Phosphorus Transport in Subsurface Flow at Beech Forest Stands: Does Phosphorus Mobilization Keep up with Transport?

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Abstract. Phosphorus (P) is a limiting factor of primary productivity in most forest ecosystems but little is known about
10 retention within and losses of P from forests. Subsurface flow (SSF) is one of the important pathways of P export but few
attempts exist to quantify it. We present results of sprinkling experiments with ca. 150 mm, 2H labelled, total rainfall conducted
at 200 m2 plots on hillslopes with slopes between 14° and 28° at three beech forests in Germany in summer and spring. We
aimed at quantifying vertical and lateral SSF and associated P transport in the forest floor, the mineral soil and the saprolite.
The study sites differed regarding soil depth, skeleton content and soil P stocks (between 678 g/m2 and 209 g/m2, in the first
15 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. 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 suggesting effective retention of P by adsorption to soil particles in all three
20 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 conditions.

25 # 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
30 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.
35 (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
40 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).

45 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
50 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 m² 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
55 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 m² 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
60 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.,

65   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

70   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 m² 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

75   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:

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., skeleton content and soil depth)

80   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 and soil P availability?

3.   Is P in the soil solution diluted during large sprinkling experiments or can P mobilization keep up with P transport (chemostatic conditions)?

85   **2. Methods**

2.1 Study Sites

For this analysis three beech 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

90   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 stocks in the upper 1 m of the soil profile is 678 g/m². The saprolite reaches a total depth of 7 m but weathering is less 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,

95   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) and their P stocks 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 has developed that is less rich in soil P stocks (209 g/m^2) than MIT und CON. The soil profile has a 20 - 40 cm deep topsoil and a

100   60 - 80 cm deep subsoil directly overlaying the fractured carbonate parent material (Tab. 2); the stone content of the top- and sub-soil is 50 % and 67 %. 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

105   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 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), but was chosen such that water draining from the forest floor (L, Of, Oh), the mineral soil (A and

110   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

115   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. 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

120   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.

125   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 before leaf senescence. The two periods were chosen to reflect potential seasonal differences in soil moisture and P supply. 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

130 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 15 to 20 mm/h over 10 to 12 h. This reflects a large rainfall event with a return period of more than 100 years for all sites (DWD, 2010). But the selected 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, run through an industrial deionizer (VE-300 (6x50 Liter), AFT GmbH

135 & Co.KG) to remove all minerals (especially P) that is also typically not found in high concentrations in natural rainfall. The deionized water had an electrical conductivity of less than 20 µS/cm. The deionized 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 6 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

140 boundary effects with the otherwise dry surrounding area. 1 kg (first sprinkling experiment) and 2 kg (second experiment) of 99.96 atom % deuterium was added while filling the 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.

145 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 hours) every 30 minutes the SSF of all TR and LY was sampled 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

150 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 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$_2$SO$_4$) and processed in an autoclave. Ptot was analyzed by the molybdenum blue photometric method based on DIN EN ISO 6878 (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

155 (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). 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 two endmember mass balance approach, also called two-component

160 isotope hydrograph separation (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.

To investigate if P mobilization was able to keep up with P transport, we plotted P-concentrations as a function of SSF in log-log space. If the statement before was correct, we would expect near chemotactic 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 flux during all sprinkling experiments. Depending on site and soil depth between 89 and 99 % of total SSF was vertically percolating to deeper depth and only < 1 to 11 % of the sprinkling water was flowing laterally towards the trench (Fig. 3). At all study sites LY1B 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 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 spring experiment, most probable due to wetter antecedent conditions (see discussion section). Maximum
195 SSF from TR2B at CON and TUT was 0.51 and 0.70 mm/h during the experiment 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 pre-event water fractions

Vertical SSF in the topsoil, subsoil and saprolite (i.e., all LY except LY1B) at all sites and during all experiments was
200 predominantly 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 % and 12 %; CON: 16 % and 10 % and TUT: 4 % and 4 % in 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 summer and spring were 88 % and 83 % at MIT, 58 % and 60 % at CON, and 83 % and 64 % at TUT. The mean pre-event water fraction in the vertical
205 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 summer and spring were 33 % and 44 % at MIT, 55 % and 63 % at CON and 47 % and 70 % at TUT. The mean pre-event water fractions during
210 the events in summer and spring of LY3B were 93 % and 95 % at MIT, 83 % and 63 % at CON 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., 74 % and 78 % for TR3B and 78 % and 86 % for LY4B at CON during the experiment in summer and spring.

3.3 Differences in SSF and response time between summer and spring

In general, SSF between the sprinkling experiments in summer and in spring differed less than between the three sites. This
215 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 summer and spring at TUT (see details above).
SSF between 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
220 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 on average was 1.3 times longer than in spring (Tab. 4). At TUT t_{rise20} in the top and subsoil was 1.8 times longer in summer than in spring but t_{rise20} in the forest floor (LY1B and TR1B) was 0.3 times shorter. At CON and TUT the t_{rise20} was not always

increasing systematically with soil depth (e.g., LY3B responded earlier than LY2B at CON in spring and TUT in summer;
225   TR3B responded earlier than TR2B at CON in 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 summer and spring.

The t_{rise20} of the event water fraction (Tab. 4), was typically longer than the t_{rise20} of SSF (Tab. 5). Only for TR2B and TR3B
at CON in 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
230   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 concentrations

Ptot concentrations of vertical SSF (LY) were up to one order of magnitude 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. 4a). This was particularly
235   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, 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 summer and spring and at TUT in summer. All Ptot
240   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 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
245   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
250   (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 summer and spring (Fig. 4b). The difference
in Ptot concentrations in the same flow component in summer and spring was not significant, except for TR1B at TUT.

3.4.1 P concentration as a function of instantaneous flow

The range in SSF was typically factors if not orders of magnitude larger than the range in Ptot concentrations during the
255   sprinkling experiments except for those flow components that yielded little SSF in general (e.g., TR1B at CON during summer

and spring, LY3B and TR2B at MIT during summer) (Fig. 5). 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. 5). 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 concentration were higher on the rising limb than on the falling limb of the SSF hydrographs, except

260   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 (Suppl. Tab. 1).

3.4.2 P concentration as a function of event water fraction

Ptot concentrations also did not change significantly with increasing event water fraction (Fig. 6). Only in the forest floor
265   (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.
270   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
275   regression slope was however close to zero. 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

280   ### 4.1 What are the main SSF paths during long, moderate intense sprinkling events?

In general, vertical SSF dominated total flow 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
285   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.

290 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) who performed a tracer experiment at the Savannah River Site (South Carolina) with 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

295 by piston flow, 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 non-sequential onset of SSF with soil depth and shorter t_{rise20} of event water 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

300 ecosystem (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 and soil bulk density of the three sites (Fig. 1). Swelling and shrinking due to the higher clay content 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

305 (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

310 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

315 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

320   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

325   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

330   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.

335   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

340   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

345   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

between 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).

350   This was particularly true at CON where the high skeleton content allowed the soils to drain quickly to field capacity. Soil

properties 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

355   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

360   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.

5. Conclusions

We present results of six sprinkling experiments conducted at 200 m² hillslopes at three beech forest sites in Germany that

365   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

370   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

375   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

380   long-term as the number of large rainfall events associated with SSF is predicted to increase under future climatic conditions.

6. Data availability

The data will be available from the data repository freiDoc. Until this is set in place please contact the authors for any requests.

7. Author contribution

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 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 soil ecological aspect of the study. MR wrote the manuscript including all figures and tables. All other co-authors discussed the results, provided valuable feedback on the text and figures.

8. Competing interests:

The authors declare that they have no conflict of interest.

9. Acknowledgments

This project was carried out in the framework of the priority program SPP 1685 "Ecosystem Nutrition: Forest Strategies for limited Phosphorus Resources" funded by DFG (grant No. WE 4598/7-2). The article processing charge was funded by the Baden-Wuerttemberg Ministry of Science, Research and Art and the University of Freiburg.

Many thanks to Emil Blattmann, Dominc Demand, Sandra Fritzsche, Maja Gensow, Nina Gottselig, Benjamin Grahler, Janine Heizmann, Barbara Herbstritt, Johannes Herre, Jakub Jeřábek, Britta Kattenstroth, Lara Kirn, Petra Küfner, Hannes Leister, Marvin Lorff, Anna Missong, Regine Nitz, Felix Nyarko, Benjamin Schima, Jonas Schwarz, Stefan Seeger, Jürgen Strub, Angela Thiemann, Delon Wagner, Monika Wirth-Lederer for support in the field, lab and admin. Thanks also to Jakob Sohrt for part of the field installations and insightful discussion; Team- FVA: Hermann Buberl, Andrea Hölscher, Martin Maier, Melanie Strecker, Philine Raubold, Lisa Rubin for good collaboration at CON; Stephan Raspe (LWF Bayern) for meteorological data of MIT. Thanks also to our local partners at the three sites: Hans Amman, Siegfried Fink, Harry Peintinger, Paul Steinhard, Fabian Reichel, Hubert Waldmann, Othmar Winterhalder.

10. References

Achat, D. L., Pousse, N., Nicolas, M., Brédoire, F. and Augusto, L.: Soil properties controlling inorganic phosphorus availability: general results from a national forest network and a global compilation of the literature, Biogeochemistry, 127(2–3), 255–272, doi:10.1007/s10533-015-0178-0, 2016.

Backnäs, S., Laine-Kaulio, H. and Kløve, B.: Phosphorus forms and related soil chemistry in preferential flowpaths and the soil matrix of a forested podzolic till soil profile, Geoderma, 189–190, 50–64, doi:10.1016/j.geoderma.2012.04.016, 2012.

Bol, R., Julich, D., Brödlin, D., Siemens, J., Kaiser, K., Dippold, M. A., Spielvogel, S., Zilla, T., Mewes, D., von Blanckenburg, F., Puhlmann, H., Holzmann, S., Weiler, M., Amelung, W., Lang, F., Kuzyakov, Y., Feger, K., Gottselig, N., Klumpp, E.,

Missong, A., Winkelmann, C., Uhlig, D., Sohrt, J., von Wilpert, K., Wu, B. and Hagedorn, F.: Dissolved and colloidal phosphorus fluxes in forest ecosystems — an almost blind spot in ecosystem research, Plant Nutr. Soil Sci., 179, 425–438, doi:doi.org/10.1002/jpln.201600079, 2016.

Braun, S., Thomas, V. F. D., Quiring, R. and Flückiger, W.: Does nitrogen deposition increase forest production? The role of
415 phosphorus, Environ. Pollut., 158(6), 2043–2052, doi:10.1016/j.envpol.2009.11.030, 2010.

Brinson, M. M.: Decomposition and Nutrient Exchange of Litter in an Alluvial Swamp Forest, Ecology, 58(3), 601–609, 1977.

Burns, D. A., Hooper, R. P., Mcdonnell, J. J., Freer, J. E., Kendall, C. and Beven, K.: Base cation concentrations in subsurface flow from a forested hillslope: The role of flushing frequency, Water Resour., 34(12), 3535–3544, 1998.

Carlyle, G. C. and Hill, A. R.: Groundwater phosphate dynamics in a river riparian zone: Effects of hydrologic flowpaths,
420 lithology and redox chemistry, J. Hydrol., 247(3–4), 151–168, doi:10.1016/S0022-1694(01)00375-4, 2001.

Cole, D. and Rapp, M.: Elemental Cycling in Forest Ecosystems, in Dynamic Properties of Forest Ecosystems, edited by D. Reichle, pp. 341–409, Cambridge University Press., 1981.

Compton, J. E. and Cole, D. W.: Phosphorus cycling and soil P fractions in Douglas-fir and red alder stands, For. Ecol. Manage., 110(1–3), 101–112, doi:10.1016/S0378-1127(98)00278-3, 1998.

425 Cox, J. W., Kirkby, C. A., Chittleborough, D. J., Smythe, L. J. and Fleming, N. K.: Mobility of phosphorus through intact soil cores collected from the Adelaide Hills, South Australia, Aust. J. Soil Res., 38(5), 973–990, doi:10.1071/SR99125, 2000.

Diaconu, D., Kahle, H. P. and Spiecker, H.: Thinning increases drought tolerance of European beech: a case study on two forested slopes on opposite sides of a valley, Eur. J. For. Res., 136(2), 319–328, doi:10.1007/s10342-017-1033-8, 2017.

DIN: DIN ES ISO 6878 - Water quality determination for phosphorus - Ammonium molybdata spectrometric method, , 1–30,
430 2004.

DIN: DIN 32645: Chemical analysis - Decision limit, detection limit and determination limit under repeatability conditions - Terms, methods, evaluation, , 1–20, 2008.

Duquesnay, A., Dupouey, J. L., Clement, A., Ulrich, E. and Le Tacon, F.: Spatial and temporal variability of foliar mineral concentration in beech (Fagus sylvatica) stands in northeastern France, Tree Physiol., 20(1), 13–22,
435 doi:10.1093/treephys/20.1.13, 2000.

DWD: Kostra Rasterdataset, 2010.

Elser, J. J., Fagan, W. F., Denno, R. F., Dobberfuhl, D. R., Folarin, A., Huberty, A., Interlandi, S., Kilham, S. S., McCauley, E., Schulz, K. L., Siemann, E. H. and Sterner, R. W.: Nutritional constraints in terrestrial and freshwater food webs, Nature, 408(6812), 578–580, doi:10.1038/35046058, 2000.

440 Elser, J. J., Bracken, M. E. S., Cleland, E. E., Gruner, D. S., Harpole, W. S., Hillebrand, H., Ngai, J. T., Seabloom, E. W., Shurin, J. B. and Smith, J. E.: Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems, Ecol. Lett., 10(12), 1135–1142, doi:10.1111/j.1461-0248.2007.01113.x, 2007.

Fuchs, J. W., Fox, G. A., Storm, D. E., Penn, C. J. and Brown, G. O.: Subsurface transport of phosphorus in riparian floodplains: Influence of preferential flow paths, J. Environ. Qual., 38(2), 473–484, doi:10.2134/jeq2008.0201, 2009.

445 Gottselig, N., Amelung, W., Kirchner, J. W., Bol, R., Eugster, W., Granger, S. J., Hernández-Crespo, C., Herrmann, F., Keizer, J. J., Korkiakoski, M., Laudon, H., Lehner, I., Löfgren, S., Lohila, A., Macleod, C. J. A., Mölder, M., Müller, C., Nasta, P., Nischwitz, V., Paul-Limoges, E., Pierret, M. C., Pilegaard, K., Romano, N., Sebastià, M. T., Stähli, M., Voltz, M., Vereecken, H., Siemens, J. and Klumpp, E.: Elemental Composition of Natural Nanoparticles and Fine Colloids in European Forest Stream Waters and Their Role as Phosphorus Carriers, Global Biogeochem. Cycles, 31(10), 1592–1607, doi:10.1002/2017GB005657,
450 2017.

Gregory, S. V: Phosphorus dynamics on organic and inorganic substrates in streams, Int. Vereinigung der Angew. und Theor. Limnol., 20, 1340–1346, 1978.

Hagedorn, F. and Bundt, M.: The age of preferential flow paths, Geoderma, 108(1–2), 119–132, doi:10.1016/S0016-7061(02)00129-5, 2002.

455 Heathwaite, A. . and Dils, R. .: Characterising phosphorus loss in surface and subsurface hydrological pathways, Sci. Total Environ., 251–252, 523–538, doi:10.1016/S0048-9697(00)00393-4, 2000.

Hill, B. H., Mccormick, F. H., Harvey, B. C., Johnson, S. L., Warren, M. L. and Elonen, C. M.: Microbial enzyme activity, nutrient uptake and nutrient limitation in forested streams, Freshw. Biol., 55(5), 1005–1019, doi:10.1111/j.1365-2427.2009.02337.x, 2010.

460 Hornberger, G. M., Bencala, K. E. and McKnight, D. M.: Hydrological controls on dissolved organic carbon during snowmelt in the Snake River near Montezuma, Colorado, Biogeochemistry, 25(3), 147–165, doi:10.1007/BF00024390, 1994.

Ilg, K., Wellbrock, N. and Lux, W.: Phosphorus supply and cycling at long-term forest monitoring sites in Germany, Eur. J. For. Res., 128(5), 483–492, doi:10.1007/s10342-009-0297-z, 2009.

Jackson, C. R., Du, E., Klaus, J., Griffiths, N. A., Bitew, M. and McDonnell, J. J.: Interactions among hydraulic conductivity
465 distributions, subsurface topography, and transport thresholds revealed by a multitracer hillslope irrigation experiment, Water Resour. Res., 52(8), 6186–6206, doi:10.1002/2015WR018364, 2016.

Jardine, P. M., Wilson, G. V., McCarthy, J. F., Luxmoore, R. J., Taylor, D. L. and Zelazny, L. W.: Hydrogeochemical processes controlling the transport of dissolved organic carbon through a forested hillslope, J. Contam. Hydrol., 6(1), 3–19, doi:10.1016/0169-7722(90)90008-5, 1990.

470 Johnson, D. W., Trettin, C. C. and Todd, D. E.: Changes in forest floor and soil nutrients in a mixed oak forest 33 years after stem only and whole-tree harvest, For. Ecol. Manage., 361, 56–68, doi:10.1016/j.foreco.2015.11.012, 2016.

Jonard, M., Fürst, A., Verstraeten, A., Thimonier, A., Timmermann, V., Potočić, N., Waldner, P., Benham, S., Hansen, K., Merilä, P., Ponette, Q., de la Cruz, A. C., Roskams, P., Nicolas, M., Croisé, L., Ingerslev, M., Matteucci, G., Decinti, B., Bascietto, M. and Rautio, P.: Tree mineral nutrition is deteriorating in Europe, Glob. Chang. Biol., 21(1), 418–430,
475 doi:10.1111/gcb.12657, 2015.

Julich, D., Julich, S. and Feger, K. H.: Phosphorus fractions in preferential flow pathways and soil matrix in hillslope soils in the Thuringian Forest (Central Germany), Zeitschrift fur Pflanzenernaehrung und Bodenkd., 180(3), 407–417, doi:10.1002/jpln.201600305, 2017a.

Julich, D., Julich, S. and Feger, K. H.: Phosphorus in preferential flow pathways of forest soils in Germany, Forests, 8(1),
480 doi:10.3390/f8010019, 2017b.

Kabeya, D., Inagaki, Y., Noguchi, K. and Han, Q.: Growth rate reduction causes a decline in the annual incremental trunk
growth in masting Fagus crenata trees, , 1444–1452, doi:10.1093/treephys/tpx081, 2017.

Kaiser, K., Guggenberger, G. and Zech, W.: Organically Bound Nutrients in Dissolved Organic Matter Fractions in Seepage
and Pore Water of Weakly Developed Forest Soils, Acta Hydrochim. Hydrobiol., 28(7), 411–419, doi:10.1002/1521-
485 401x(20017)28:7<411::aid-aheh411>3.0.co;2-d, 2000.

Kirschbaum, M. U. F.: The temperature dependence of soil organic matter decomposition, and the effect of global warming
on soil organic C storage, Soil Biol. Biochem., 27(6), 753–760, doi:10.1016/0038-0717(94)00242-S, 1995.

Kunimatsu, T., Hamabata, E., Sudo, M. and Hida, Y.: Comparison of nutrient budgets betwen three forested mountain
watersheds on granite bedrock, Water Sci. Technol., 44(7), 129–140, doi:10.2166/wst.2001.0406, 2001.

490 Lang, F., Krüger, J., Amelung, W., Willbold, S., Frossard, E., Bünemann, E. K., Bauhus, J., Nitschke, R., Kandeler, E., Marhan,
S., Schulz, S., Bergkemper, F., Schloter, M., Luster, J., Guggisberg, F., Kaiser, K., Mikutta, R., Guggenberger, G., Polle, A.,
Pena, R., Prietzel, J., Rodionov, A., Talkner, U., Meesenburg, H., von Wilpert, K., Hölscher, A., Dietrich, H. P. and Chmara,
I.: Soil phosphorus supply controls P nutrition strategies of beech forest ecosystems in Central Europe, Biogeochemistry,
136(1), 5–29, doi:10.1007/s10533-017-0375-0, 2017.

495 Makowski, V., Julich, S., Feger, K., Breuer, L. and Julich, D.: Leaching of dissolved and particulate phosphorus via preferential
flow pathways in a forest soil : An approach using zero-tension lysimeters Leaching of dissolved and particulate phosphorus
via preferential flow pathways in a forest soil : An approach usi, J. Plant Nutr. Soil Sci., doi:10.1002/jpln.201900216, 2020.

Mayerhofer, C., Meißl, G., Klebinder, K., Kohl, B. and Markart, G.: Comparison of the results of a small-plot and a large-plot
rainfall simulator – Effects of land use and land cover on surface runoff in Alpine catchments, Catena, 156, 184–196,
500 doi:10.1016/j.catena.2017.04.009, 2017.

Missong, A., Holzmann, S., Bol, R., Nischwitz, V., Puhlmann, H., v. Wilpert, K., Siemens, J. and Klumpp, E.: Leaching of
natural colloids from forest topsoils and their relevance for phosphorus mobility, Sci. Total Environ., 634, 305–315,
doi:10.1016/j.scitotenv.2018.03.265, 2018a.

Missong, A., Bol, R., Nischwitz, V., Krüger, J., Lang, F., Siemens, J. and Klumpp, E.: Phosphorus in water dispersible-colloids
505 of forest soil profiles, Plant Soil, 427(1–2), 71–86, doi:10.1007/s11104-017-3430-7, 2018b.

Mulholland, P. J. and Hill, W. R.: Seasonal patterns in streamwater nutrient and dissolved organic carbon concentrations:
Separating catchment flow path and in-stream effects, Water Resour. Res., 33(6), 1297–1306, doi:10.1029/97WR00490, 1997.

Qualls, G. Haines, L.: Geochemistry of Dissolved Organic Nutrients in Water Percolating through a Forest Ecosystem, Soil
Sci. Soc. Am. JournalSci, 1112–1123, 1991.

510 Qualls, R. G., Haines, B. L., Swank, W. T. and Tyler, S. W.: Retention of soluble organic nutrients by a forested ecosystem,
Biogeochemistry, 61(2), 135–171, doi:10.1023/A:1020239112586, 2002.

Schindler, D. and Nighswander, J.: Nutrient supply and primary production in Clear Lake, Eastern Ontario, J. Fish. Res. Board

Canada, 27(11), 2009–2036, doi:doi.org/10.1139/f70-226, 1970.

Sklash, M. and Farvolden, R.: The role of groundwater in storm runoff, J. Hydrol., 43, 45–65, 1979.

515 Sohrt, J., Lang, F. and Weiler, M.: Quantifying components of the phosphorus cycle in temperate forests, Wiley Interdiscip. Rev. Water, 4(6), e1243, doi:10.1002/wat2.1243, 2017.

Sohrt, J., Puhlmann, H. and Weiler, M.: Phosphorus transport in lateral subsurface flow at forested hillslopes, SOIL Discuss., (June), 1–20, doi:10.5194/soil-2018-13, 2018.

Sohrt, J., Uhlig, D., Kaiser, K., von Blanckenburg, F., Siemens, J., Seeger, S., Frick, D. A., Krüger, J., Lang, F. and Weiler,
520 M.: Phosphorus Fluxes in a Temperate Forested Watershed: Canopy Leaching, Runoff Sources, and In-Stream Transformation, Front. For. Glob. Chang., 2(December), 1–14, doi:10.3389/ffgc.2019.00085, 2019.

Steegen, A., Govers, G., Takken, I., Nachtergaele, J., Poesen, J. and Merckx, R.: Factors controlling sediment and phosphorus export from two Belgian agricultural catchments, J. Environ. Qual., 30(4), 1249–1258, doi:10.2134/jeq2001.3041249x, 2001.

Stelzer, R. S., Heffernan, J. and Likens, G. E.: The influence of dissolved nutrients and particulate organic matter quality on
525 microbial respiration and biomass in a forest stream, Freshw. Biol., 48(11), 1925–1937, doi:10.1046/j.1365-2427.2003.01141.x, 2003.

Tayor, A., Edwards, E. and Simpson, E.: Nutrients in Streams Draining Woodland and Farmland near Coshocton, Ohio, Water Resour. Res., 7, 81–89, doi:doi.org/10.1029/WR007i001p00081, 1971.

Timmons, D. R., Verry, S., Burwell, R. E. and Holtz, R. F.: Nutrient Transport in Surface Runoff and Interflow from an Aspen-
530 Birch Fores, , 6(2), 1977.

Vanek, V.: Transport of groundwater-borne phosphorus to Lake Bysjön, South Sweden, Hydrobiologia, 251(1–3), 211–216, doi:10.1007/BF00007180, 1993.

van Verseveld, W. J., McDonnell, J. J. and Lajtha, K.: A mechanistic assessment of nutrient flushing at the catchment scale, J. Hydrol., 358(3–4), 268–287, doi:10.1016/j.jhydrol.2008.06.009, 2008.

535 Weiler, M. and Mcdonnell, J.: Testing nutrient flushing hypotheses at the hillslope scale: A virtual experiment approach, J. Hydrol., 319(1–4), 339–356, doi:10.1016/j.jhydrol.2005.06.040, 2006.

Zhang, Z., Fukushima, T., Shi, P., Tao, F., Onda, Y., Gomi, T., Mizugaki, S., Asano, Y., Kosugi, K., Hiramatsu, S., Kitahara, H., Kuraji, K., Terajima, T. and Matsushige, K.: Baseflow concentrations of nitrogen and phosphorus in forested headwaters in Japan, Sci. Total Environ., 402(1), 113–122, doi:10.1016/j.scitotenv.2008.04.045, 2008.

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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)**

	MIT	CON	TUT
Soil [a]	Hyperdystric chromic folic cambisol (Humic. Loamic. Nechic)	Hyperdystric skeletic folic cambisol (Hyperhumic. Loamic)	Rendzic Leptosols
Humus form [b] (thickness in cm)	Moder (8)	Mor-like Moder (13)	Mull (12)
Soil texture (topsoil/subsoil) [a]	Loam/Sany Loam	Loam/Sany Loam	Clay/Clay
Stone content (topsoil/subsoil) [%]	23/26	87/62	50/67
Ptot stocks forest floor [g/m^2]	7	13	19
Ptot stocks mineral soil [g/m^2]	624	230	189
Parent material [b]	Paragneiss	Paragneiss	Carbonate Rock
Slope [°]	14°	28°	23°
Elevation [m a.s.l.]	1023	840	835
Dominant vegetation (mean age)	Fagus sylvatica (131 [b])	Fagus sylvatica (132 [b])	Fagus sylvatica (90 [d])
Annual precipitation [mm/year]	1299 [b]	1749 [b]	900 [d]

545 **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 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	LY4B, TR3B

550 **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 summer and spring. NA indicates that this lysimeter or trench does not exist (TUT) or 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

555 **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} could not be calculated due to missing data at the beginning of the event (MIT in spring); NA indicate that this LY or TR was not existing (TUT) or 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.**

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

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Tab. 5: Time to 20% event water fraction (t_{rise20}) in [min] for all flow components and all sprinkling experiments; X indicates that t_{rise20} could not be calculated due to missing data at the beginning of the event (MIT in spring); NA indicate that this LY or TR was not existing (TUT) or 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

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Tab. 6: Slope of the linear regression between Ptot concentration and fraction of event water for the lysimeters (LY) and the trench (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

570 **that are not chemostatic and not significantly different from slopes describing simple dilution. Therefore, these relations are likely governed by proportional mixing.**

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

575
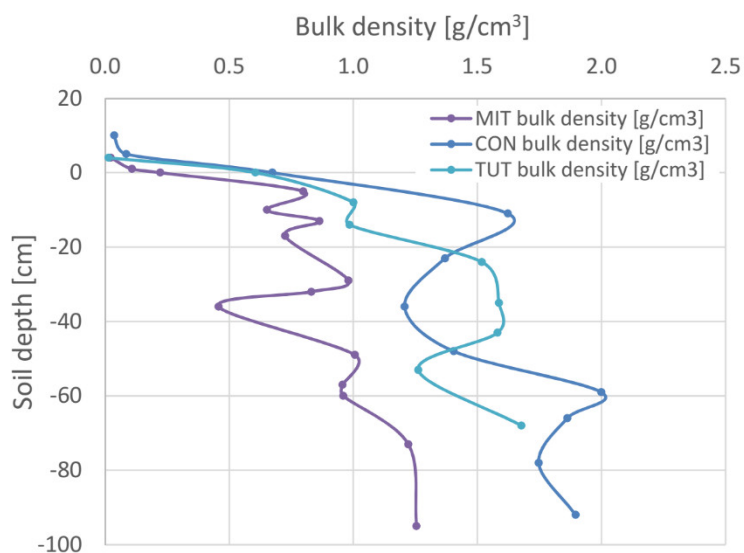
Fig. 1: Depth profile of soil bulk density of the three experimental sites Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT).

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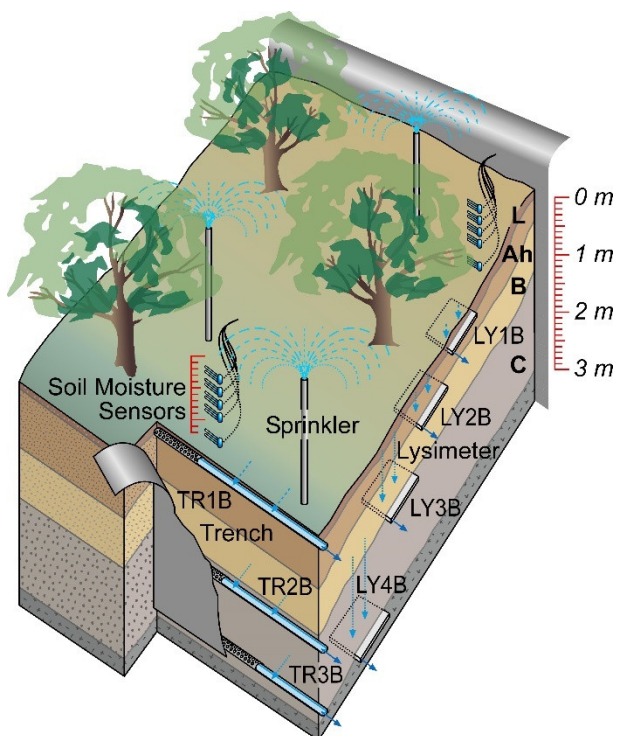
Fig. 2: Experimental setup to measure vertical and lateral subsurface flow with drainage pipes at three soil depth in a trench (TR1B, TR2B, TR3B) and zero tension lysimeters (i.e. steel piling plates pushed in the undisturbed soil profile from the side of the hillslope; LY1B, LY2B, LY3B, LY4B). "B" indicates the sprinkling plot and is used to be consistent with other publications from the same
585 **sites.**
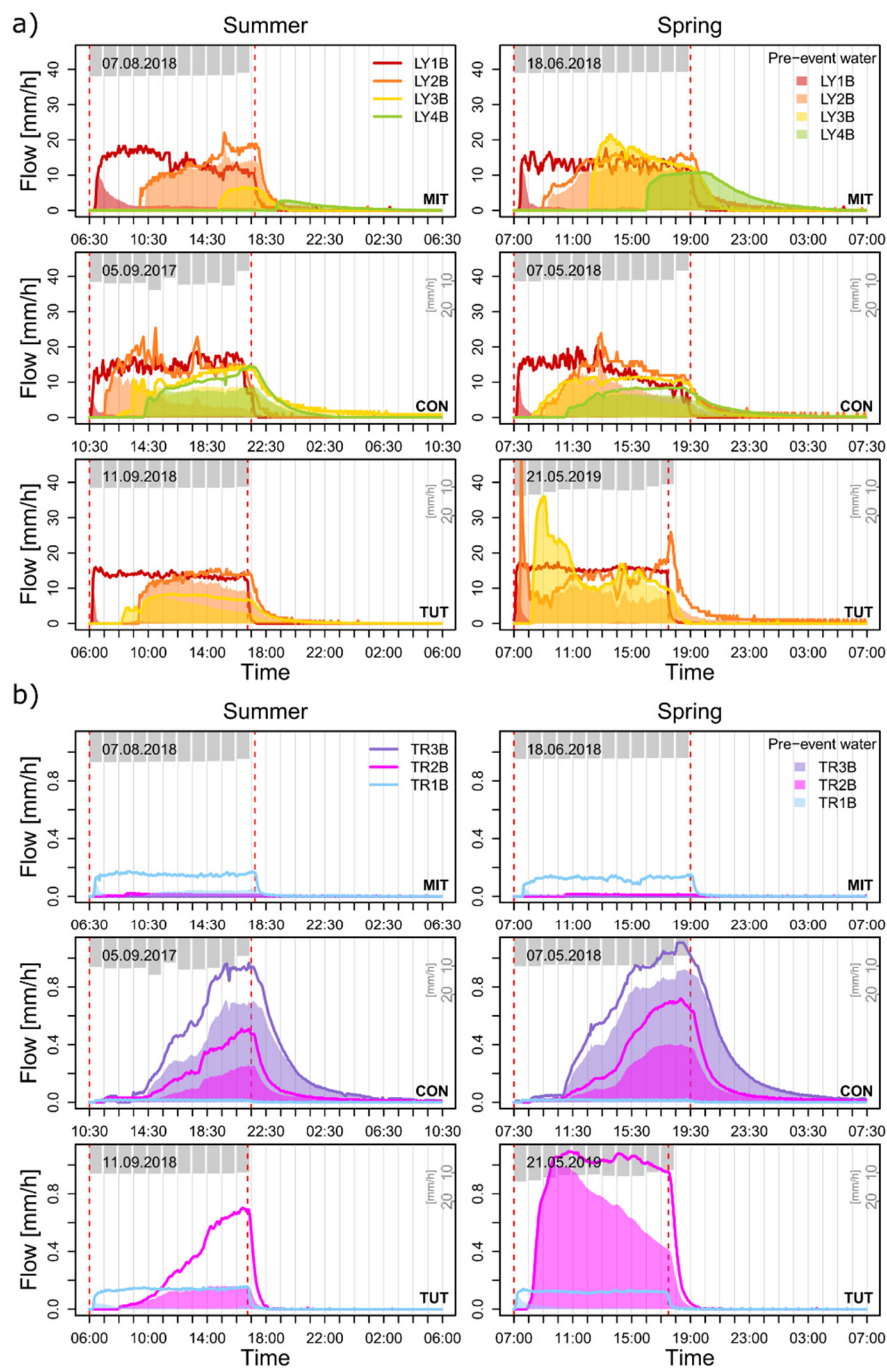
Fig. 3: a) Vertical subsurface flow from the lysimeters (LY1B, LY2B, LY3B, LY4B) and b) lateral subsurface flow from the trench (TR1B TR2B, TR3B) for the sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT) sorted in rows from P-rich to P-poor forest ecosystems and for summer (left column) and spring (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.
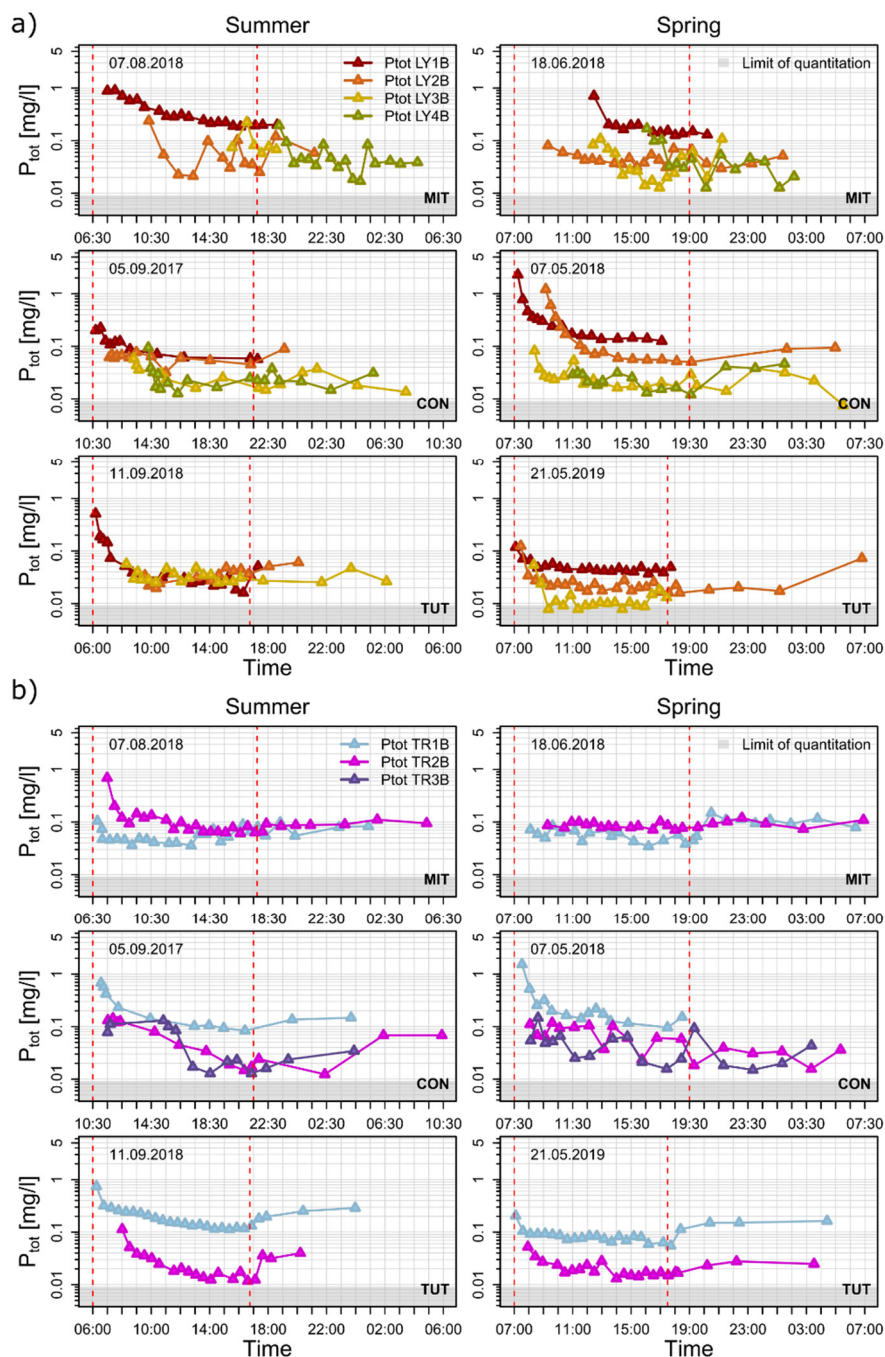
Fig. 4: Ptot concentrations of (a) vertical subsurface flow and (b) lateral subsurface flow for the sprinkling experiments in Mitterfels
595 **(MIT), Conventwlad (CON) and Tuttlingen (TUT) sorted in rows from P-rich to P-poor forest ecosystems for summer (left column)**
and spring (right column). Red dashed lines indicate the start and end of the sprinkling experiment, light gray area indicates range
of Ptot below limit of quantitation.
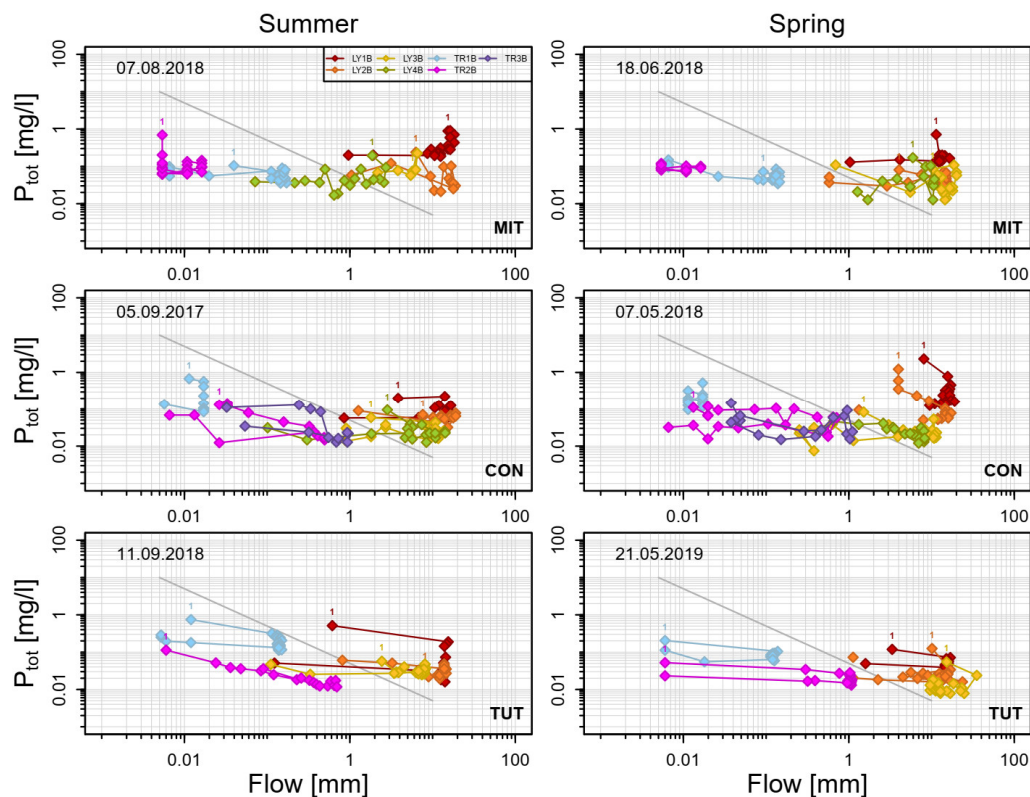
Fig. 5: Ptot concentrations as a function of subsurface flow in the lysimeters (LY) and the trench (TR) during the sprinkling experiments in Mitterfels (MIT), Conventwald (CON) and Tuttlingen (TUT). The gray diagonal line indicates simple dilution ($C \sim 1/Q$). Labels "1" indicates the first data point of each flow component.
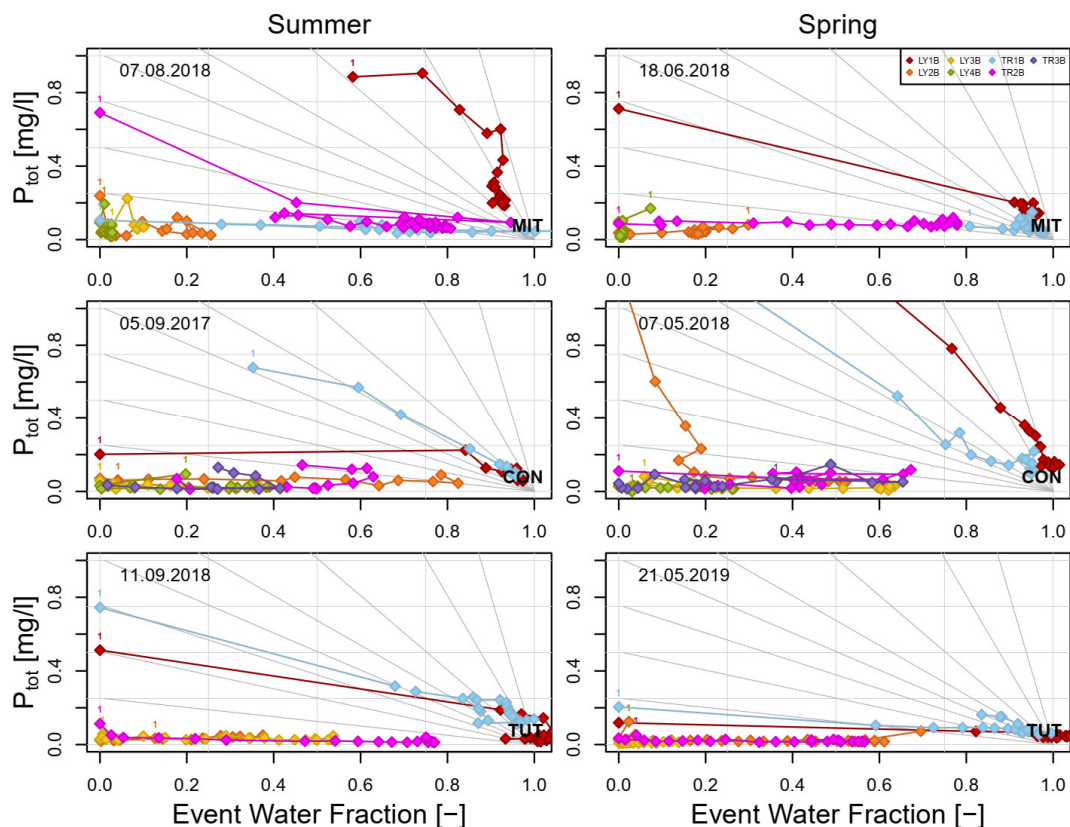
Fig. 6: Ptot concentration as a function of event water fraction calculated for the sprinkling experiments. Gray lines indicate a
605 **selection of possible lines that describe the theoretical change in Ptot concentration assuming simple dilution (i.e., proportional mixing of event and pre-event water according to the event water fraction). Labels "1" indicates the first data point of each flow component.**