revised version we just uploaded we have addressed every comment of both reviewers (see separate uploads). Here we summarize the most substantial changes:

As suggested, we have changed the focus of the paper manuscript and present our results on subsurface storm flow and P concentrations in the light of P turnover an P buffering capacity of soils. As the term “turnover” is commonly used in the context of biotic processes but our study analysis the net effect of both biotic and abiotic processes we decided to use the term *rate of P replenishment* as a more general and neutral term. This term and its meaning is explained in the beginning of the introduction section.

- We have changed the title in line of the suggestion by reviewer #1
- we have rewritten the abstract
- and a substantial part of the introduction
- have added information on P stocks and isotopically exchangeable P

- have added a whole new section in the discussion that reflects the change in focus of the paper.
- We have included a rough estimation of the annual P flux for CON which is the only site we have data for more than one year. We compare it to a study by Sohrt et all. (2019) who estimated annual P fluxes from be-weekly groundwater samples. We also compare our annual P fluxes with input fluxes by dry and wet deposition and rock weathering from this site and from other studies and conclude that P fluxes will be relevant for longer time scales as they are in a similar order than dry and wet depositions or mineral weathering.
- The conclusions are rewritten as suggested to less summarize the results but highlight the new findings.

With this substantial revision we think that we have adequately responded to the feedback and suggestions by both reviewers. They have stated that the content we presented in the first submission was interesting to the scientific community. Including the reviewers comments has certainly improved the paper but we aim to convince that presenting results from a field study like this is relevant to the scientific community. Results on subsurface flow and their interpretation in terms of runoff generation mechanisms and response time and old and new water is still missing in the field and is limiting advance in process understanding. There has been recently an explicit call for more field based experimental work in our field (Burt & McDonnell, 2015). Our study has a profound experimental setup that allowed to measure not only lateral but also vertical subsurface flow – something that has been neglected in previous studies. The experiments also improve on limitations of earlier (smaller-scale, shallower depth) studies that are (more) likely affected by soil heterogeneity and boundary effects.

We also think that our quantitative, hypothesis driven attempt to test whether subsurface flow in various soil depth is subject to dilution/enrichment or is chemostatic across a spectrum of soils with different properties is worth presenting and will not be obsolete in the future. As one of the reviewers mentions, a study like ours has implications on the land surface modelling community that seeks for simple but realistic (and therefore empirically tested) assumptions to model long-term nutrient cycling. To better illustrate the relevance of our outcome and its potential use we now include an example of a rough estimation of the annual P flux for one of our sites where we have the necessary data. We think that the revised paper manuscript and title is now more focused to our aim and offers convincing evidence that this is a relevant contribution to the scientific community.

Best regards,

Response to the Reviewers:

Reviewer 1

Anonymous Referee #1:

Interactive comment on“Phosphorus Transport inSubsurface Flow at Beech Forest Stands: Does Phosphorus Mobilization Keep up withTransport?”by Michael Rinderer et al.Anonymous Referee #1 Received and published: 15 May 202

We thank the reviewer1 for his/her assessment of our paper manuscript and the useful comments to improve the text. To make it more convenient for the second round of reviews we have provide this updated version of our original response to the reviewers. This way we hope to guarantee a better readability.

General remark: The authors present data from sprinkling experiment in three forest sites, performed during two different seasons, where they analyzed water flow and soil solution P con-centrations. The paper is generally well-written and easy to follow, and the results are interesting. However, from my point of view the motivation/objective of the paper is not yet properly addressed with the results. This needs to be addressed before the manuscript can be published. The stated objective of this paper to quantify P losses via subsurface flow (abstract, as well as l. 75 of introduction). This sets the reader up to expect to learn about phosphorus fluxes. More information on subsurface flow P losses would indeed be very interesting, also for the land surface modelling community, which is struggling to incorporate P cycling into C, N models. However, in the results no soil P fluxes [g Pm-2 time-1] are presented, only P concentrations [mg P L-1 water]. I suggest authors to bring the paper in line with the objectives. Firstly, in the introduction by introducing what are typical soil P stocks in forest ecosystems (see e.g. (Achat et al. 2016; Hou etal. 2018)), and further what are orders of magnitude for P flux losses (e.g. in g P m-2yr-1) as determined by earlier studies (see e.g. (Vitousek 2004) and others authors would have to search the literature a bit here). Perhaps also comparing to other Pfluxes in forest ecosystems such as dust deposition, rock weathering, etc. This will set the scene for talking about P fluxes in forest ecosystems. I’m guessing that the losses will be several orders of magnitude lower than the stocks, and it will have to be argued why (if?) they are still important. Secondly, no P flux data is presented in the results. Is it possible to multiply water flowby P conc. to get P flux? Why is this not done?

The focus of the paper has now changed and our aim is now better reflected by a new title.

One of our findings was that chemostatic transport conditions were prevalent in the mineral soil for most of the experiment, even for very high rainfall amounts. This would suggest that annual P flux from forest stands could be approximated by simply knowing the average P concentration and the water balance of the site. To illustrate this, we have now included a new section in the discussion where we estimated the annual P flux for CON. We compare our annual P flux with results from other studies and with P inputs by dry and wet deposition and weathering.

The discussion should be developed further also. How do the results from this study tie into what we already know about P cycling in forests, and P loss pathways? At the moment the discussion mostly explains the results, but it needs to go further to show readers what has been learned. Again, given the setup of the paper, the focus should be on P fluxes. What do the results mean in terms of fluxes? What do we learn about P cycling in forest ecosystems?

We have revised the discussion section to better match the new title. We focus a) on process understanding of subsurface flow (SSF) mechanisms and on b) the P-transport conditions during the experiments. We hope to now better discuss and explain why findings like presented in this paper are useful. We do this (among others) by presenting a rough estimate of an annual P flux that is based on the assumption of chemostatic transport conditions.

Just thinking out loud (authors may choose to followup on this or not): Apart from the nutrient flush in the first 1-2 hours, P concentrations were relatively constant regardless of SSF. On a methodological note, does this imply that we can (roughly) approximate annual P losses via SSF given the water balance of the site and the soil solution P concentration? What would that imply in terms of annual P loss [g P m-2 yr-1] for these sites? How does that compare to the forest stocks and orders of magnitude that can be expected for other loss and input pathways such as dust deposition, weathering and erosion (Chadwick et al. 1999; Hartmann et al. 2014; Tipping et al. 2014; Aciego et al. 2017) ?

We have now included a rough estimation of the annual P flux for CON which is the only site we have data for more than one year. We compare it to a study by Sohrt et all., (2019) at the same site who estimated annual P fluxes with a different approach. We also compare our annual P fluxes with input fluxes by dry and wet deposition and rock weathering from this site.

Specific comments

Title: This is up to the authors, but if they want their article to also reach hydrologists, the title (and abstract?) should be revised. A good portion of the results and discussion as well as the conclusion focus on water flow, which I did not expect from reading the title. E.g. something along the lines o f "Beech forest stands sprinkling experiments: effects on sub-surface flow and phosphorus dynamics"

We change the title in line with the suggestion and rewrote the abstract.

l. 23 Jumping on the "climate change" bandwagon here is unwarranted. There is no discussion of climate change in the article. Also, the data rather show that P conc. is constant and thus only dependent on water balance, right?

The idea of this sentence was to put the paper in a very broad, general context but we agree that climate change is not a main theme in the following paper analysis. Still, precipitation is predicted to change as a consequence of climate change, and by this will have an effect on SSF and thus P-transport. We have deleted this sentence.

l. 29 How much P is in forest soils? How big are these losses?

We have deleted this sentence as we focus on P dynamics

l. 32 remove period after "SSF"

We have rewritten this part

l. 34 remove period after "nutrients"

We have address that

l. 45 The way this sentence is written makes it sound like it was done in this study. I suggest to change tense to "has been" or state "in previous studies"

We have address that

l. 52 add ", USA"

We have address that

l. 54-62 This is too detailed and should be condensed

We have address that

l. 66 "In biopores..." ?

We have corrected this

l. 74 "We performed....to capture potential differences in P fluxes." However, in the research questions the focus is on dynamics of P concentrations. This should be aligned.

We have change it to P dynamics

l. 99 231 g at CON is very similar to 209 g at TUT, especially given heterogeneity inherent to soils. I don't think you can argue that TUT is "less rich in soil P" than CON.

We realize that the way we wrote the sentence is maybe misleading to the reader. We have stated the P-content but not rank it relative to each other.

l. 99 So that the reader can put these numbers into relation (is 209 – 678 g P m2 really a large range in P, justifying calling one P poor and the other P rich?), I again suggest presenting orders of magnitude ranges in soil P stocks in forests (see comment l. 29)

We avoid the terms "rich" and "poor" in the paper and just state the numbers.

l. 102 Add period before "Bulk". I stop correcting spelling / grammar mistakes at this point, but there are more in the remaining text. Please proof read the next version carefully.

We have worked on that.

l. 136 I'm no expert here, but I'm guessing rain water is far from de-ionized. How do you think using deionized water affected the results? Does that need to be discussed?

Collecting 60.000 L of rainfall for the experiment was not an option. So, we were left with using groundwater from the drinking supply system. We argue that using untreated groundwater as sprinkling water would have been unacceptable from an experimental design point of view simply because it is an unnatural source of hydrochemical compounds (including P) to the system. We think that the term "deionized" might make some readers think of purified water like in a lab environment. To show that this was not the case, we had added in L 136 (first submission) that the water had an electrical conductivity of 20 µS/cm. This is comparable to the EC of natural rainfall. The 20 µS/cm is a result of the efficiency of the industrial deionizer and processing 60.000 L of water. In order to avoid irritation, we have removed the term "deionized water".

l. 170-177 Nice setup to let the reader now what to expect, look for and interpret in the results! That's an example of great scientific writing .

We appreciate your positive comment

Table 1: Please also add pH to the table. pH is an important indicator of soil P forms and dynamics and may be important to explain the results, e.g. the difference between TUT, CON and MIT.

We have added pH to Tab 1

Fig. 1 and others: colors are not grayscale print-friendly:

We argue to keep figures in color-scheme as showing all in gray scale is hard to indicate the information included in the plots.

Fig. 2 Very nice overview figure. This makes it a lot easier to understand what was done.

We appreciate your positive comment

Fig 3. also this is a nice figure. I suggest to move spring before summer. I understand that spring experiments were carried out a year later, and that's ok since you have the dates there and it can be noted in the figure caption. But it makes more sense to have the plots in seasonal order for interpreting the plots

We have changing the order of the panels.

Section 3.4 It would have been interesting to measure inorganic and organic P as opposed to only total P.

We see the reviewer's point but Ptot is the data that we have at hand.

Results section 3.5: multiypling conc. by water flow = element flux. Why not present these data in a section 3.5 "Soil P fluxes"

The focus of the paper has changes to better reflect our aim to study SSF and P -dynamics. The fact that we observed chemostatic conditions during most of the duration of the experiment is a useful information for the community. To illustrate our point we have includes a rough estimation of the annual P flux based on SSF data and chemostatic transport conditions; an assumptions motivated by parts of our results.

l. 256-260 (p. 8-9): I'm not surprised that P conc. in the soil solution remains relatively constant. If we consider the very fast turnover time of P in the soil solution of only seconds to minutes (Helfenstein et al. 2018).

We thank the reviewer for the literature suggestions. We have now included turnover (or more general rate of replenishment) and buffering in the introduction and the discussion section (4.2).

l. 300 What about biopores? Is there evidence to suggest that CON and TUT have more earthworms or other large soil fauna

Biopores can also make a contribution to preferential flow. We have included this now in the text. Due to the low pH, there are no or only a few earthworms to be found at MIT and CON. In TUT, earthworms are present. However, due to the clay content, we consider it very likely that cracks originating from shrinking and swelling processes make the largest contribution to preferential flow.

section 4.2 This section could be re-written to make it more focused. At the moment there is a mix of rather trivial findings, such as that P stocks are higher in the forest floor than in the mineral soil, while the interesting things are not discussed in-depth enough. The discussion of P concentration dynamics should be better linked to existing literature, e.g. what is known about turnover time of P in the soil solution and phosphate buffering capacity. Phosphorus-buffering capacity (PBC) is defined as the ability of soil to moderate changes in the concentration of soil solution P (Beckett and White1964; Olsen and Khasawneh 1980; Barrow 1983; Pypers et al. 2006), and would be interesting to bring in here. Soil solution P turnover, a related concept, has been shown to be negatively correlated with P conc. in the soil solution (Helfenstein et al. 2018),which authors might consider discussing as well. (i.e. the more P in the soil solution(forest floor), the slower the turnover time; the less P in the soil solution (mineral soil),the faster the turnover time.

Thank you for your valuable input. We agree that including the mentioned issues have improve the discussion. Section 4.2. has undergone major revisions along the lines suggested.

l. 347 As with the plots, I would take spring before summer.

We have changed this for all figures

l. 364 not exactly true that you have six different experiments. It's one experiment carried out on three sites and at two time points.

We have change this.

l. 371 It's quite well known that soil solution P concentrations are lower with increasing soil depth. I would rather focus on novel findings in the conclusion.

We have rewritten the text.

l. 372 “it was especially strong...” What is it?l. 373 It is obvious that P concentrations are highest in the P-rich site. Again, the conclusion should focus on the novel findings.

We have rewritten the text.

l. 374 “Particulalry high”. Please be concrete. How much higher? Are we talking 1.5x,2x or 10x higher than during the rest of the experiment?

We have added the actual value

l. 375 – 379 This is interesting and in my opinion the main finding of the study. This should be placed more prominently and discussed appropriately.

We have extended this part

l. 380 Conclusion not supported by the data. There was no discussion of climatechange in the article.

The last sentence of the paper has been removed.

l. 436 “DWD, 2010” please provide complete citation reference

Reference is updated

References

Thanks for pointing out to these references. They were valuable for our paper revisions

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Beckett PHT, White RE (1964) Studies on thephosphate potentials of soils. Plant Soil 21:253–282. doi: 10.1007/bf01377744

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Helfenstein J, Jegminat J,McLaren TI, Frossard E (2018) Soil solution phosphorus turnover: derivation, inter-pretation, and insights from a global compilation of isotope exchange kinetic studies. Biogeosciences 15:105–114. doi: 10.5194/bg-15-105-2018

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Reviewer 2

Anonymous Referee #2:

General comments: The manuscript entitled " Phosphorus Transport in Subsurface Flowat Beech Forest Stands: Does Phosphorus Mobilization Keep up with Transport? ",written by Michael Rinderer, Jaane Krüger, Friederike Lang, Heike Puhlmann, and Markus Weiler, presents valuable results that contribute to the understanding of phosphorus transport in and phosphorus losses from the soil. The topic falls into the scope of Biogeosciences. The manuscript comprises results from large sprinkling experiments at three beech forest sites in Germany. The methods are adequate to test the research questions. The results are described in detail and can be used to answer the research question. The text is easily understandable, tables and figures are well-arranged and the conclusions are sound. Hence, I would recommend to consider this manuscript for publication in Biogeosciences after minor revision.

We thank the reviewer2 for his/her positive assessment of our paper manuscript and the useful comments to improve the text. In the following we respond to each comment. Our response is similar to the one we had uploaded earlier.

Specific comments

L 14 The values differ from those in Tab. 1.

We corrected the values to match Table 1.1

L75/76 The time of the two experiments was not well chosen if microbial conditions – like soil moisture, temperature, litter fall – should differ. Rather late autumn/early winter (november; wet, cold, a lot of litter) and summer (july/august; dry, warm, less litter) should have been chosen.

We agree that a stronger contrast in seasonality would have been better to evaluate seasonal effects. However, this was a sub-ordinate part of the study and is therefore not listed as a separate research question in our paper. When choosing our days of sprinkling we were restricted to the vegetation period (i.e., the time when trees had leaves and active photosynthesis) as we were also monitoring tree water uptake and P-transport in trees during the subsequent 4 to 6 weeks after the sprinkling experiment.

However, we will rewrite the text as follows:

"We performed two sprinkling experiments at each site to capture potential differences in P fluxes within the vegetation period (i.e., between summer/fall and spring). …"

And we deleted the part "…and litter fall is not evenly distributed across the year." from the manuscript.

L 227 trise20 of the event water fraction is in Tab. 5 and trise20 of SSF in Tab.4

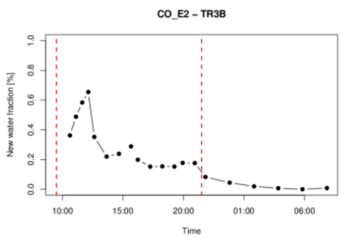
Thanks for pointing this out. We corrected it.

L 233-252 Results of the statistical analyses are not displayed anywhere and the statistical approach is not described in the materials and methods section.

In addition to Figure 4 we have added another figure (Figure 5) that presents the results in form of boxplots that better illustrated what we describe in the text. We also add information in 2.3 Data Analysis (second paragraph).

L 295/296 "A peak of high event water at the beginning of the sprinkling experiments,...“ I could not find this result in the presented data (Fig. 3?).

We agree that this is difficult to see as the total SSF at the beginning of the event is small in general. Here is an example that shows high new water fraction at the onset of response. However, cases like this were few. In general, the high fraction of pre-event water in SSF during the experiment suggests, that preferential flow is a secondary process.



L 302 Tab. 1 (skeleton content) and Fig. 1 (soil bulk density)

We will add/correct the cross-references.

L 315-317 Why is the Ptot concentration from the mineral soil in vertical SSF in MIT lower than from the forest floor?

Probably your question aims at the fact that only in MIT the Ptot concentration in LATERAL SFF from mineral soil is higher than in the LATERAL SFF from the humus layer. A possible explanation was given in Line 317-320 of the first submission version of the paper: “*This is explained by the difference in P-stocks of the forest floor and mineral soil of the three sites. While Ptot stocks in the forest floor at MIT are only 7 g/m^2 it is almost 2 times higher at CON (13 g/m^2) and almost three times higher at TUT(19 g/m^2). On the contrary the Ptot stocks in the mineral soil at MIT (624 g/m^2) are almost 3 times higher than at CON (230 g/m^2) and more than three times higher than at TUT (189 g/m^2)”.*

In addition, lateral SSF from the forest floor at MIT was larger than lateral SSF from the mineral soil while this is not the case for CON and TUT (see Fig. 3b).”

Reviewer #1 suggested to rewrite the section 4.2 to better match the new focus and outcome of the paper. Therefore, the sentence is no longer part of the paper.

L 339/340 This is only true for vertical SSF, isn’t it?

Section 4.2. has undergone major rewriting and the original sentence has been changed.

L345 This is predominately the case for LY1B, isn’t it (Suppl. Tab.1)?

Yes, we will delete the sentence in L345f and fit the information at the end of section 3.4.1.

L 350/351 Which soil properties?

We add e.g. , drainable porosity

L 361 It is unlikely that adsorption explains the difference, since adsorption is very small in the forest floor. How large was the P flow from the 3 sites in g/m2 (in cormparison to the soil P stocks of the 3 sites)? Compare it with values from the literature that you cited in the introduction (L 30 and others).

In the course of rewriting section 4.2. this part has gone but we think that most of the lateral SSF from the forest floor is likely to occur at the contact face between the relatively high permeable forest floor and the lesser permeable mineral soil. So TR1B likely receives water that was flowing at or near the surface of the mineral soil towards the trench. Along this surface of the mineral soil, adsorption can happen. LY1B is installed below the forest floor but on top of the mineral soil and therefore collects water that did not have contact with the mineral soil.

Tab.2 Why was the soil depth of the installations in the subsoil in CON different from MIT?

The depth of installation was adapted to the depth of the soil horizons that differ between sites.

Tab. 4 and 5: You abbreviate both variables with trise20; better add "in SSF“ in Tab. 4 and "in event water fraction“ in Tab. 5.

We have deleted the abbreviation in Table 4 and 5.

Fig. 3b Reorder the labels (TR1B, TR2B, TR3B)according to the labels in Fig. 3a (from forest floor to saprolite).

We have change this.

Fig. 5 The unit of the flow on the x-axis is mm/h, isn‘ it?

Yes, have change that. Thank you!

Technical corrections:

Thanks for pointing these issues out. We have address all technical corrections.

L 29 forest ecosystem -> forest ecosystems

L 66 In biopores-> Biopores

L 171 chemotatic -> chemostatic

L 225 paranthesis is missing

L 287suction caps -> suction cups

L 332 Makoswski et al -> Makowski et al.

L 338 suggest-> suggests

L 345 was -> were

L 357 and 370 expect -> except

Tab. 1 Dominant vegetation and Annual precipitation for TUT: d -> c

Fig. 5 and Fig. 6 Labels -> Label

Manuscript with tracked changes:

~~Phosphorus Transport in~~ Subsurface Flow ~~at~~ and Phosphorus Dynamics in Beech Forest ~~Stands: Does~~Hillslopes during Sprinkling Experiments: How fast is Phosphorus ~~Mobilization Keep up with Transport~~replenished?

Michael Rinderer[1], Jaane Krüger[2], Friederike Lang[2], Heike Puhlmann[3], Markus Weiler[1]

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Correspondence to: Michael Rinderer (michael.rinderer@hydrology.uni-freiburg.de)

Abstract. The Phosphorus (P) ~~is a limiting factor~~concentration of ~~primary productivity in most forest ecosystems but little~~soil solution is of key importance for plant nutrition. During large rainfall events, the P concentration is ~~known about retention~~altered by lateral and vertical subsurface storm flow (SSF) that facilitates P mobilization, redistribution within the soil profile and ~~losses of~~potential P export from ~~forests. Subsurface flow (SSF) is one of the important pathways~~the ecosystem. These processes are not well studied under field conditions. Important factors of the replenishment of P ~~export but few attempts exist to quantify it.~~concentrations in soil solutions are the rate of P replenishment (by biotic and abiotic processes) and P buffering capacity of soils. Lab experiments have shown that replenishment rates can vary between minutes and months. The question remains how P concentrations in lateral and vertical SSF vary under natural field conditions. We present results of large-scale sprinkling experiments ~~with ca.~~simulating 150 mm~~, 2H labelled, total rainfall conducted~~ throughfall at 200 m^2 plots on hillslopes ~~with slopes between 14° and 28°~~ at three beech forests in Germany ~~in summer and spring.~~. We aimed at quantifying lateral and vertical ~~and lateral~~ SSF and associated P ~~transport~~ concentrations in the forest floor, the mineral soil and the saprolite~~. The study~~ during sprinkling events in spring and summer. The sites differed ~~regarding~~mainly in terms of soil depth, skeleton content and soil P stock~~s~~ (between ~~678~~ 189 g/m^2 and ~~209~~ 624 g/m^2~~,~~ in the ~~first~~top 1 m soil depth). Vertical SSF in the mineral soil and in the saprolite was at least two orders of magnitude larger than lateral SSF in the same depth.

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Vertical and lateral SSF consisted mainly of pre-event water that was replaced by sprinkling water (piston flow mechanism). Short spikes of event water at the beginning of the experiment at two of the sites with high skeleton content indicate that preferential flow occurred in parallel to matrix flow. We observed a significant decrease in P concentrations in SSF with increasing soil depth. Higher P concentrations in SSF in the first 1 to 2 h after onset of SSF indicated nutrient flushing, but P concentrations in the mineral soil and saprolite were nearly constant thereafter for most of the experiment despite strong increase in SSF. This suggests that P in the soil solution at all three sites was replenished fast by mineral or organic sources. The observed chemostatic transport conditions suggest that annual P losses at the lateral and vertical boundary of a forest plot can be approximated by knowing the average P concentration and the water fluxes in forest soils. A rough estimation of the annual P loss based on this simplified assumption for one of our sites with longer SSF data, resulted in an annual P loss of 3.16 mg/m^2/a from the forest plot. This P loss is similar to estimates from a previous study at the same site using be-weekly groundwater samples. Our approximated annual P loss in SSF was similar to P input by dry and wet deposition and by mineral weathering, suggesting effective retention of P by adsorption to soil particles in all three forest ecosystems. Higher P concentrations in SSF at the beginning of the experiments indicate nutrient flushing but P concentrations were nearly constant thereafter despite strong increase in SSF. P concentrations did also not change significantly with increasing share of event water in SSF. These chemostatic transport conditions suggests that P mobilization rates were similar to transport rates in both, P-rich and P-poor sites. The observed first flush effect implies that P export by SSF will increase as rainfall events with high transport capacity are predicted to occur more frequent under future climatic conditionsP losses by SSF to become relevant particularly at long time scales.

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1 Introduction

Phosphorus (P) is a major component of plant nutrition and has been reported to be a potential limiting factor of primary productivity in forest ecosystems (Achat et al., 2016; Elser et al., 2000, 2007). In the last decades a decrease of foliar P concentrations and an increase in the Nitrogen to Phosphorus (N:P) ratio has been observed in forests (Braun et al., 2010; Duquesnay et al., 2000; Jonard et al., 2015; Kabeya et al., 2017). One of the largest loss of P from forest ecosystem is occurring via surface or subsurface flow (SSF) (Jardine et al., 1990; Kaiser et al., 2000; Missong et al., 2018b; Sohrt et al., 2018). Changes in elemental composition of SSF provide insight into the processes along the flow paths (e.g., dilution, enrichment, precipitation) and whether P mobilization can keep up with P transport caused by SSF. (Bol et al., 2016; Heathwaite and Dils, 2000; Julich et al., 2017b; Steegen et al., 2001). Soil internal measurements of lateral and vertical SSF and associated elemental concentrations are the prerequisite to characterize ecosystems in terms of their overall capacity to retain available nutrients. (Lang et al., 2017).

Unlike for agricultural land only very few studies exist that quantify the fluxes of P in forest environments, as elemental concentrations are low and measuring vertical and lateral SSF is challenging (Bol et al., 2016; Mayerhofer et al., 2017). In a small number of field studies water was sampled from surface waters i.e., forest streams (Cole and Rapp, 1981; Gottselig et al., 2017; Kunimatsu et al., 2001; Schindler and Nighswander, 1970; Tayor et al., 1971; Zhang et al., 2008) or from groundwater wells in riparian zones near the stream that are easier accessible than SSF (Carlyle and Hill, 2001; Fuchs et al., 2009; Vanek, 1993). But elemental composition of stream water represents an integrated signature of the entire catchment and is therefore not appropriate for a detailed process identification within soil compartments. Stream water is also subject to in-stream retention and mobilization of P and therefore not necessarily representative of transport conditions in the hillslopes (Gregory, 1978; Hill et al., 2010; Mulholland and Hill, 1997; Sohrt et al., 2019; Stelzer et al., 2003).

Therefore, soil solution below forest stands was collected using suction lysimeters (Cole and Rapp, 1981; Compton and Cole, 1998; Kaiser et al., 2000; Qualls et al., 2002) but these samples alone do not allow to calculate P fluxes. A very limited number of studies used a trench to measure fluxes and P concentration in lateral SSF. Timmons et al. (1977) were among the first; they installed a 1.8 m long drainage at the intersection of the A and B horizon in 33 cm soil depth to measure P concentrations and water fluxes in an aspen-birch forest in Minnesota. Sohrt et al. (2018) used 10 m wide trenches to measure and collect lateral subsurface flow from the forest floor, mineral soil and saprolite at three beech forest stands in Germany during natural rainfall events. Jackson et al. (2016) performed an artificial sprinkling experiments on a 200 m^2 plot on a hillslope at the Savannah River Research Site (South Carolina) covered by pine trees and measured water flux and P concentrations in lateral SSF at 1.25 m soil depth. They applied dye- and conservative tracers to identify dominant flow paths and fractions of event and pre-event water involved with solute transport. Makowski et al (2020) were the first to measure vertical SSF use zero-tension lysimeters of a size of 40 x 50 cm installed at 10 cm and 35 cm soil depth at two soil profiles of a beech forest in Saxony, Germany. The mineral soil of the Haplic Cambisol had an average P content between 194 and 221 mg/kg (Makowski et al., 2020). They performed artificial sprinkling experiments on 3 m^2 plots above the lysimeters with a sprinkling intensity of 20 mm/h over 4 hours. They reported significantly higher P concentrations in the sampled vertical SSF in the first 2 hours (P-flushing), followed by a decrease and finally constant P-concentrations for the rest of the experiment. Initial P-concentrations as well as SSF- and P-fluxes where higher in the subsoil than the topsoil. They reported strong differences on water and P fluxes between the two soil profiles which is likely caused by soil heterogeneity and can only be optimized by larger lysimeter and sprinkling plot size.

From field studies as described above and from lab experiments using soil columns we know, that transport of P during high intensity rainfall events does occur mainly along preferential flow paths (Cox et al., 2000; Fuchs et al., 2009; Julich et al., 2017b; Missong et al., 2018a). Preferential flow path allow subsurface flow to bypass the soil matrix that otherwise has been shown to effectively retain P (Compton and Cole, 1998; Ilg et al., 2009; Johnson et al., 2016; Qualls et al., 2002). In biopores (e.g., earthworm borrows, root channels) have been identified as biochemical hotspots that can show significantly higher P concentrations than the soil matrix especially in fine textured soils (Backnäs et al., 2012; Hagedorn and Bundt, 2002). Preferential flow paths can extent below the rooting depth and thus are considered as one important pathway of P losses from forest ecosystems (Julich et al., 2017a; Sohrt et al., 2017).

Here we present a comparative study based on hillslope-scale artificial sprinkling experiments on 200 m^2 plots at three beech (*Fagus sylvatica*) forest sites that differ in terms of their soil depth, skeleton content and SSF flowpaths as well as their soil P stocks. We used an experimental setup that allowed to measure soil depth specific lateral and vertical flow of water and transported hydrochemicals (incl. P). We performed two sprinkling experiments at each site to capture potential differences in P fluxes between summer/fall and spring. The rational is that microbial activity, which is responsible for P mineralization, is strongly dependent on moisture and temperature conditions and litter fall is not evenly distributed across the year (Brinson, 1977; Kirschbaum, 1995). In particular, we address the following research questions:

What are the main runoff generation mechanisms, flow paths and temporal delays in runoff response (lateral versus vertical, shallow versus deep) in three forest stands with different soil properties (i.e., skeleton content and

1 Introduction

Phosphorus (P) is a major component of plant nutrition and has been reported to be a potential limiting factor of primary productivity in forest ecosystems (Achat et al., 2016; Elser et al., 2000, 2007). In the last decades a decrease of foliar P concentrations and an increase in the nitrogen to phosphorus (N:P) ratio has been observed in forests (Braun et al., 2010; Duquesnay et al., 2000; Jonard et al., 2015; Kabeya et al., 2017). From a plant nutrition perspective the replenishment of the P concentration of the soil solution is of key importance for the fertility of a soil (Lambers et al., 2008). The replenishment is a function of the exchange rate between the soil solution and the solid phase including biotic and abiotic processes. Helfenstein et al. (2018) present a worldwide compilation of P turnover for 217 soils that range between 10^{-2} to 10^{6} minutes for which the majority have a turnover of 1 to 100 minutes. The same authors also found a negative relation between the concentration of the soil solution P and the turnover. In our paper we us the term P *replenishment* to describe the replenishment of P by both, biotic and abiotic processes. A high rate of replenishment implies that the soil has many potential binding sites where P can be sorbet or precipitated or the soil has many microorganisms that can immobilize or mobilize P. Thus, the P replenishment is also related to the P buffering capacity which is defined as the ability of a soil to moderate changes in the concentration of soil solution P (Pypers et al., 2006; White and Beckett, 1964).

The P dynamic in subsurface storm flow (SSF) is an indicator for dilution and enrichment processes along the lateral and vertical flow paths in the soil (Bol et al., 2016; Heathwaite and Dils, 2000; Julich et al., 2017a; Steegen et al., 2001). Unlike for agricultural land only very few studies exist that quantify the dynamics of P concentrations in forest ecosystems, as concentrations are low and measuring vertical and lateral SSF is challenging. In a small number of field studies water was sampled from surface waters i.e., forest streams (Cole and Rapp, 1981; Gottselig et al., 2017; Kunimatsu et al., 2001; Schindler and Nighswander, 1970; Tayor et al., 1971; Zhang et al., 2008) or from groundwater wells in riparian zones near the stream that are easier accessible than SSF (Carlyle and Hill, 2001; Fuchs et al., 2009; Vanek, 1993). But elemental composition of stream water represents an integrated signature of the entire catchment and is therefore not appropriate for a detailed process identification within soil compartments. Stream water is also subject to in-stream retention and mobilization of P and therefore not necessarily representative of transport conditions in hillslopes (Gregory, 1978; Hill et al., 2010; Mulholland and Hill, 1997; Sohrt et al., 2019; Stelzer et al., 2003).

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From field studies as described above and from lab experiments using soil columns we know that transport of P during high intensity rainfall events occurs mainly along preferential flow paths (Cox et al., 2000; Fuchs et al., 2009; Julich et al., 2017b; Missong et al., 2018a). Preferential flow paths allow subsurface flow to bypass the soil matrix that otherwise has been shown to effectively retain P (Compton and Cole, 1998; Ilg et al., 2009; Johnson et al., 2016; Qualls et al., 2002). Moreover, biopores (e.g., earthworm borrows, root channels) have been identified as biochemical hotspots that can show significantly higher P concentrations than the soil matrix, especially in fine textured soils (Backnäs et al., 2012; Hagedorn and Bundt, 2002). Preferential flow paths can extent below the rooting depth and are therefore considered an important pathway of P losses from forest ecosystems (Julich et al., 2017a; Sohrt et al., 2017).

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1. What are the main runoff generation mechanisms, flow paths and temporal delays in runoff response (lateral versus vertical, shallow versus deep) in three forest stands with different soil properties (i.e., soil texture, skeleton content, soil depth) during long, moderate intense sprinkling events?
2. What is the dynamic of P concentrations in lateral and vertical SSF, measured at different soil depths during artificial sprinkling events and does it differ seasonally and among sites with different ~~skeleton content~~physical soil properties and soil P availability?

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3. Is P in the soil solution diluted during large sprinkling experiments or ~~can~~is the rate of P ~~mobilization keep up with~~replenishment at all sites and all soil depths high enough to facilitate constant P ~~transport~~concentrations (chemostatic conditions)?

2. Methods

2.1 Study Sites

For this analysis three beech (*Fagus sylvatica*) forest sites in Germany with contrasting soil hydrological properties (i.e., skeleton content and soil depth) and P stocks were selected. Their site characteristics ~~and source of references~~ are summarized in Tab. 1. Mitterfels (MIT) (48° 58' 32" N; 12° 52' 37" E) is located ca. 70 km east of Regensburg in the Bavarian Forest at 1023 m a.s.l. It~~'~~s mean annual precipitation is 1299 mm. The site is characterized by Hyperdystric chromic, folic cambisol with a loamy topsoil (0~~—~~ – 35 cm) and a sandy-loamy subsoil (35 – 130 cm). The stone content in the top- and subsoil is 23 % and 26 % and the P stock~~s~~ in the upper 1 m of the soil profile is 678 g/m^2. The saprolite reaches a total depth of 7 m but ~~weathering~~ is less weathered below 2 m depth. The parent material below the saprolite is paragneiss. Conventwald (CON) (48° 01' 16" N; 7° 57' 56" E) is located 20 km east of Freiburg in the Black Forest at 840 m a.s.l. and has a mean annual ~~total~~ rainfall of 1749 mm. The parent material, main soil type and vegetation is similar to MIT but soils considerably differ in the skeleton content (CON: 87 % topsoil, 67 % subsoil), the depth of the saprolite (CON 17~~-~~ m but less weathered below ~~3m~~3 m) and the~~ir~~ P stock~~s~~ in the upper 1 m of the soil profile (CON: 231 g/m^2). Tuttlingen (TUT) (47° 58' 42" N; 8° 44' 50" E) is located 125 km south of Stuttgart at 835 m a.s.l. and has 900~~-~~ mm annual rainfall. Due to its carbonatic parent material a rendzic Leptosol with a clayey top- and ~~sub-soil~~subsoil has developed ~~that is less rich in soil~~ (P stock~~s~~ ~~(~~209 g/m^2~~) than MIT und CON.~~)). The soil profile has a 20 - 40 cm deep topsoil and a 60 - 80 cm deep subsoil directly overlaying the fractured carbonate parent material ~~(Tab. 2); the~~. The stone content of the top- and ~~sub-soil~~subsoil is 50 % and 67 ~~%.~~%, respectively. The site is also covered by beech ~~(Fagus sylvatica)~~ but the stand is younger than at MIT and CON ~~(TUT: 100 years)~~. Bulk density of CON and TUT is more similar than compared to MIT, but all three soil profiles show considerably variation in the bulk density with depth (Fig. 1~~)~~).

2.2. Experimental Setup and Lab~~-~~ Analysis

At each of the three sites we delineated an experimental plot of 200 m^2 (10 by 20 m) which was separated from its uphill neighboring area by a plastic foil inserted into the soil profile. At the downhill side of the plots, a trench (TR) was dug down into the saprolite ~~until refusal of~~with a hydraulic shovel excavator, and drainage matts and drainage pipes were installed in three (MIT, CON) or two (TUT) depths (Fig. 2) to collect lateral flow. The actual depth of the pipes varied according to the site-specific soil profile (~~Tab. 2~~Tab. 2), but was chosen such that water draining from the forest floor (L, Of, Oh), the mineral soil (A and B horizon) and the saprolite (Cw) could be sampled. Plastic foil was installed across the entire 10 m width of the trench and down to the depth of the three soil compartments so that all water flowing laterally towards the trench was captured in the appropriate drainage pipe. To measure also vertical flow, we installed zero tension lysimeters (LY) for which we used steel piling plates with a dimension of 1.0 by 0.6 m. To install them, a trench was dug at the side of the hillslope and the steel piling plates were pushed from the side into the undisturbed soil profile with heavy duty hydraulic jacks. By this, effects on soil structure by excavation and refill were prevented and the mixing of soil P stocks from different soil horizons was avoided. We installed the LY in similar depth as the TR. At MIT and CON we installed an additional LY right below the A-horizon in 30 to 40 cm soil depth; at TUT the shallow soil depth did not allow installing a LY below 60 cm. In the following, the TR and LY are numbed as TR1B to TR3B and LY1B to LY4B with increasing soil depth (Tab. 2Tab. 2). (“B” indicates the sprinkling plot and allows distinction from another dataset not used in this paper but mentioned in subsequent publications). All trenches were backfilled after installation.

In the upper and lower half of the hillslope, volumetric water content and soil temperature were monitored at two soil profiles in 20, 40, 60, 80 and 120 cm soil depth (no 120 cm measurements at TUT due to shallower soil). We used SMT100 sensors (Truebner GmbH) attached to CR1000 data loggers (Campbell Scientific) and monitored volumetric water content and soil temperature in 5 min time intervals.

At each plot we performed two artificial sprinkling experiments – one in spring at the start of the growing season and one in late summer/early fall during or towards the end of the growing season but well before leaf senescence. The two periods were chosen to reflect potential seasonal differences in soil moisture and P supply. I, i.e., higher soil moisture content after snowmelt but less P mineralization due to colder temperature in spring, versus drier soil moisture conditions and advanced P mineralization towards the end of the growing season with warmer soil temperature in summer/fall. The mean difference of the median volumetric water content over the 7 days preceding the experiment was 8 vol %, 3 vol % and 8 vol % between summer and spring for MIT, CON and TUT, respectively. We sprinkled the 200 m^2 plots with a mean rainfall intensity of 1510 to 20 mm/h over 10 to 12 h. ThisAlthough, this reflects a large rainfall event with a return period of more than 100 years for all sites (DWD, 2010). But the selected(DWD Climate Data Center, 2010), the chosen rainfall intensity allowed all irrigation water to infiltrate into the soil and did not generate surface runoff. 60’000 liters of water were trucked to the site, and run through an industrial deionizer (VE-300 (6x50 Liter), AFT GmbH & Co.KG) to remove all minerals (especially P) that is alsoreduce the mineral content to low levels, typically not found in high concentrations in natural rainfall. The deionizedIn our case the sprinkling water had an electrical conductivity of less thanaround 20 µS/cm. The deionizedsprinkling water was stored in a large water pillow (60.000 L, Sturm Feuerschutz GmbH) between 50 and 100 m above the sprinkling plot. The resulting hydrostatic gradient was sufficient to run the sprinkling without a pump. The 6six radial sprinklers (Xcel-wobbler and pressure regulator manufactured by Senninger) installed at a height of 2 m sprinkled 60 % of the total water onto the 200 m^2 plot and 40 % outside to reduce boundary effects with the otherwise dry surrounding area. 1 kg (first sprinkling experiment) and 2 kg (second experiment) of 99.96 atom % deuteriumoxid was added while filling the water pillow to elevate the natural background deuterium isotopic signature of the sprinkling water by ca. 100 permille. Water samples before and after adding the deuterium and during sprinkling (collected with totalizers on the experimental plot) were collected to measure the background isotopic composition and to check for a constant isotopic label signature over the course of the experiment.

The subsurface flow (SSF) from the TR and LY was routed outside the hillslope via a pipe system to tipping buckets that recorded the flow volume of SSF in 5 min time intervals. The pipe system had been flushed via access tubes the day before the experiments to guarantee function and cleanness. Over the course of the sprinkling experiment (ca. 10 to 12 hours) ~~every 30 minutes~~ the SSF of all TR and LY was sampled every 30 minutes into 100 ml brown glass bottles using automatic samplers (custom made by the Chair of Hydrology, University of Freiburg). The sampling was continued for 12 hours after the end of the sprinkling with a sampling interval of 2 h. The water samples were transported in cooling boxes to the lab directly after the experiment for subsequent hydrochemical and stable isotope analysis. To measure total phosphorus concentrations (Ptot), 50 ml of the sample was digested by adding 0.5 ml 4.5 M sulfuric acid (H_2SO_4) and processed in an autoclave. Ptot was analyzed by the molybdenum blue photometric method based on DIN EN ISO 6878 ~~(DIN, 2004)~~(DIN, 2004) using a Unicam AquaMate photometer (Spectronic Unicam) with a 5 cm-cuvette at 700 nm. We determined the limit of quantitation for Ptot (0.026 mg/l,) and the limit of detection (0.013 mg/l) based on DIN 32645 with a significance bound of 99 % for the limit of quantitation and 77 % for the limit of detection ~~(DIN, 2008).~~(DIN, 2008). The remaining 50 ml of each sample was filtered with a 0.45 µm cellulose filter (PERFECT-FLOW, WICOM) and used for analysis of ^{18}O and ^{2}H stable water isotopes using a Cavity Ring-Down L2130-i Isotopic Liquid Water Analyser (Picarro Inc.). Based on the background isotopic signature and the isotopic signature of the sprinkling water, event- and pre-event water fractions were calculated using a simple two endmember mass balance approach, also called two-component isotope hydrograph separation ~~(Sklash and Farvolden, 1979)~~(Sklash and Farvolden, 1979).

2.3 Data Analysis

TR and LY volumetric measurements were scaled to 1 m^2 ~~of~~ plot area and expressed as mm/h, which allows direct comparison with the incoming sprinkling water. We determined the time lag between the start of the sprinkling experiment and the time when 20 % of the total rise in SSF had been reached or exceeded (called t_{rise20} in SSF). In a similar way we determined the time lag between the start of the sprinkling event and the time when the event water fraction had reached 20 % (called t_{rise20} in event water fraction). The threshold of 20 % has been chosen as clear indication of first response in the change of SSF and event water fraction. If t_{rise20} in event water fraction is longer than t_{rise20} in SSF this indicates that the flow celerity is faster than flow velocity. If t_{rise20} in event water fraction is shorter than the t_{rise20} in SSF, this indicates preferential flow as velocity is faster than celerity.

We plotted P concentrations of vertical and lateral SSF as a function of time but also as boxplots to allow a better comparison between the experiments. To investigate if ~~P mobilization was able to keep up with P transport~~the P concentrations of each flow component was higher during the first 2 h after onset of flow than during the remaining time of the experiments, we plotted our data separately for the two periods of each event. As some of the flow components had only few samples in the flushing period a statistical test of significance was not meaningful. This was also the reason why we did not further differentiate between flushing period and remaining part for the following analysis.

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We plotted P- concentrations as a function of SSF in log-log space~~. If the statement before was correct~~ to investigate whether P mobilization was able to keep up with P transport. In this case, we would expect near chemostactic conditions, i.e., P-concentrations would not vary much with increasing or decreasing SSF. In this case data points were expected to plot parallel to the x-axis ~~in log-log space.~~. In case of simple dilution, P concentrations would decrease proportional with increasing SSF and the data points would be aligned on a 1:1 line ~~in log-log space.~~. As a further measure we calculated the ratio between the range in observed SSF values and the respective range in observed P concentration. Under predominantly chemostatic conditions, the range in SSF would be much larger than the range in P- concentrations.

In addition, we investigated the relation between Ptot concentrations and event water fractions and checked whether the slope of a linear regression based on the datapoints was significantly different from zero. We also tested if this regression slope was significantly different from a slope describing dilution due to proportional mixing of pre-event water with our ~~deionized~~ sprinkling water using a Mann-Whitney test. The slope describing simple dilution was determined by a regression with the best linear fit through the data points and the additional constrain of Ptot = 0 mg/l for event water fraction = 1. ~~All data analyses were performed in R 3.4.2 (R Developer Team).~~

3. Results

3.1 Lateral versus vertical SSF

In general, vertical SSF (measured by LY) dominated total water fl~~ux~~ow during all sprinkling experiments. Depending on site and soil depth, between 89 % and 99 % of total SSF ~~was~~percolated vertically ~~percolating~~ to deeper depths and only < 1 to 11 % of ~~the sprinkling water~~total SSF was flowing laterally towards the trench (Fig. 3). At all study sites LY1B (below the forest floor) yielded steady flow with a mean rate of 10 to 15 mm/h, which is identical to the sprinkling rate. This confirms that the LY, even if positioned at the boundary of the hillslope, were experiencing rainfall intensities that are representative for the rest of the hillslope. The LY at deeper soil depth showed a slower increase in SSF than LY1B ~~below the forest floor~~, but also reached a mean flow rate of 10 to 15 mm/h towards the end of the sprinkling experiment (except LY3B and LY4B at MIT in spring and summer ~~and spring~~ and LY3B at TUT in summer) (Fig. 3). Lateral SSF (measured by TR) was at least two orders of magnitude lower than vertical SSF but increased constantly towards the end of the sprinkling experiments. An exception was TR2B at TUT which reached a plateau (ca. 1 mm/h) during the experiment in spring ~~experiment~~, most probable due to wetter antecedent conditions ~~(see discussion section).~~. Maximum SSF from TR2B at CON and TUT was 0.~~51~~72 and ~~0.70~~1.09 mm/h during the experiment in spring and 0.51 and 0.70 mm/h in summer ~~and 0.72 and 1.09 mm/h in spring~~. TR3B at MIT yielded neither vertical nor lateral SSF in any of the two sprinkling experiments. This is likely attributed to lower skeleton content and higher storage capacity of the soil in MIT.

3.2 Event and ~~p~~Pre-event ~~water fractions~~Water Fractions

Vertical SSF in the topsoil, subsoil and saprolite (i.e., all LY except LY1B) at all sites and during all experiments was ~~predominantly~~dominated by pre-event water (Fig. 3, Tab. 3). In contrast, the mean pre-event water fraction from the forest floor (i.e., LY1B) during the sprinkling events was low (i.e., MIT: ~~15~~12 % and ~~12~~15 %; CON: 10 % and 16 % and ~~10 % and~~ TUT: 4 % and 4 % in spring and summer ~~and spring~~, respectively). The mean pre-event water fractions of vertical SSF increased with depth at all sites and all events and already in 35 to 40 cm soil depth (LY2B) the mean pre-event water fractions during the events in spring and summer ~~and spring~~ were 83 % and 88 % ~~and 83 %~~ at MIT, 60 % and 58 % ~~and 60 %~~ at CON, and 64 % and 83 % ~~and 64 %~~ at TUT. The mean pre-event water fraction in the vertical SSF further continued to increase with soil depth (see LY3B and LY4B in Tab. 3).

For the lateral SSF a similar increase of the pre-event water fraction with soil depth was observed. However, the mean pre-event water fractions of lateral SSF in the subsoil (i.e., TR2B) were typically smaller than the pre-event water fractions in vertical SSF at similar depth (i.e., LY3B). The mean pre-event water fractions of TR2B during the events in spring and summer ~~and spring~~ were 44 % and 33 % ~~and 44 %~~ at MIT, 63 % and 55 % ~~and 63 %~~ at CON and 70 % and 47 % ~~and 70 %~~ at TUT. The mean pre-event water fractions during the events in spring and summer ~~and spring~~ of LY3B were 95 % and 93 % ~~and 95 %~~ at MIT, 63 % and 83 % ~~and 63 %~~ at CON and 95 % and 70 % ~~and 95 %~~ at TUT. Mean pre-event water fractions of the lateral and vertical SSF in the saprolite (i.e., TR3B and LY4B) were similar (i.e., 78 % and 74 % ~~and 78 %~~ for TR3B and 86 % and 78 % ~~and 86 %~~ for LY4B at CON during the experiment in spring and summer ~~and spring~~.

3.3 Differences in SSF and ~~response time~~Response Time between ~~summer~~Spring and ~~spring~~Summer

In general, SSF differed less between the sprinkling experiments in ~~summer~~spring and in ~~spring differed less~~ summer than between the three sites. This was particularly true for the peak flows in SSF ~~at all sites~~. An exception was TUT where peak flows for LY2B and LY3B were higher in spring compared to summer which is likely due to pronounced differences in antecedent wetness between spring and summer ~~and spring~~ at TUT ~~(see details above).~~ .

SSF between spring and summer ~~and spring~~ differed mainly in respect to the response time of the individual TR and LY. This was most pronounced at MIT and least at CON (Fig. 3, Tab. 4). At MIT the t_{rise20} in SSF was on average 1.9 times longer in summer than in spring (except TR2B: 0.7 times shorter) (Tab. 4). At CON it was the opposite i.e., the t_{rise20} in SSF was on average 0.7 times shorter in summer than in spring; except for SSF in the saprolite (i.e., LY3B, LY4B, TR3B) for which t_{rise20} in SSF was on average ~~was~~ 1.3 times longer than in spring (Tab. 4). At TUT t_{rise20} in SSF in the top and subsoil was 1.8 times longer in summer than in spring but t_{rise20} in SSF in the forest floor (LY1B and TR1B) was 0.3 times shorter. At CON and TUT the t_{rise20} in SSF was not always increasing systematically with soil depth (e.g., LY3B responded earlier than LY2B at CON in spring and TUT in summer; TR3B responded earlier than TR2B at CON in spring and summer ~~and spring.)~~. At MIT in summer LY4B did not respond until 1.5 h after the end of the sprinkling experiment and TR3B did not respond at all in spring and summer ~~and spring~~.

The t_{rise20} of the event water fraction (~~Tab. 4~~Tab. 5), was typically longer than the t_{rise20} of SSF (~~Tab. 5)~~,Tab. 4). Only for TR2B and TR3B at CON in spring and summer~~ and spring~~ and TR2B at TUT in summer t_{rise20} of event water fraction was shorter than t_{rise20} of SSF. These differences in t_{rise20} of event water fraction and t_{ries20} of SSF for these TR was in the range of 3 hours to almost 4 hours for CON and 1 h for TUT. At MIT, SSF from LY3B and LY4B did not reach 20% event water fraction during the experiments in summer and spring. The same was true for LY3B at TUT in spring.

3.4 Dynamics of P ~~c~~Concentrations

Median Ptot concentrations of vertical SSF (LY) ~~were up to one order of magnitude higher~~ during the first flush (i.e., the first 1 to 2 hours after the first response of each flow component~~-~~) were up to 6 time higher than during the remaining time of the sprinkling experiment (Fig. 5); some of the Ptot concentrations of the very first water samples were even up to one order of magnitude higher (Fig. 4a). This was particularly true for the vertical flow from the forest floor and the topsoil (LY1B and LY2B) but also apparent, albeit less ~~than one order of magnitude~~distinct, for the subsoil and the saprolite (LY3B, LY4B). The Ptot concentrations of vertical flow from the forest floor was significantly higher than in the topsoil (in 30 to 40 cm soil depth), except for CON and TUT in summer (Fig. 4a). Similarly, the Ptot concentrations of vertical flow in the subsoil and saprolite was significantly lower than in the topsoil except for MIT in spring and summer and ~~spring and~~ at TUT in summer. All Ptot concentrations were above the limit of quantitation (i.e., 0.009 mg/l).

The Ptot concentrations in the same vertical flow component were not significantly different between spring and summer~~ and spring~~, except for LY1B and LY3B at MIT; LY1B at CON; LY1B, LY2B and LY3B at TUT. However, the Ptot concentrations of the same flow component (e.g., LY1B) at ~~the P-rich site~~ MIT was generally significantly higher than at ~~the P-poorer sites~~ CON and TUT, except for LY2B in spring. In a similar vein, Ptot concentration at CON were significantly higher than at TUT except, LY3B in summer.

Ptot concentrations in the lateral flows (TR) also showed a sharp decline during the first 1 to 2 hours after the onset of flow of each component, except for TR1B and TR2B at MIT in spring and TR2B at CON in spring (Fig. 4b). Ptot concentrations of TR2B at CON in summer showed a steady decline and for TR3B the decline occurred with more delay (i.e., 5 h after first response) compared to the other experiments. At CON and TUT the Ptot concentrations in the lateral flow of the forest floor (TR1B) were significantly higher than in the subsoil (TR2B). Contrarily at MIT the Ptot concentrations in lateral flow of the forest floor (TR1B) were significantly lower than in the subsoil (TR2B), both in spring and summer ~~and spring~~ (Fig. 4b). The difference in Ptot concentrations in the same flow component in spring and summer~~ and spring~~ was not significant, except for TR1B at TUT.

3.4.1 P ~~c~~Concentration as a ~~f~~Function of instantaneous ~~f~~Flow

The range in SSF was typically several factors if not orders of magnitude larger than the range in Ptot concentrations during the sprinkling experiments except for those flow components that yielded little SSF in general (e.g., TR1B at CON during spring and summer ~~and spring~~, LY3B and TR2B at MIT during summer) (Fig. 6). Data points were in general aligned rather parallel to the x-axis in the log-log plots of Ptot concentration versus SSF and not along the 1:1 line (Fig. 6). This suggests that transport in most subsurface flow components was rather chemostatic than diluted. However, we observed weak anti-clockwise hysteresis effects i.e., median Ptot concentrations were higher on the rising limb than on the falling limb of the SSF hydrographs, except for TR1B at MIT in summer, LY4B at CON in spring and LY2B at TUT in summer. Most differences in Ptot concentrations during rising and falling limbs were, however, small (e.g., 0.007 mg/l, 0.028 mg/l and 0.081 mg/l for the 25%-, 50%- and 75% quantile of all differences) indicating that the hysteresis effect was small and only for LY1B this difference was consistently significant across all sites (Suppl. Tab. 1).

3.4.2 P ~~c~~Concentration as a ~~f~~Function of ~~event water fraction~~Event Water Fraction

Ptot concentrations also did not change significantly with increasing event water fraction (Fig. 7). Only in the forest floor (LY1B and TR1B), the slopes of the linear regression lines fitted to the Ptot concentrations as a function of event water fractions were significantly different from zero, except LY1B at CON in summer and TR1B at MIT in spring (Tab. 6). The transport conditions in the lateral and vertical SSF in the forest floor were therefore predominantly non-chemostatic. In the mineral soil and saprolite lateral and vertical SSF was dominantly chemostatic (except LY2B and LY4B at MIT in spring, LY3B at CON in summer, LY2B at CON in spring, LY2B and TR2B at TUT in summer and TR2B at TUT in spring) (Tab. 6). Most of these regression slopes were however close to zero.

For the regression slopes indicating non-chemostatic behavior, we tested if they were not significantly different form a slope describing proportional mixing. In general this was not the case, except for some flow components from the forest floor (i.e., LY1B at MIT and TUT in summer, TR1B at CON in spring; see italic values in Tab. 6) and from the mineral soil (i.e, TR2B at MIT in summer, TR2B at TUT in summer and spring and LY3B at CON in summer). At least for the latter three the regression slope was however close to zero~~.~~ (Tab. 6). As a rough generalization one could summarize: The regression slopes of most lateral and vertical SSF in the mineral soil and saprolite tended to be small or close to zero (the majority was chemostatic) whereas regression slopes of flow components from the forest floor were typically significantly different from zero (some indicating proportional mixing).

4. Discussion

4.1 ~~What are the main~~Main SSF ~~paths~~Flow Paths during long, moderate intense ~~sprinkling events?~~Sprinkling Events

In general, vertical SSF dominated total ~~flow~~SSF during all ~~of our~~ sprinkling experiments and lateral SSF was at least two orders of magnitude lower than vertical SSF (Fig. 3)~~. This finding implies that previous studies at trenched hillslopes at sites with well drained soils and moderately permeable bedrock likely missed out to measure and sample an important loss term of the water and nutrient balance (Jackson et al., 2016; Sohrt et al., 2018; Timmons et al., 1977). This is partly due to different research foci of these studies but mainly attributed to the technical difficulty to measure and sample vertical SSF. The use of our large zero tension lysimeters is a successful way to capture vertical flow. They yield more representative results than~~

~~traditional small size lysimeters of a few cm^2 or suction caps that are more likely to be affected by soil heterogeneity. The steel piling plates that we pressed into the undisturbed soil profile from the side of the hillslope also allow to preserve the natural soil profile with its soil texture and structure and horizon-specific P stocks. However, their installation comes with high effort.~~). This finding implies that previous studies at trenched hillslopes at sites with well drained soils and moderately permeable bedrock missed out to quantify the important loss term of the water balance (e.g., Jackson et al., 2016; Sohrt et al., 2018; Timmons et al., 1977). This is partly due to different research foci of these studies but mainly attributed to the technical difficulty to measure and sample vertical SSF. The use of our large zero tension lysimeters is a successful way to capture vertical flow in heterogeneous soils. They yield more representative results than traditional small size lysimeters of a few cm^2 or suction cups that are more likely to be affected by soil heterogeneity. The steel piling plates that we pressed into the undisturbed soil profile from the side of the hillslope also allow to preserve the natural soil profile above with its soil texture and structure and horizon-specific P stocks. However, their installation comes with high technical and man-power effort.

Vertical and lateral SSF in the mineral soil and saprolite at all sites and all experiments was predominantly pre-event water (Fig. 3, Tab. 3). This is generally in agreement with Jackson et al. ~~(2016)~~(2016) who performed a tracer experiment at the Savannah River Site (South Carolina~~) with~~, USA) in a loamy sand topsoil overlaying a sandy clay-loam subsoil. However, their maximum pre-event water fraction was 50 % while it was typically higher in our study (mean pre-event water fraction in vertical and lateral SSF of the mineral soil was 83 % and 63 %, see also Tab. 3). Our findings suggest that SSF runoff generation was dominated by ~~piston flow~~initiation of water already stored in the soil, i.e., incoming event water is pushing pre-event water down into the soil profile initiating SSF. ~~A peak of high event water at the beginning of the sprinkling experiments, a~~A non-sequential onset of SSF with soil depth and shorter t_{rise20} of event water fraction than t_{rise20} of SSF in some lateral flow components at CON and TUT suggest, however, that preferential flow occurred in parallel to matrix flow at CON and TUT.

Occurrence of preferential flow is important as it allows P bypassing the soil matrix in its soluble and colloidal form and is therefore considered to be a very prominent pathway of P loss from the ecosystem ~~(Jardine et al., 1990; Kaiser et al., 2000; Missong et al., 2018b).~~(Jardine et al., 1990; Kaiser et al., 2000; Missong et al., 2018b). The fact that we observed indications of preferential flow predominantly at CON and TUT but not at MIT and mainly in lateral flow and less in vertical flow may be explained by differences in soil properties, especially skeleton content (Tab. 1) and soil bulk density (Fig. 1) of the three sites ~~(Fig. 1).~~. Swelling and shrinking due to the higher clay content and more biopores due to higher earthworm abundance (due to higher soil pH) at TUT might be another mechanism leading to preferential flowpaths. At MIT the lateral and vertical flow components in the saprolite did not respond or responded with strong ~~time~~ delay to the sprinkling experiments (Fig. 3, Tab. 4) which suggests very efficient storage of the sprinkling water in the soil. Therefore, at MIT characterized by relatively low skeleton content but high soil storage capacity, the shallow flowpaths were most important and the deeper flowpaths did not yield much or any flow. At CON it was the opposite. The higher skeleton content allowed water to reach deeper soil depth and therefore deeper flowpaths yielded more flow than shallower ones. At TUT the clay-rich topsoil led to more lateral flow at shallow depth than CON but as the total soil depth was much smaller than at the two other sites, the storage capacity was less and therefore also the mineral soil yielded significant amounts of flow.

4.2 P concentration dynamics in vertical and lateral SSF

Median Ptot concentrations in vertical SSF from the forest floor were significantly higher than those from the mineral soil and saprolite (Fig. 4). This suggests that P losses from the forest floor were efficiently retained by adsorption in the mineral soil. This was the case for all three sites regardless of natural P-soil stocks. The rate at which adsorption occurred appeared to be faster than the flow rate. The Ptot concentrations in vertical and lateral SSF declined with increasing soil depth, except for MIT were the median Ptot concentration in lateral SSF of the mineral soil was higher than from the forest floor during both sprinkling experiments. This is explained by the difference in P-stocks of the forest floor and mineral soil of the three sites. While Ptot stocks in the forest floor at MIT are only 7 g/m^2 it is almost 2 times higher at CON (13 g/m^2) and almost three times higher at TUT(19 g/m^2) (see Tab. 1). On the contrary the Ptot stocks in the mineral soil at MIT (624 g/m^2) are almost 3 times higher than at CON (230 g/m^2) and more than three times higher than at TUT (189 g/m^2). The only other study on P concentrations in vertical SSF, did not find a significant difference in Ptot concentrations of vertical SSF at 10 cm and 35 cm soil depth at their beech forest site in Saxony, Germany with Haplic Cambisol (Makowski et al., 2020).

While in our study P concentrations in SSF decreased with soil depth at all three sites, the Ptot concentrations of the same flow component at different sites was typically highest at the P-rich site (MIT) and lowest at the P-poorest site (TUT), except lateral flows from the forest floor (TR1B) at MIT. This suggests that similar P leaching and retention mechanisms are occurring in soils at P-rich and P-poor sites but the actual P concentrations in SSF differ between forest ecosystems with different P-availability.

Ptot concentrations in vertical and lateral SSF at all soil depth were typically significantly higher during the first 1 to 2 hours after the first response of each flow component than during the remaining time of the sprinkling experiment (Fig. 4). This nutrient flushing effect (Hornberger et al., 1994) has been describe also in other studies as a prominent feature of lateral export of nutrients; mainly N (DON) and C (DOC) (Qualls, G. Haines, 1991; van Verseveld et al., 2008; Weiler and Mcdonnell, 2006) but also P (DOP) (Burns et al., 1998; Missong et al., 2018a; Qualls et al., 2002; Sohrt et al., 2018). Makowski et al (2020), who measured P concentrations in vertical SSF, also reported nutrient-flushing in the first 2 hours of their sprinkling experiments.

In our study Ptot concentrations after the nutrient flushing were, relatively constant regardless of further increasing SSF. The change in Ptot concentrations were several factors smaller than the change in SSF (Fig. 5) and the regression slope of Ptot concentration versus SSF was not significantly different from zero for most flow components in the mineral soil (Tab. 6). This suggest that P transport in SSF in the mineral soil was chemostatic, which further suggests that P mobilization could keep up with P transport. This was different in the forest floor as Ptot concentrations in SSF continued to decline at a slow rate after the flushing phase (Fig. 4). However the slope of a regression line of Ptot concentration versus event water fraction of most flow components in the forest floor was significantly different from simple dilution that would be the case if a given amount of P is leached and diluted by an increasing amount of event water in SSF (Fig. 6, Tab. 6). It is therefore likely that dilution happed at a lower rate than assumed by proportional mixing, i.e., the rate at which P was supplied by biogeochemical processes was equal or slightly lower than transport capacity. This would also fit our other observation that the median Ptot concentrations was higher on the rising than on the falling limb of all flow components (Fig. 5, Suppl. Tab. 1).

4.3 Seasonal differences in SSF and P concentrations

The differences in SSF between the sprinkling experiment in summer and spring were smaller than the difference in SSF betweenThe differences in SSF between the sprinkling experiment in spring and summer were smaller than the difference in SSF among the three sites and mainly related to SSF response timing (Fig. 3, Tab. 4). The reason is likely due to relatively small differences in soil moisture antecedent conditions between the two sprinkling experiments in summer and spring (except TUT). This was particularly true at CON where the high skeleton content allowed the soils to drain quickly to field capacity. Soil properties, such as drainable porosity, likely also explain, why seasonal differences in SSF response timing were more pronounced at MIT than CON (Tab. 4, Tab. 5). At TUT the difference in SSF dynamics between the experiment in summer and spring is likely more related to differences in antecedent wetness conditions. The 7-day-average of the median volumetric water content of the soil profile at TUT during spring was 23 vol % compared to 15 vol % at TUT during summer. Still, the dominance of pre-event water fractions during both events at TUT suggest that not fast preferential flow but piston flow was the dominant process during both experiments at TUT.

P concentrations in the same SSF flow component were typically not significantly different between summer and spring, expect for vertical flow from the forest floor at all sites. This might be an effect of the different biogeochemical processes and degree of decomposition of the litter material between summer and spring, as we had hypothesized. The reason why we did not measure a significant difference in Ptot concentration in lateral flow from the forest floor (TR1B) could be that lateral flow has typically a longer flow distance to the trench along which adsorption can occur. While the mean lateral distance to the trench is 10 m the total vertical soil depth is 3 m.

4.2 P Concentration Dynamics in vertical and lateral SSF

Ptot concentrations in vertical and lateral SSF, particularly in the forest floor and to a lesser degree in the mineral soil, were typically significantly higher during the first 1 to 2 hours after the first flow response of each component than during the remaining time of the sprinkling experiment (Fig. 4, Fig. 5). This so-called nutrient flushing effect (Hornberger et al., 1994) has been described also in other studies as a prominent feature of lateral export of nutrients; mainly for N (DON) and C (DOC) (Qualls and Haines, 1991; van Verseveld et al., 2008; Weiler and Mcdonnell, 2006) but also for P (DOP) (Burns et al., 1998; Missong et al., 2018a; Qualls et al., 2002; Sohrt et al., 2018). Makowski et al. (2020), who measured P concentrations in vertical SSF, also reported nutrient-flushing in the first 2 hours of their sprinkling experiments.

In our study Ptot concentrations in SSF from the forest floor continued to decline after the flushing phase or were relatively constant (Fig. 4). The slope of a regression line of Ptot concentration versus event water fraction of most flow components in

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the forest floor was significantly different from simple dilution that would be the case if a given amount of P was leached and diluted by an increasing amount of event water in SSF (Fig. 7, Tab. 6). A likely better explanation could be that the rate of P replenishment in the P-rich forest floor was not high enough to facilitate the high P concentrations measured at the beginning of the event. This is in line with Helfenstein et al (2018) who found a negative relation between P concentration in the soil solution and the turnover rate based on isotopic exchange kinetic experiments on 217 soil samples collected worldwide. The P concentrations from the forest floor (except MIT during summer) were more constant towards the end of the experiment, when the P concentrations from the forest floor where low in general.

In the mineral soil the flushing effect was less distinct. The P concentrations in SSF were generally lower compared to the forest floor. The change in Ptot concentrations in the mineral soil were several times smaller than the change in SSF (Fig. 6) and the regression slope of Ptot concentration versus SSF was not significantly different from zero for most flow components in the mineral soil (Tab. 6). This suggests that P transport in SSF in the mineral soil was chemostatic. It also suggests that the rate of P replenishment in SSF of the mineral soil was high during the experiments. This was even true towards the end of the sprinkling experiments when the fraction of new water increased. For this new sprinkling water, it is clear that the contact time was short (i.e., in the order of minutes to hours) and still the P concentration was not significantly lower than for water samples with a higher pre-event water fraction. The relatively constant P concentrations in the mineral soil also suggest, that P leached from the forest floor was efficiently buffered in the mineral soil and that this buffering effect is also apparent in soils with preferential flow paths (CON and TUT) that we considered as a potential bypass of the soil matrix. We could not see significant differences between the experiments in spring and summer at the three sites. An exception were P concentrations of vertical flow from the forest floor at all sites. Our findings are therefore in line with Kaiser et al. (2003) and Sohrt et al. (2019) who also reported higher P concentrations of leachate from the forest floor during the vegetation period. Sohrt et al (2019) also reported an increase in P concentrations in throughfall towards the end of the vegetation period but the average P-concentration was 0.15 mg/l and therefore more than 4 times smaller compared to the enrichment caused by leaching form the forest floor (average 0.7 mg/l).

4.3 Estimates of annual P Fluxes

Our finding that P transport in the SSF after the first flush was chemostatic would suggest that annual P losses from forest stands could be roughly estimated by knowing the amount of annual SSF and an average P concentration. This would be particularly relevant for long-term soil evolution modeling (Chadwick et al., 1999; Vitousek, 2004). While we lack the data to fully test this hypothesis, we performed a rough estimation of the annual P fluxes in SSF at CON, which is the only site where we have SSF data for more than 1 year. We chose the period between 10.05.2018 to 09.05.2019 because the water balance is not affected by the sprinkling experiments. The sum of SSF and the calculated Ptot flux for this period is shown in Tab. 7. The mean P concentration used to calculate the annual P flux is based on the P concentrations measured after the first flush during the experiment at CON in spring 2018 (i.e., 07.05.2018). This is the sprinkling experiment most recent to the one-year period of interest. Our estimation of annual Ptot fluxes (sum of vertical and lateral) yielded 45 $mg/m^2/a$ from the forest floor and

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14 mg/m^2/a from the topsoil, 6 mg/m^2/a from the subsoil and 2 mg/m^2/a from the saprolite. Considering only the outside boundaries of the hillslope (LY4B, TR1B, TR2B, TR3B) we estimated a total P-loss of 3.16 mg/m^2/a. While we expect these values to be different for every year, the order of magnitude can be compared to values presented by Sohrt et al. (2019) who have sampled P concentrations in lateral SSF at CON on a bi-weekly interval between 01.03.2015 and 25.02.2016. Their mean P concentration from the organic layer (only lateral flow) was 0.57 mg/l, the annual water flux from the organic layer was 0.002 mm/a and the calculated annual P flux was 0.001 mg/m^2/a. For the mineral soil they reported a mean P concentration of 0.043 mg/l, a total annual water flux of 446 mm/a and a resulting P flux of 20 mg/m^2/a. While the annual flux from the mineral soil matches well between Sohrt et al. (2019) and our study, the annual P flux from the forest floor differs by several orders of magnitude. This is likely due to differences in the estimated water fluxes (Tab. 7). Sohrt et al. (2019) estimated the annual water flux by End Member Mixing Analysis (EMMA) based on samples from the lateral flow components (TR1B) only. Our annual water flux from the forest floor comprises both, the lateral (TR1B) and the vertical (LY1B) flow component of which the latter has been shown to be orders of magnitude lager then the lateral flow (see Fig. 3). For that reason it is likely better to compare the estimated P fluxes from the outside boundaries of the hillslope in our study (3.16 mg/m^2/a) to the P fluxes in the groundwater (2.5 mg/m^2/a) in the study of Sohrt et al. (2019). These two P fluxes represent the P loss from the entire hillslope. These two fluxes match well.

Our annual P fluxes are also in line with the range of fluxes published in a number of other studies or reviews. Bol et al. (2016) estimated that potential total P loss through leaching into subsoils and export by forest streams was less than 50 mg P/m^2/a. But the range of published data varies more than one order of magnitude (e.g., 4 mg Ptot/m^2/a in Benning et al. (2012); 32 mg/m^2/a in Julich personal communication, cit. in Bol et al. (2016)). Other studies reported only annual fluxes of Dissolved Organic Phosphorus (DOP). While these numbers are difficult to be directly compared to our Ptot data, the published values are in a similar order of magnitude (e.g.: 15 to 62 mg DOP/m^2/a in leachate from the organic layer and 1.7 to 38 mg DOP/m^2/a from the mineral soil (Kaiser et al., 2000, 2003; Qualls et al., 2000). In another meta-analysis Sohrt et al. (2017) presented annual P fluxes that are higher: (e.g., mean: 226 mg/m^2/a (sd: 389 mg/m^2/a) and mean: 119 mg/m^2/a (sd: 279 mg/m^2/a) for water samples from the organic layers and the mineral soils). A possible reason for the higher values compared to our results could be that we used mean P concentration measured after the first flush which results in rather conservative estimates. Julich (personal communication cit. in Boll et al. (2016)) reported that up to 40 % of the annual P flux might occur during single events, which would suggest that the fluxes during the first flush have an important share on the annual flux and cannot be neglected.

Despite the considerable range of published annual P fluxes in SSF from different forest environments it becomes clear that annual P fluxes are orders of magnitude lower than soil P stocks (Achat et al., 2016; Hou et al., 2018). In the case of CON soil P stocks are estimated to be 13 g/m^2 and 230 g/m^2 for the forest floor and the mineral soil, respectively (Lang et al., 2017). Therefore, export of P from forest ecosystems by SSF becomes only relevant on the time-scale of millennia (Bol et al., 2016; Vitousek, 2004). The annual P losses are also well compensated by atmospheric deposition and mineral weathering (Aciego et al., 2017; Hartmann et al., 2014; Tipping et al., 2014). For instance, at CON the annual P flux by dry deposition, canopy

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leaching, bulk throughfall and mineral weathering are 10, 43, 60 and 76 mg/m^2/a (Sohrt et al., 2017; Uhlig and von Blanckenburg, 2019) and therefore in a similar order as the estimated annual P fluxes by SSF in our study. It is also important to note that the internal ecosystem P fluxes due to forest litter fall (100 to 500 mg/m^2/a, Sohrt et al. (2017)) are up to one order of magnitude larger than the annual P fluxes in SSF. This is an indication of an efficient nutrient recycling in forest ecosystems (Lang et al., 2017).

5. Conclusions

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We present results of six sprinkling experiments conducted at 200 m^2 hillslopes at three beech forest sites in Germany that differ in their soil depth, skeleton content and soil P stocks to quantify vertical and lateral SSF and associated P transport. Vertical SSF in the mineral soil and saprolite was at least two orders of magnitude larger than lateral SSF and consisted mainly of pre-event water that was likely replaced by sprinkling water (piston flow mechanism). Short spikes of event water at the beginning of the experiment at CON and TUT however indicate that preferential flow occurred in parallel at sites with a higher skeleton content. No or very delayed SSF from the saprolite at MIT also showed the importance of soil storage capacity in terms of retaining water and nutrients; differences between seasons were however minor at all sites, expect for vertical flow from the forest floor. We observed a significant decrease in Ptot concentrations in SSF with increasing soil depth in all three forest ecosystems. It was especially strong in the mineral topsoil, which suggest efficient retention of P by adsorption. However, the actual P concentrations in SSF were highest at the P-rich site (MIT) and lowest at the P-poor site (TUT). Ptot concentrations in SSF at all soil depth and all sites were particularly high in the first 1 to 2 h after the first response of each flow component. For the remaining time of the experiments, transport conditions in the mineral soil and saprolite were however close to chemostatic. E.g., the change (decrease) in Ptot concentrations was factors if not orders of magnitude smaller than the change in SSF during the experiments. Ptot concentrations in the mineral soil did also not change significantly with increasing fraction of event water in SSF towards the end of the experiments. This suggests that the P mobilization in the mineral soil could keep up with P transport. As P transport is closely linked to SSF, P export from forest stands will likely increase on the long-term as the number of large rainfall events associated with SSF is predicted to increase under future climatic conditions. We present results of sprinkling experiments conducted at 200 m^2 hillslopes at three beech forest sites in Germany that differ in their soil depth, skeleton content and soil P stocks, to quantify the dynamic of vertical and lateral SSF and associated P concentrations. Vertical SSF in the mineral soil and saprolite was at least two orders of magnitude larger than lateral SSF and consisted mainly of pre-event water that was likely replaced by sprinkling water (piston flow mechanism). This suggest that previous studies that only measured lateral flow have likely missed the major hydrological export flux from forest stands. Median Ptot concentrations in SSF from the forest floor, and to a lesser extent also at deeper soil depth, at all sites were up to 6 times higher in the first 1 to 2 h after the first response of each flow component (first flush). For the remaining time of the experiments (ca. 10 h), transport conditions in the mineral soil and saprolite were however close to chemostatic. This suggests that the rate of P replenishment at all three sites was high (in the order of minutes to hours). Our finding, that chemostatic transport conditions were prevalent in the mineral soil for most of the experiment even for very high rainfall amounts, would suggest that annual P flux from forest stands could be approximated by simply knowing the average P concentration and the water balance of the site. A quick test of this assumption in the form of a rough approximation of the annual P flux at one of our sites (CON) yielded comparable results to an earlier study at the same site (Sohrt et al., 2019) and some other sites (Benning et al., 2012; Bol et al., 2016). In terms of a P budget our approximated annual P fluxes in SSF at CON were in a similar order than P inputs by dry and wet deposition and mineral weathering and several orders of magnitude smaller than the total P stocks of the mineral soil. This suggest that the annual P fluxes are only relevant on long time scale and that forest ecosystem appear to be relative efficient in retaining P in their nutrient cycle.

6. Data availability

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~~The data will be available from the data repository freiDoc. Until this is set in place please contact the authors for any requests.~~ Data is available under: https://1drv.ms/u/s!AhSOi7EfJ5qmfyJQrT3O65OLBuE?e=BrwfFp

7. Author contribution

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MW wrote the grant; MR was responsible for the~~ project management,~~ experimental design and field installations together with two technicians. MR planed and organized the sprinkling experiments and lab-analysis ~~which was conducted by~~together with two technicians. MR was responsible for data pre-processing and all analysis. JK and FL provided the data on soil characterization and valuable thoughts and discussion on the soil ecological aspect of the study. HP provided meteorological data, supported the long-term monitoring of SSF at CON and provided valuable feedback on the paper manuscript. MR wrote the manuscript including all figures and tables~~. All~~ and all other co-authors discussed the results, provided valuable feedback on the text and figures.

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8. Competing interests:

The authors declare that they have no conflict of interest.

9. Acknowledgments

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Tab. 1: Characteristics of the three experimental sites Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT). [a] classified based on WRB, 2015, [b] (Lang et al., 2017), [c] (Diaconu et al., 2017), all other information is based on own data)

: Characteristics of the three experimental sites Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT). [a] classified based on WRB, 2015, [b] (Lang et al., 2017), [c] (Diaconu et al., 2017), all other information is based on own data)

Properties	MIT	CON	TUT
Soil [a]	Hyperdystric chromic folic cambisol (Humic. Loamic. Nechic)cambisol[a]	Hyperdystric skeletic folic cambisol (Hyperhumic. Loamic)cambisol[a]	Rendzic Leptosols [a]
Humus form [b] (thickness in cm)	Moder (8) [b]	Mor-like Moder (13) [b]	Mull (12)
Soil texture (topsoil/subsoil) [a]	Loam / Sandy Loam [b]	Loam / Sandy Loam [b]	Clay / Clay
Stone content (topsoil/subsoil) [%]	23/26	87/62	50/67
pH(H_2O) (forest floor/ topsoil /subsoil)	3.53 / 3.57 / 4.61 [b]	3.46 / 4.03 / 4.61 [b]	6.00 / 7.23 / 7.84
Ptot stocks forest floor [g/m^2]	7 [b]	13 [b]	19
Ptot stocks mineral soil [g/m^2]	624 [b]	230 [b]	189
isotopically exchangeable P within 1 min (Ah, BA) [mg P/kg]	9.2/3.0 [b]	4.0/5.0 [b]	- / -
Parent material [b]	Paragneiss [b]	Paragneiss [b]	Carbonate Rock
Slope [°]	14°	28°	23°
Elevation [m a.s.l.]	1023 [b]	840 [b]	835
Dominant vegetation (mean age)	Fagus sylvatica (131 [b])	Fagus sylvatica (132 [b])	Fagus sylvatica (90 [dc])
Annual precipitation [mm/year]	1299 [b]	1749 [b]	900 [dc]

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Tab. 2: Depth [cm] of lysimeter (LY) and trench (TR) installations aligned with the main soil horizons for Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT); "B" indicates the sprinkling plot and is used to be consistent with other publications from the same sites. ~~NA~~X indicates that LY4B and TR3B could not be installed at TUT due to shallow soil depth.

Depth in [cm]	MIT	CON	TUT	Lysimeter, Trench
Forest floor	0	0	0	LY1B, TR1B
Topsoil	35	40	40	LY2B
Subsoil	130	100	60	LY3B, TR2B
Saprolite	190	290	~~NA~~X	LY4B, TR3B

Tab. 3: Mean pre-event water fraction [%] of the different flow components (trench: TR1B, TR2B, TR3B and lysimeter: LY1B, LY2B, LY3B, LY4B) and the 6 sprinkling experiments in Mitterfels (MIT), Conventwald (CON), Tuttlingen (TUT) in spring and summer ~~and spring. NA.~~ X indicates that this lysimeter or trench does not exist (TUT~~) or~~), NA indicates that it has not yielded any flow (MIT).

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Summer	15	88	93	98	33	33	NA
MIT Spring	12	83	95	99	9	44	NA
CON Summer	16	58	83	78	24	55	74
CON Spring	10	69	63	86	19	63	78
TUT Summer	4	83	70	NA	14	47	NA
TUT Spring	4	64	95	NA	14	70	NA

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Spring	12	83	95	99	9	44	NA
MIT Summer	15	88	93	98	33	33	NA
CON Spring	10	69	63	86	19	63	78
CON Summer	16	58	83	78	24	55	74
TUT Spring	4	64	95	X	14	70	X
TUT Summer	4	83	70	X	14	47	X

Tab. 4: Time to 20% rise in SSF ~~(t_{rise20})~~ in [min] for all flow components and all sprinkling experiments. ~~X~~"–" indicates that ~~t_{rise20}~~time to 20% rise in SSF could not be calculated due to missing data at the beginning of the event (MIT in spring); ~~NA~~X indicate that this LY or TR was not existing (TUT~~) or~~), NA indicates that it yielded no flow (MIT). Numbers in bold indicate cases where a flow component at deeper soil depth responded earlier than a flow component at shallower soil depth.

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Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Summer	30	210	535	765	20	55	NA
MIT Spring	X	95	290	515	10	80	NA
CON Summer	10	70	180	235	25	305	**265**
CON Spring	15	160	**105**	220	35	315	**235**
TUT Summer	15	205	**140**	NA	20	295	NA
TUT Spring	70	85	140	NA	70	145	NA

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Spring	–	95	290	515	10	80	NA
MIT Summer	30	210	535	765	20	55	NA
CON Spring	15	160	**105**	220	35	315	**235**
CON Summer	10	70	180	235	25	305	**265**
TUT Spring	70	85	140	X	70	145	X
TUT Summer	15	205	**140**	X	20	295	X

Tab. 5: Time to 20% event water fraction ~~(trise20)~~ in [min] for all flow components and all sprinkling experiments; ~~X~~"–" indicates that ~~trise20~~time to 20% event water fraction could not be calculated due to missing data at the beginning of the event (MIT in spring); ~~NA indicate~~X indicates that this LY or TR was not existing (TUT~~) or~~), NA indicates that it yielded no flow (MIT~~). Empty cells~~), "/" indicate that 20 % event water fraction was not reached during the event; numbers in bold indicate that the time to 20% event water fractions was shorter than the time to 20% rise in SSF (see Tab. 4).

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Summer	45	565			30	75	NA
MIT Spring	X	110			15	250	NA
CON Summer	15	115	285	390	35	**70**	**80**
CON Spring	25	295	325	515	45	**80**	**65**
TUT Summer	15	480	370	NA	20	**235**	NA
TUT Spring	75	300		NA	80	380	NA

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Spring	–	110	/	/	15	250	NA
MIT Summer	45	565	/	/	30	75	NA
CON Spring	25	295	325	515	45	**80**	**65**
CON Summer	15	115	285	390	35	**70**	**80**
TUT Spring	75	300	/	X	80	380	X
TUT Summer	15	480	370	X	20	**235**	X

Tab. 6: Slope of the linear regression between Ptot concentration and fraction of event water for the lysimeters (LY) and the trenches (TR) and for all sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT), Bold values are not significantly different from a slope = zero and therefore the relation likely chemostatic (Alpha = 0.95). Italic values indicate slopes that are not chemostatic and not significantly different from slopes describing simple dilution. Therefore, these relations are likely governed by proportional mixing. X indicate that this LY or TR was not existing (TUT), NA indicates that it yielded no flow (MIT).

Events	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Summer	*-2.3*	**-0.3**	**-0.6**	**-1.5**	-0.1	*-0.5*	NA
MIT Spring	-0.6	0.1	**0.0**	1.8	**-0.2**	**0.0**	NA
CON Summer	**-0.1**	**0.0**	*-0.1*	**0.0**	-1.0	**0.1**	**0.1**
CON Spring	-2.2	-1.2	**0.0**	**0.0**	*-1.4*	**0.1**	**0.1**
TUT Summer	*-0.5*	0.1	**0.0**	NA	-0.6	*-0.1*	NA
TUT Spring	-0.1	**0.0**	**0.0**	NA	-0.1	*0.0*	NA

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Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Spring	-0.6	0.1	**0.0**	1.8	**-0.2**	**0.0**	NA
MIT Summer	*-2.3*	**-0.3**	**-0.6**	**-1.5**	-0.1	*-0.5*	NA
CON Spring	-2.2	-1.2	**0.0**	**0.0**	*-1.4*	**0.1**	**0.1**
CON Summer	**-0.1**	**0.0**	*-0.1*	**0.0**	-1.0	**0.1**	**0.1**
TUT Spring	-0.1	**0.0**	**0.0**	X	-0.1	*0.0*	X
TUT Summer	*-0.5*	0.1	**0.0**	X	-0.6	*-0.1*	X

Tab. 7: Estimation of annual Ptot fluxes in vertical (LY) and lateral (TR) SSF at CON calculated based on the mean P concentration after the first flush during the sprinkling experiment at CON in summer (see Fig. 5) and the water flux between 10.05.2018 and 10.05.2019. This period has been chosen in order to not include the sprinkling experiments. No long-term measurements available for MIT and TUT.

	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
Water fux [mm/a]	295	239	320	102	2	4	1
Median P concentration after first flush [mg/l]	0.17	0.07	0.02	0.03	0.15	0.05	0.04
Estimated P flux [kg/ha/a]	0.49	0.16	0.07	0.03	0.00	0.00	0.00

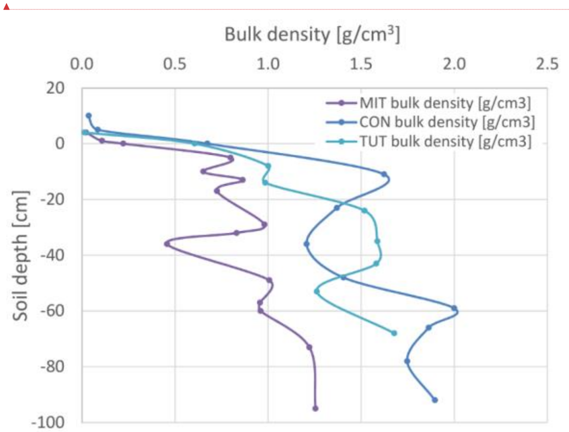

Fig. 1: Depth profile of soil bulk density of the three experimental sites Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT).

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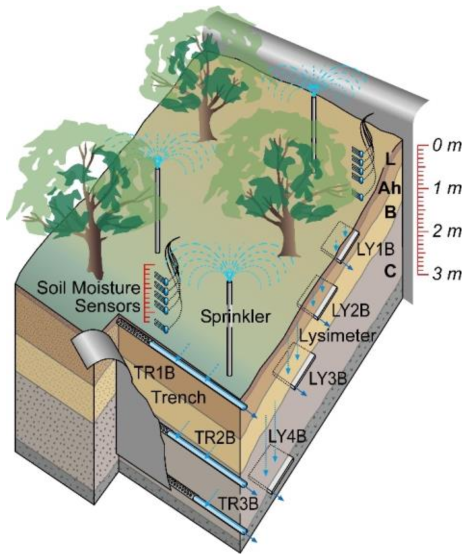

Fig. 2: Experimental setup to measure vertical and lateral subsurface flow with drainage pipes at three soil depth in a trench (TR1B, TR2B, TR3B) and zero tension lysimeters (i.e. steel piling plates pushed in the undisturbed soil profile from the side of the hillslope; LY1B, LY2B, LY3B, LY4B). "B" indicates the sprinkling plot and is used to be consistent with other publications from the same sites.

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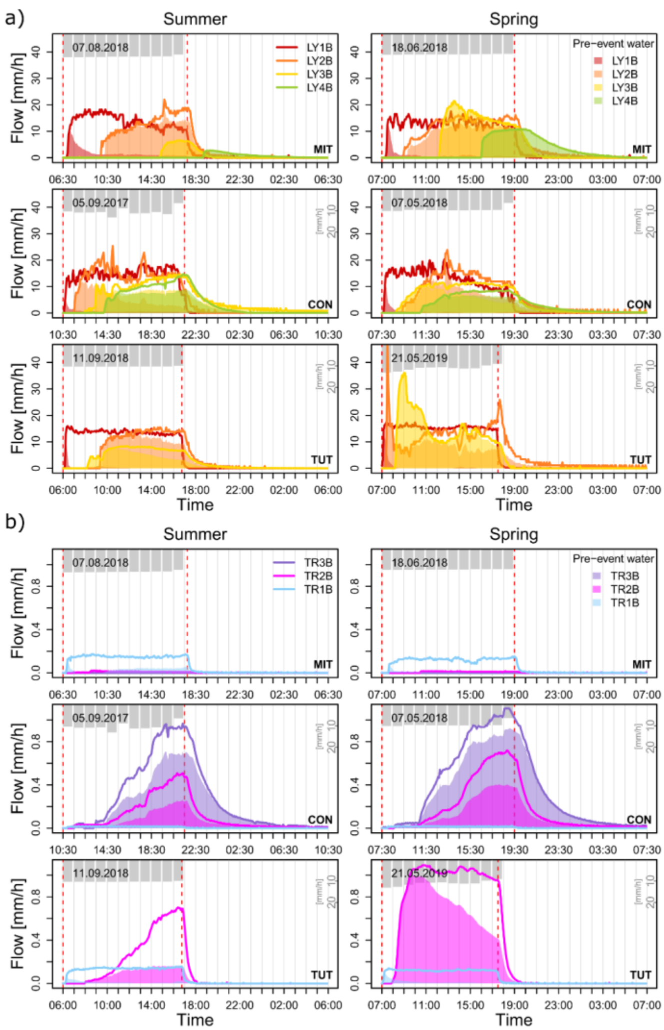

a)
Summer
Spring
07.08.2018
18.06.2018
LY1B
LY2B
LY3B
LY4B
Pre−event water
MIT
05.09.2017
07.05.2018
CON
11.09.2018
21.05.2019
TUT
Flow [mm/h]
Time
[mm/h]
b)
07.08.2018
18.06.2018
TR3B
TR2B
TR1B
05.09.2017
07.05.2018
11.09.2018
21.05.2019

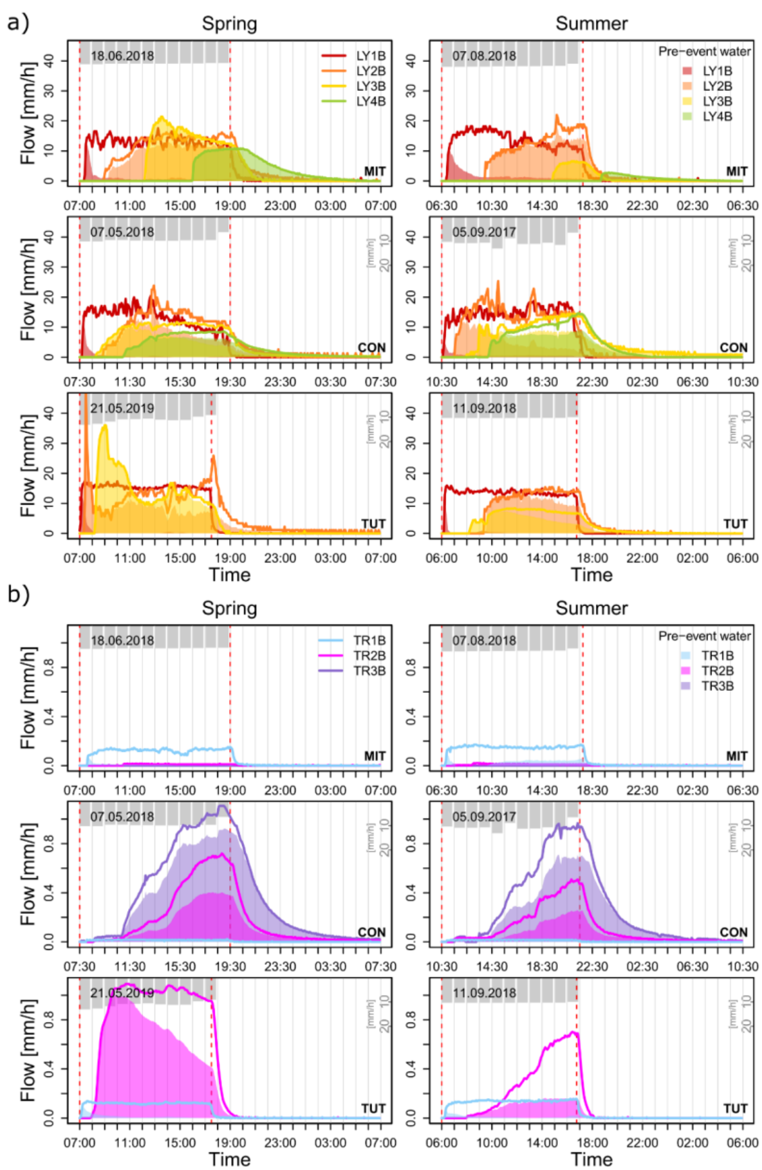
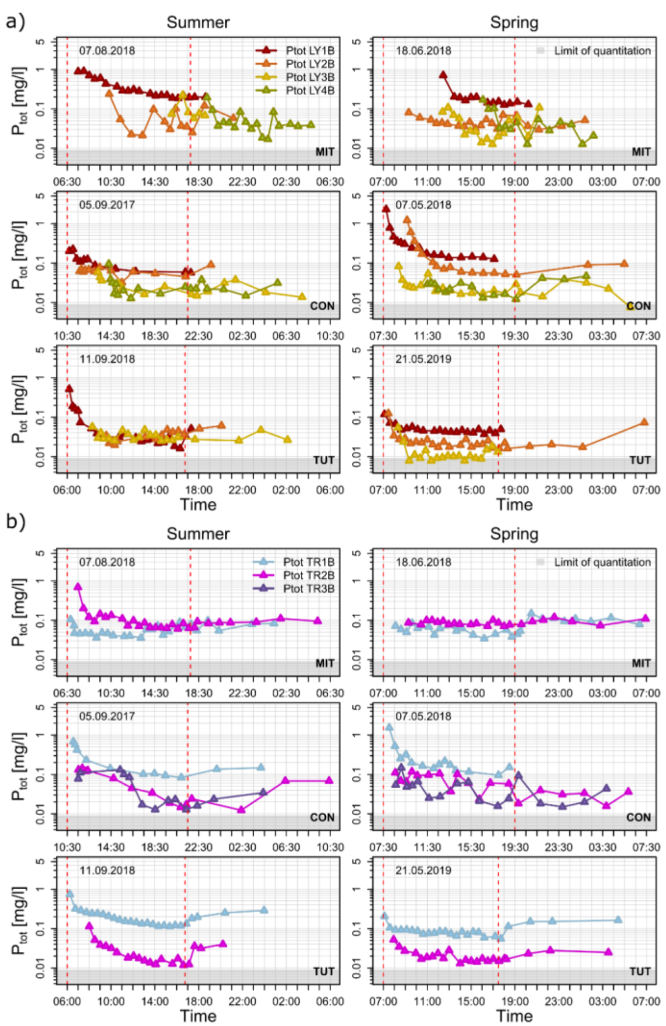

a)
Spring
Summer
18.06.2018
07.08.2018
LY1B
LY2B
LY3B
LY4B
Pre−event water
MIT
07.05.2018
05.09.2017
CON
21.05.2019
11.09.2018
TUT
Flow [mm/h]
Time
b)
TR1B
TR2B
TR3B

Fig. 3: a) Vertical subsurface flow from the lysimeters (LY1B, LY2B, LY3B, LY4B) and b) lateral subsurface flow from the trench (TR1B TR2B, TR3B) for the sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT) sorted in rows ~~from P-rich to P-poor forest ecosystems~~ and for ~~summer~~spring (left column) and ~~spring~~summer (right column). Color-shaded areas show the pre-event water contribution to total discharge of each component (calculated from the Deuterium tracer). Red dashed lines indicate the start and end of the sprinkling experiment. Gray bars show sprinkling input during the experiments.



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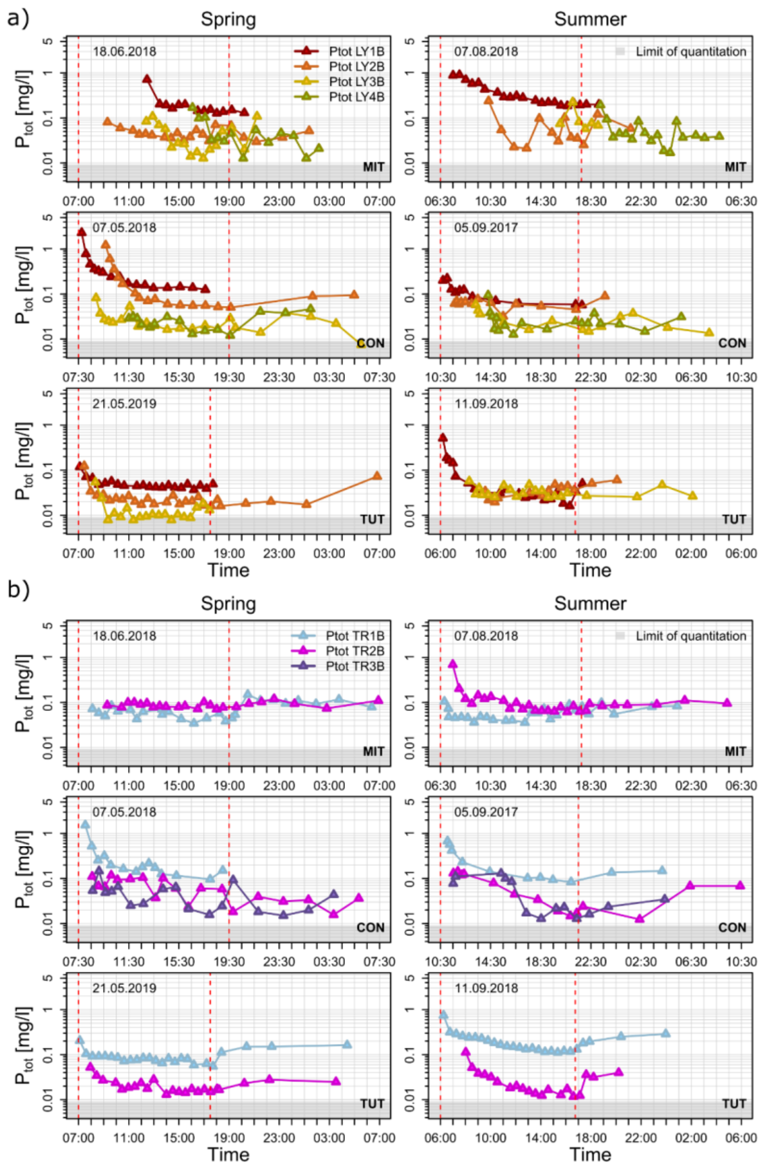
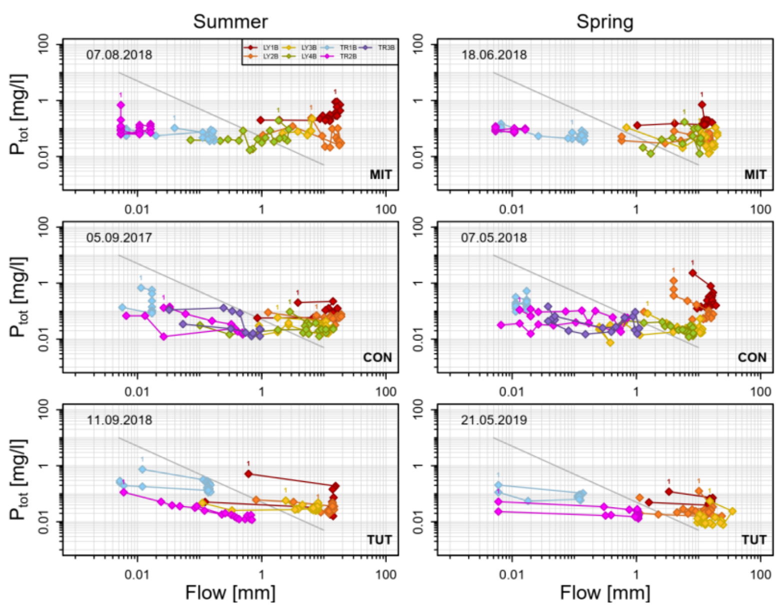

a)
Spring
Summer
Ptot LY1B
Ptot LY2B
Ptot LY3B
Ptot LY4B
Limit of quantitation
18.06.2018
07.08.2018
07.05.2018
05.09.2017
21.05.2019
11.09.2018
MIT
CON
TUT
P_tot [mg/l]
Time
b)
Spring
Summer
Ptot TR1B
Ptot TR2B
Ptot TR3B
Limit of quantitation
18.06.2018
07.08.2018
07.05.2018
05.09.2017
21.05.2019
11.09.2018
MIT
CON
TUT
P_tot [mg/l]
Time

Fig. 4: Ptot concentrations of (a) vertical subsurface flow and (b) lateral subsurface flow for the sprinkling experiments in Mitterfels (MIT), Conventwlad (CON) and Tuttlingen (TUT) sorted in rows ~~from P-rich to P-poor forest ecosystems~~ and for ~~summer~~spring (left column) and ~~spring~~summer (right column). Red dashed lines indicate the start and end of the sprinkling experiment, light gray area indicates range of Ptot below limit of quantitation.



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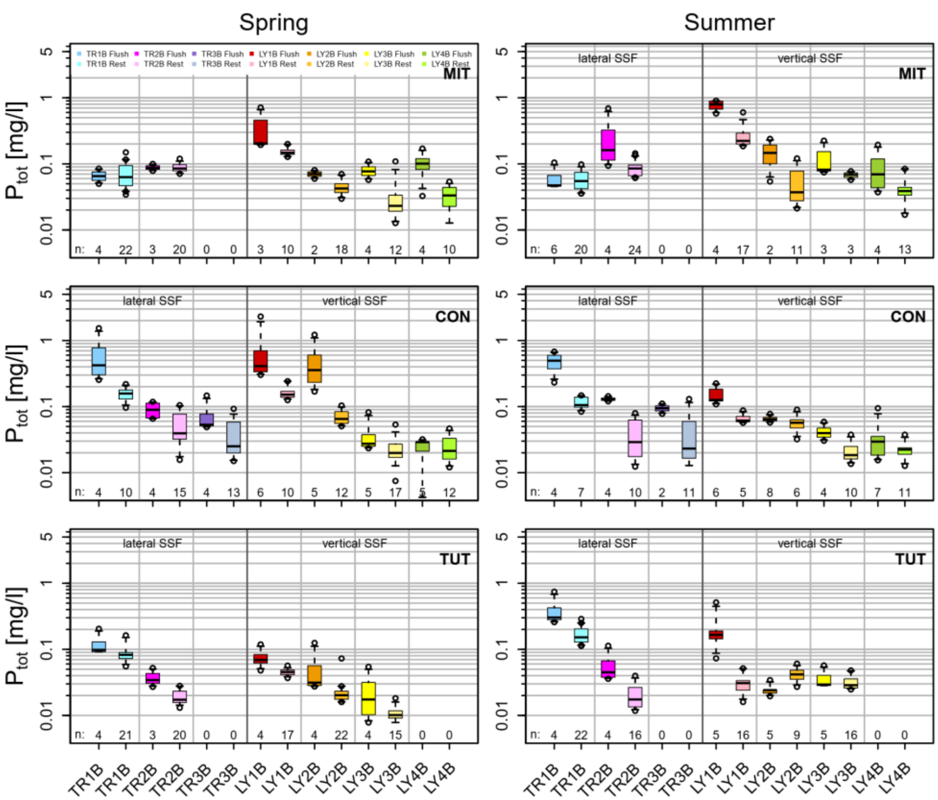

Fig. 5: Ptot concentrations of vertical and lateral subsurface flow (SSF) during the flushing period defined as the first 2 h after onset of each flow component (darker colors) and the remaining part of the event (10 to 12 h) in lighter colors for the sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT).

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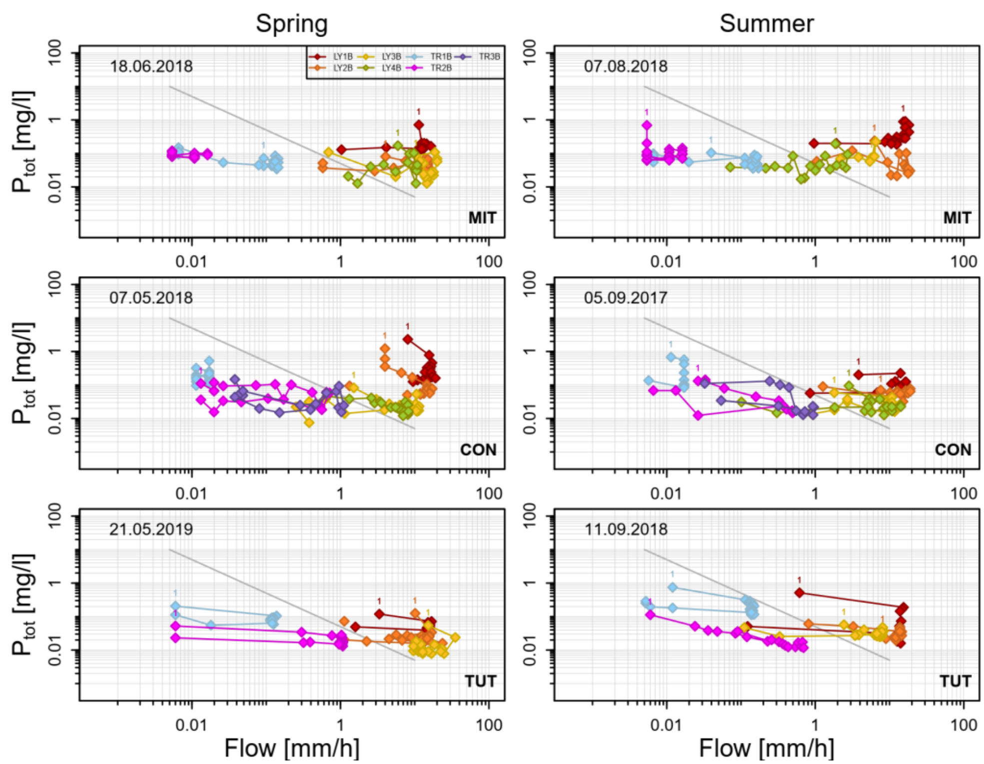

Fig. 6: Ptot concentrations as a function of subsurface flow in the lysimeters (LY) and the trench (TR) during the sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT). The gray diagonal line indicates simple dilution (C ~ 1/Q). Labels "1" indicates the first data point of each flow component.

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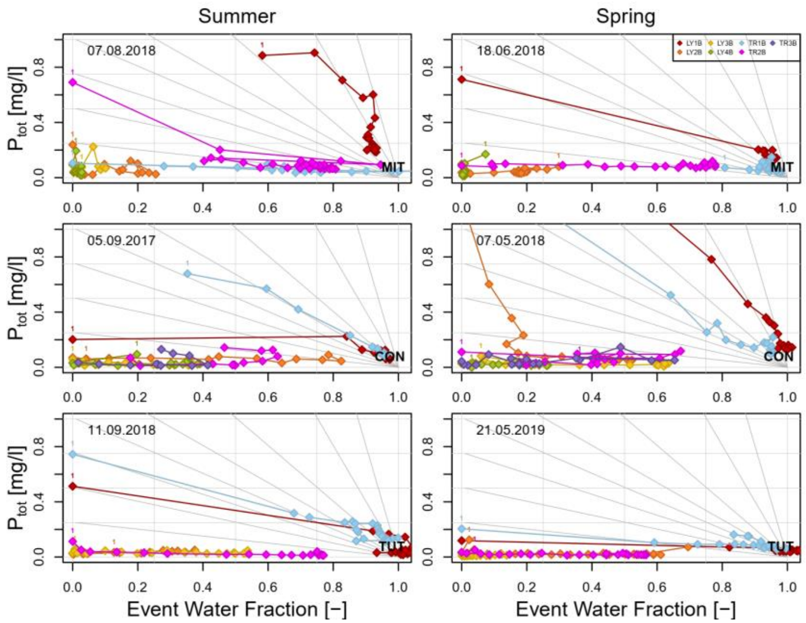
Summer
Spring
07.08.2018
18.06.2018
05.09.2017
07.05.2018
11.09.2018
21.05.2019
MIT
CON
TUT
LY1B
LY2B
LY3B
LY4B
TR1B
TR2B
TR3B
P_{tot} [mg/l]
Event Water Fraction [−]

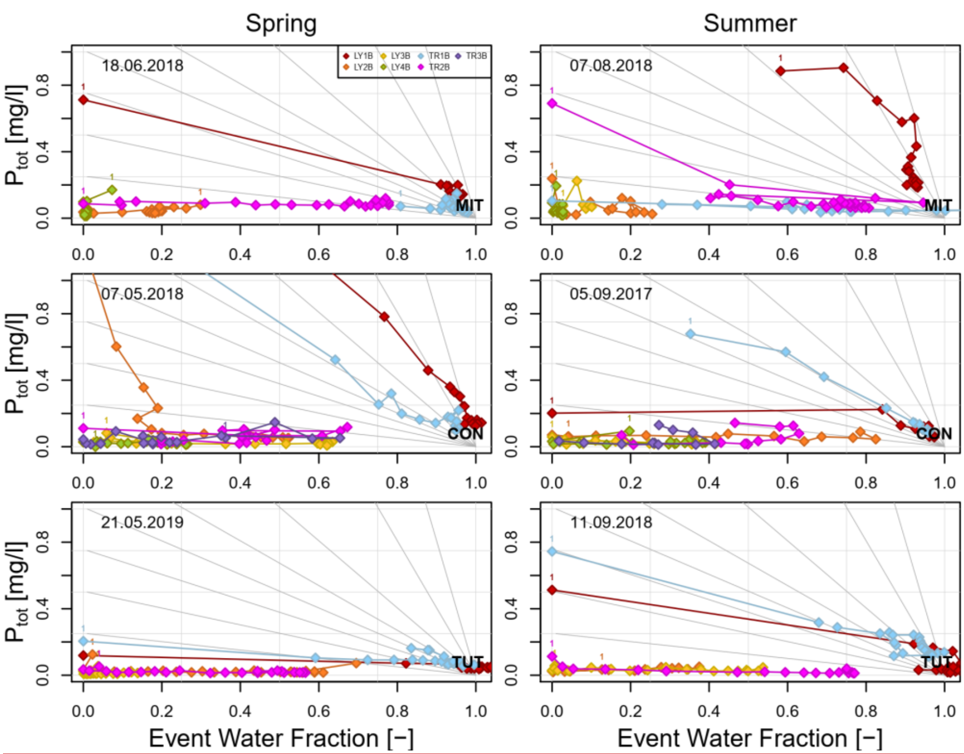

Fig. 7: Ptot concentration as a function of event water fraction calculated for the sprinkling experiments. Gray lines indicate a selection of possible lines that describe the theoretical change in Ptot concentration assuming simple dilution (i.e., proportional mixing of event and pre-event water according to the event water fraction). Labels "1" indicates the first data point of each flow component.

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11. Supplement

Suppl. Tab. 1: Difference between the median Ptot concentration on the rising and falling limb of the sprinkling experiments in [mg/l]. Positive values indicate that Ptot concentrations on the rising limb are higher than on the falling limb and negative values vice versa. Small values indicate that the difference between the median Ptot concentration on the rising and falling limb is small which means that the hysteresis is small and vice versa. Bold values indicate that the difference between Ptot concentration on the rising and falling limb is significantly different. ~~NA~~X indicates that this lysimeter or trench does not exist (TUT~~) or~~), NA indicates that it has not yield any water sample during the experiment (MIT).

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Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Summer	**0.642**	0.010	0.072	**0.104**	-0.007	**0.117**	NA
MIT Spring	**0.051**	0.007	**0.059**	0.011	0.008	0.017	NA
CON Summer	**0.063**	0.004	0.012	0.000	0.488	0.059	0.062
CON Spring	**0.176**	**0.113**	**0.007**	-0.005	0.349	**0.035**	0.027
TUT Summer	**0.319**	**-0.019**	0.003	NA	**0.106**	0.006	NA
TUT Spring	**0.049**	0.003	**0.029**	NA	**0.072**	**0.008**	NA

Event	LY1B	LY2B	LY3B	LY4B	TR1B	TR2B	TR3B
MIT Spring	**0.051**	0.007	**0.059**	0.011	0.008	0.017	NA
MIT Summer	**0.642**	0.010	0.072	**0.104**	-0.007	**0.117**	NA
CON Spring	**0.176**	**0.113**	**0.007**	-0.005	0.349	**0.035**	0.027
CON Summer	**0.063**	0.004	0.012	0.000	0.488	0.059	0.062
TUT Spring	**0.049**	0.003	**0.029**	X	**0.072**	**0.008**	X
TUT Summer	**0.319**	**-0.019**	0.003	X	**0.106**	0.006	X

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