

Global Biogeochemical Cycles[®]



RESEARCH ARTICLE

10.1029/2024GB008467

Natural Nanoparticles and Colloids in Forested Streams Across Europe: Seasonal Patterns and Impact of Soil Groups

Key Points:

- Catchments with dystrophic and eutrophic soils had peak colloid concentrations in spring and summer, respectively
- Streams draining Gleysols exhibited irregular colloid concentration peaks that were up to 30x higher than the annual average
- Colloidal org C export was 10x higher in spring versus autumn (dystrophic soils), and 6x higher in summer versus autumn (eutrophic soils)

Supporting Information:

Supporting Information may be found in the online version of this article.

Correspondence to:

H. Schimmel,
heike.schimmel@uni-bonn.de

Citation:

Schimmel, H., Amelung, W., Gottselig, N., Sebastia, M.-T., Keizer, J. J., Martins, M. A. S., et al. (2025). Natural nanoparticles and colloids in forested streams across Europe: Seasonal patterns and impact of soil groups. *Global Biogeochemical Cycles*, 39, e2024GB008467. <https://doi.org/10.1029/2024GB008467>

Received 14 JAN 2025

Accepted 28 MAY 2025

Author Contributions:

Conceptualization: Nina Gottselig, Maria-Teresa Sebastia, Jan Jacob Keizer, Martinho A. S. Martins, Annalea Lohila, Christin Müller, Hjalmar Laudon, Erwin Klumpp







Data curation: Heike Schimmel

Funding acquisition: Nina Gottselig

Investigation: Heike Schimmel, Wulf Amelung, Maria-Teresa Sebastia, Jan Jacob Keizer, Martinho A. S. Martins, Annalea Lohila, Christin Müller, Hjalmar Laudon, Melanie Braun

Methodology: Nina Gottselig, Erwin Klumpp

Project administration: Nina Gottselig, Melanie Braun

Heike Schimmel¹ , Wulf Amelung^{1,2} , Nina Gottselig¹, Maria-Teresa Sebastia^{3,4} , Jan Jacob Keizer⁵ , Martinho A. S. Martins⁵ , Annalea Lohila⁶ , Christin Müller^{7,8}, Hjalmar Laudon⁹ , Erwin Klumpp² , and Melanie Braun¹

¹Soil Science and Soil Ecology, Institute of Crop Science and Resource Conservation (INRES), University of Bonn, Bonn, Germany, ²Institute of Bio- and Geosciences (IBG-3) Agrosphere, Forschungszentrum Jülich, Jülich, Germany, ³Group GAMES and Department of Agricultural and Forest Sciences and Engineering, School of Agrifood and Forestry Engineering and Veterinary Medicine, University of Lleida, Lleida, Spain, ⁴Laboratory of Functional Ecology and Global Change (ECOFUN), Forest Science and Technology Centre of Catalonia (CTFC), Solsona, Spain, ⁵Department Environment and Planning, GeoBioTec & CESAM (Centre for Environmental and Marine Studies), University of Aveiro, Aveiro, Portugal, ⁶Climate System Research, Finnish Meteorological Institute, Helsinki, Finland, ⁷Department Catchment Hydrology, Helmholtz Centre for Environmental Research GmbH – UFZ, Halle (Saale), Germany, ⁸Département des sciences de la Terre et de l'atmosphère, L'Université du Québec à Montréal (UQAM), Montréal, QC, Canada, ⁹Department of Forest Ecology and Management, Swedish University of Agricultural Sciences, Umeå, Sweden

Abstract The element loss from forested ecosystems is largely reflected by the so-called dissolved phase (<450 nm) in stream waters. However, natural colloids (1 nm–1 µm), including natural nanoparticles (NNP, 1–100 nm), constitute a major but largely unexplored part of this dissolved phase. We hypothesized (a) that there is a systematic seasonal variation in colloidal concentrations across European catchments, and (b) that the colloidal concentrations are mainly controlled by site conditions, that is, prevalent reference soil groups. To test these hypotheses, the base flow of seven stream waters was sampled from a transect across Europe from Northern Finland to Portugal at monthly intervals for 1 year. Three colloidal size fractions (1–20 nm, >20–60 nm, and >60 nm) were identified using field flow fractionation coupled with inductively coupled plasma mass spectrometry and an organic carbon detector for the size-resolved detection of Al, Si, P, Ca, Mg, Mn, Fe, Zn, and organic C. Colloids accounted for up to 90%–100% of total element concentrations, emphasizing the importance of colloids for element cycling. Element concentrations showed seasonal patterns in stream waters. However, these patterns differed between the catchments with distinct soil groups: the catchments with dystrophic terrestrial soils exhibited peak flows in colloidal element concentrations during spring, whereas those with eutrophic terrestrial soils did during summer. By contrast, catchments with Gleysols showed season-unrelated single peaks of high element export. Peak concentrations were also influenced by the climatic regime, which in turn impacted colloid abundance.

Plain Language Summary Nutrient and element losses from forests are often assessed through the “dissolved phase” in stream water, which is defined as particles smaller than 450 nm. However, this fraction includes natural colloids, that is, particles between 1 nm and 1 µm in size that can carry high amounts of nutrients but remain understudied. In this study, we investigated how colloidal element concentrations vary seasonally across European forested streams. We collected monthly stream water samples over a 1-year period, covering a climate transect reaching from arctic Finland to Portugal. Element concentrations in the size range of colloids were analyzed. Our results show that colloids can account for up to 100% of the total element concentrations but also that their occurrence shows seasonal trends, which differed between soils: catchments with acidic soils had their mean colloid export peak in spring, while sites with neutral to alkaline pH had it in summer. Groundwater-influenced soils showed single peaks and no seasonal patterns. These seasonal patterns were also influenced by local climate conditions, underlining the complex interplay between soil type, climate, and colloid-mediated element transport.

© 2025. The Author(s).

This is an open access article under the terms of the [Creative Commons Attribution License](https://creativecommons.org/licenses/by/4.0/), which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

[Attribution License](https://creativecommons.org/licenses/by/4.0/), which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

1. Introduction

Forest ecosystems are well-recognized for their efficient biogeochemical cycling of elements, although some elements are lost from these ecosystems through leaching and transport into stream water. Here, the elements are commonly assessed through filtration to 450 nm to obtain the operationally defined so-called “dissolved phase”

Software: Nina Gottselig
Supervision: Wulf Amelung,
 Melanie Braun
Visualization: Heike Schimmel
Writing – original draft:
 Heike Schimmel
Writing – review & editing:
 Wulf Amelung, Maria-Teresa Sebastiá,
 Jan Jacob Keizer, Martinho A. S. Martins,
 Annalea Lohila, Christin Müller,
 Hjalmar Laudon, Erwin Klumpp,
 Melanie Braun

(Binkley et al., 2004; Espinosa et al., 1999). However, this term neglects the occurrence of very fine particles in the size range $<1\ \mu\text{m}$ in the form of natural nanoparticles (1–100 nm; NNP) and colloids (1 nm–1 μm , both of which are summarized here as colloids; Wilkinson & Lead, 2006). Due to their high specific surface area (Baalousha et al., 2009), these particles can be highly reactive, which means they are important in the binding and cycling of elements in surface waters (Gottselig, Amelung, et al., 2017; Hill & Aplin, 2001; Jarvie et al., 2012). In freshwater systems, more than 90% of iron (Fe), aluminum (Al), manganese (Mn), organic carbon (org C) and phosphorus (P) can be associated with colloids, while smaller sized but large colloidal fractions are still found for Ca and Mg (Baalousha et al., 2009; Gottselig, Amelung, et al., 2017). Hence, colloid transport can become the dominant carrier of nutrients in ecosystems. Given this significance for element export, particularly of Al, Si, P, Ca, Mg, Mn, Fe, Zn, and organic C, a better understanding of spatiotemporal patterns of colloidal concentrations and their environmental drivers is essential in order to accurately assess element losses from forest ecosystems.

Most colloids in stream water derive from rock weathering and decomposition of soil organic matter (SOM), frequently forming secondary organo-mineral complexes. While the conditions controlling the formation of these particles are still poorly known, they are likely to differ across climatic regions (Gottselig, Amelung, et al., 2017). This variation might be caused by seasonal variations in parameters such as temperature and precipitation, although the role of these has remained largely unexplored. For instance, temperature affects both plant productivity and microbial activity and thereby soil weathering and SOM decomposition rates (Conant et al., 2011). Precipitation and the soil moisture regime, in turn, control the dissolution of elements and their transport toward the streams. Precipitation may also lead to riverbank erosion, where phyllosilicates in colloidal size ranges enter the streams (Missong, Bol, et al., 2018). Under wet soil conditions, colloids can be released from soils by the reduction of metal(hydr)oxides; by contrast, dried soil conditions favor the cementation and particle bonding, thus inhibiting the release of colloids (X. Jiang et al., 2015; Kjaergaard, Moldrup, et al., 2004; Kjaergaard, Poulsen, et al., 2004). Frequent changes in the soil moisture regime may also induce biological and physical processes that can induce colloid release by means of fragmentation of soil aggregate (Gu et al., 2018). As the formation of colloids in soils as well as their release from soils are linked to precipitation and temperature, it is likely that the colloidal concentrations in stream waters also show clear seasonal patterns.

Previous studies have indicated that soils and groundwater can be major sources of colloids in stream waters (Gottselig et al., 2020; Gu et al., 2018; Missong, Bol, et al., 2018). Especially for the mobile and potentially leachable fraction of soil colloids, so-called water dispersible colloids may reach adjacent streams. The amount and composition of such colloids are variable for different soil horizons, varying, for instance, between organic layers and mineral soil horizons (Huang et al., 2016; Missong, Bol, et al., 2018; Séquaris et al., 2013). Furthermore, Missong, Bol, et al. (2018) reported that NNP concentration decreased with increasing soil depth, being highest under low pH values while the concentrations of fine and medium-sized colloids were at their maximum in the mineral topsoil. The same authors found that colloid composition varied between soil horizons and hence depth. For example, the NNPs of the organic horizons were dominated by organic C and Al, whereas the NNPs of the mineral horizons mainly consisted of Fe, Mn und Si. The elemental composition of soil colloids has also been found to differ between main types of land cover, for example, between forest, grassland and arable land (C. Jiang et al., 2013; X. Jiang et al., 2015; Regelink et al., 2015). The formation as well as the potential mobilization and stability of natural colloids in soil are related to the physical (e.g., moisture and temperature) and chemical (for instance, pH and ionic strength) properties of the soil (C. Jiang et al., 2013; Klitzke & Lang, 2009; Tsao et al., 2011). For example, in forest soils, colloid formation can be promoted by low pH and positively charged metal oxide surfaces as they strongly bind with negatively charged nutrients or SOM (Liang et al., 2010). This can lead to enhanced colloid availability for transport under suitable hydrological conditions. Hence, it is likely that the prevalence of colloids in stream water is affected by the properties of the ecosystem, mostly dependent on the colloidal pattern of soil.

In a former study, Gottselig, Amelung, et al. (2017) showed systematic differences in colloid occurrence and fractionation in stream waters across Europe, depending on dominant forest type, acidity and lithology, following a transition gradient from Ca dominance in the south to a Fe dominance in the north. The same study also pointed out that there are likely to be interactive effects of climate and soil properties due to the negative correlation of temperature and SOM content (Kirschbaum, 2006), providing a higher input of colloidal SOM the further north the stream is located. Consequently, it can be assumed that both climate and ecosystem properties affect the prevalence of colloids in surface waters. What remains unknown is how differences in climate combined with differences in soil properties affect the seasonal pattern of colloids in stream water.

To identify and characterize colloids in the so-called dissolved phase in aqueous and terrestrial systems, field flow fractionation (FFF) has become the method of choice for analyzing the elemental composition of NNP and colloids within their specific size ranges. An advantage of size fractionation of colloids and NNP via FFF is that almost no pre-treatment is necessary, which can alter the particle composition or size range. Accordingly, this method has been used in several studies for colloids analyses in aqueous and soil samples (e.g., Gottselig et al., 2014; Missong, Holzmann, et al., 2018; Moradi et al., 2020), allowing for the size-resolved resolution of elements and org C when coupled online to an inductively coupled plasma mass spectrometry (ICP-MS) and an organic C detector (OCD).

The aim of the present study was to investigate the seasonal pattern of colloidal element concentrations on a continental scale. We hypothesized that there is a systematic seasonal variation of colloidal discharge that is influenced by climatic factors such as precipitation and temperature. Furthermore, we hypothesized that this seasonal variation is controlled by ecosystem properties, that is, by the predominant soil group. Therefore, we sampled seven streams along a climatic transect across Europe, from Arctic Finland in the north to Portugal in the south, covering three major soil groups (dystrophic soils, eutrophic Cambisols and Gleysols). Samples were taken monthly for 1 year to estimate seasonal variations in colloid concentration and composition, and to analyze their dependence on temperature and precipitation. The colloid analysis was performed using FFF coupled with OCD and ICP-MS.

2. Material and Methods

2.1. Sampling Sites

Stream water samples were taken from seven headwater catchments across Europe. Although the sites differed in catchment size, they all had low-intensity forest management practice, high tree coverage and no inflows from urban or agricultural settings, ensuring comparable undisturbed forested conditions. The sites are located along a transect from Northern Finland to Portugal (Figure 1, Table 1), covering a wide range of different mean annual temperatures (MAT) and precipitation (mean annual precipitation (MAP)). The MAT ranged from -1.4°C (Pallas, Finland) to 15.6°C (Ribera Salada, Spain) and the MAP ranged from 484 mm (Pallas, Finland) to 2,293 mm (Alpthal, Switzerland). Monthly temperature and precipitation data were recorded by site operators, and in the case of Bode, data were derived from the German Weather Service (DWD).

The sampling sites comprise different main soil groups, which we categorized into dystrophic soils (Podzols and dystrophic Cambisols), eutrophic Cambisols (Cambisols with a $\text{pH} > 6.5$) and Gleysols (semiterrestrial soils; Table 1). The streams differ further in terms of pH as three sites were eutrophic ($\text{pH} > 6.5$), while four sites were acidic ($\text{pH} \leq 6.5$) (Table 1; Gottselig, Amelung, et al., 2017).

2.2. Sampling

At each site, stream water samples were taken at monthly intervals from September 2018 to August 2019. The exact sampling location was defined by the site operators, following Gottselig, Amelung, et al. (2017). In our previous study, we sampled two transects across Europe that exhibit pH-dependent colloidal element distribution, with an increased presence of Fe and Al colloids in the northern sites and an increased presence of Ca colloids in the southern sites. Of these two transects, the one with the longest distance between the northernmost and southernmost sites was chosen for a more detailed, monthly resolved investigation in the present study. This transect had seven sites, which were continuously monitored by site operators and for which detailed information about site characteristics was available (Table 1).

The sampling was conducted during base flow conditions in the center of the streams without sediment disturbance and upstream, from the outlet to the headwaters, each with between two and four site replicates. Up to four samples of unprocessed stream water were taken manually per site in pre-cleaned polypropylene containers with a volume of 200 ml. Additional samples for organic C analysis were taken with pre-cleaned and pre-equilibrated glass vials. Before sampling, containers were preconditioned with stream water in triplicates. Samples were kept cool during transport and storage until the analyses were conducted, which was as soon as possible after sampling but always within 1 week after sampling. Pre-tests showed that there are no significant alterations in colloid composition in the first week of storage after sampling (Table S11 in Supporting Information S2).

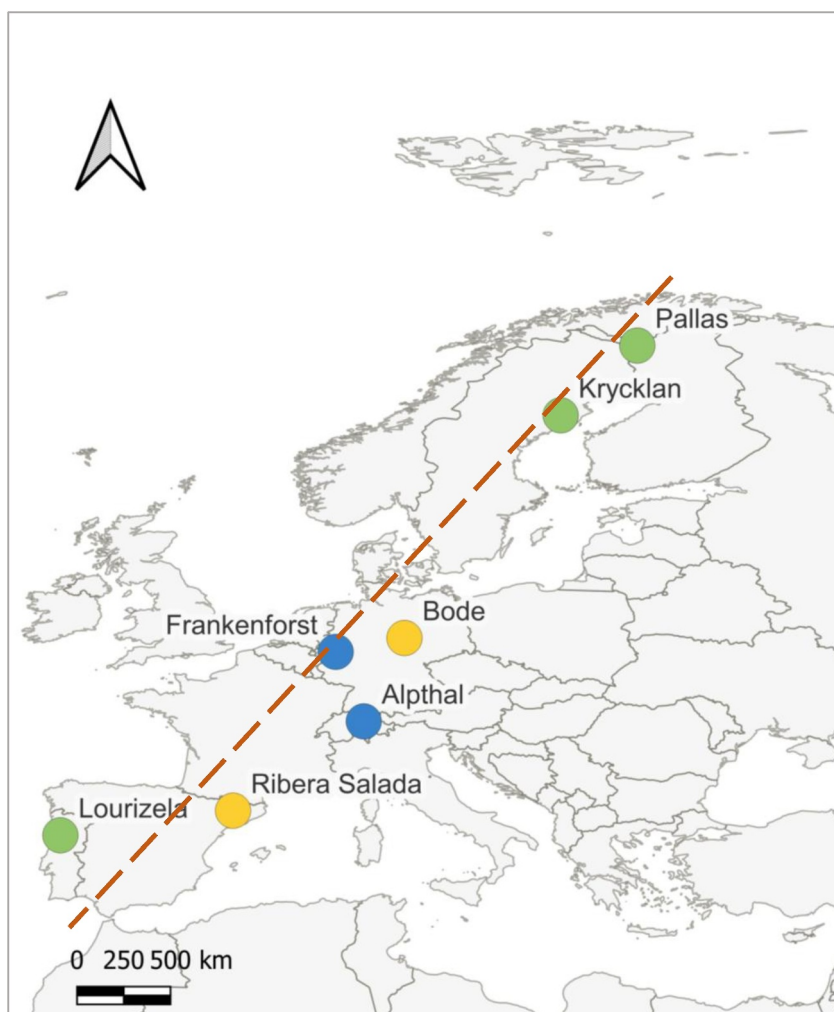


Figure 1. Location of the sampling sites across Europe. Each dot represents a sampling site and the color represents different adjacent soil groups: green = dystrophic soils (including Podzols and Cambisols), yellow = eutrophic Cambisols and blue = Gleysols (Table 1).

Table 1

Characteristics of the Sampling Sites From North to South

Sampling site	MAP	MAT	Soil class	Bedrock type	pH _{stream}	pH _{soil}	Catchment size [km ²]
Pallas	484	−1.4	Dystrophic soil	Granodiorite	6.5	4.8	5.15
Krycklan ^a	614	1.8	Dystrophic soil	Greywacke	4.6	4.8	68
Bode	1,600	7.1	Eutrophic Cambisol	Shale and Greywacke	7.8	6.8	1.27
Frankenforst	650	8.7	Gleysol	Loess	6.2	3.8	n.d.
Alpthal	2,293	6.0	Gleysol	Flysch	7.5	7.1	3.6
Lourizela	1,300	13.8	Dystrophic soil	Schist	6.1	3.9	0.65
Ribera Salada	800	15.6	Eutrophic Cambisol	Carbonatic	8.2	7.4	n.d.

Note. Dystrophic soils include one dystrophic Cambisol (Lourizela) and Podzols (Pallas and Krycklan). MAP = mean annual precipitation; MAT = mean annual temperature (Gottselig, Amelung, et al., 2017). ^aFor further information see Laudon et al. (2021).

2.3. Asymmetric Field Flow Fractionation

Before FFF analysis, the samples were homogenized by shaking them carefully to resuspend any material that had settled and then filtered through a 5 μm cellulose nitrate filter. To avoid excluding particles that were not specific to the given size range and to avoid membrane clogging, we preceded filtering at 0.45 μm . Fractionation of colloids was performed via Asymmetric FFF (AF⁴) following Gottselig, Amelung, et al. (2017), using a 0.5 mm spacer, a 1 kDa polyether sulfone membrane and a 25 μM NaCl eluent. Approximately 5 mL of the samples were injected into the AF⁴ system (see Gottselig, Amelung, et al. (2017) for details).

Colloidal size and turbidity were monitored online via UV detection (254 nm) and dynamic light scattering. Element-specific detection (Al, Si, P, Ca, Mn, Mg, Zn, and Fe) was performed by quadrupole ICP-MS (Agilent 7,500), and org C was detected using an org C Detector (OCD; DOC-Labor, Karlsruhe). Both ICP-MS and OCD were coupled online to AF⁴. OCD measurements used a 1 mL injection and a 10 min focusing time. Blank runs were carried out regularly revealing no contamination. Additional methodological details are available in Gottselig, Amelung, et al. (2017).

2.4. Data Analysis

The ICP-MS raw data were detected in counts per second (cps) using the MassHunter Workstation Software (Agilent Technologies, Japan), while the OCD raw data were recorded in volts detector signal (V) via the AF⁴ analytical software (Postnova Analytics, Landsberg, Germany). Baseline correction, peak integration, and conversion of peak areas to concentrations through multipoint linear calibration were performed using Excel® (Microsoft Corporation, Redmond, USA). Three different pools of elements were considered in this study: (a) elemental concentrations assigned to the first, second, or third size fraction of NNP and fine colloids; (b) all colloidal elemental concentrations – that is, the sum of the concentrations in the first, second, and third fractions combined; and (c) the total concentration of an element in the sample prior to fractionation. Concentrations are primarily given in $\mu\text{mol L}^{-1}$ for Al, Si, P, Mg, Zn, Ca, Mn, and Fe and in mmol L^{-1} for org C.

Graphical and statistical analysis was performed in SigmaPlot (13.0: Systat Software GmbH) and R (version 4.2.2), respectively, using the stats (R Core Team, 2022), tidyverse (Wickham et al., 2019), lme4 (Bates et al., 2015), car (Fox & Weisberg, 2019), lmerTest (Kuznetsova et al., 2017) and emmeans (Lenth, 2023) packages. Assumptions of normality and homoscedasticity of the residues were tested using the Shapiro-Wilk test and the Brown-Forsythe test, respectively. When assumptions were not met, data were log-transformed. All statistical tests involved an alpha of 0.05.

To find a clustering of elements with similar temporal behavior, a principal component analysis (PCA) was performed, where data points were centered and scaled. The correlation between colloidal concentration and total element concentration as well as the correlation between monthly temperature and precipitation and the colloidal concentrations were assessed with log₁₀-transformed data and the Pearson *r* correlation. When comparing the colloidal concentrations of different pH classes (acidic and alkaline), a Wilcoxon test was carried out. To compare the seasonal behavior of colloidal concentrations, the element concentrations per season were tested for significant differences between seasons by carrying out a one-way analysis of variance (ANOVA) with repeated measures, followed by Holm-Sidak's post hoc test. Seasons were defined as follows: spring = March, April, and May; summer = June, July, and August; autumn = September, October, and November; winter = December, January, and February. To find interactions between seasons, soils and colloidal element concentrations, a two-way ANOVA with repeated measures was carried out for each element, followed by a pair-wise comparison using estimated marginal means with Holm correction for multiple testing. We are aware that the requirements for ANOVA analyses are not completely met as our sample sizes are not equal for all groups. Therefore, we additionally used a generalized linear mixed effects model (GLMM) with soil and season as factors. Sampling site and field replicate were used as random intercepts with field replicates nested within sampling sites. We used the glmer function in R:

$$\text{response} \sim \text{as.factor(soil)} * \text{season} + (1|\text{site/replicate}) \quad (1)$$

For Gleysols, we also analyzed whether there are significant differences between months and not only between seasons due to the temporal behavior of Gleysol sites. To test for significant effects of the factors season and soil and their interaction, a type III ANOVA was carried out. For R codes and the data set, please also see Schimmel

et al. (2025). We compared these results with the results of the two-way ANOVA with repeated measures and when a result was significant for both tests, we considered it significant, while we considered it to be a trend when only one of the two results was significant.

3. Results

3.1. Fractionation of Nanoparticles and Colloids

Using AF⁴, three distinct fractions of NNP and fine colloids could be identified for the investigated elements. The first fraction of small-sized NNP ranged from 1 to 20 nm, and the second fraction consisted of intermediate-sized NNP from 20 to 60 nm. Larger-sized NNP and fine colloids fall into the third fraction, with a size range of 60–300 nm.

The median percentages of elements associated with colloids were 27% for Fe (within an interquartile range of 11%–39%), 14% for Al (5%–24%), 11% for org C (4%–22%), but only 9% for Mn (3%–22%), 5% for Zn (2%–10%), 2% for P (1%–5%), 1% for Ca (1%–9%), 1% for Mg (1%–4%) and 0% for Si (0%–1%). For specific sites and months, a maximum of 90%–100% of the total element concentrations for all investigated elements, except for Mg with a maximum of 13%, were associated with colloids. Thus, colloids contributed substantially to the total element concentrations of natural stream waters.

For specific elements, the colloidal concentration was linked to the total element concentration. The strongest correlation was observed for Fe ($r = 0.83$, $p < 0.001$) and Al ($r = 0.75$, $p < 0.001$, Figure 2), especially in the case of the large NNP and fine colloids ($r = 0.87$, $p < 0.001$ and $r = 0.73$, $p < 0.001$, respectively; Table S3 in Supporting Information S2). Similarly, org C ($r = 0.69$, $p < 0.001$) and Mn ($r = 0.65$, $p < 0.001$) showed strong correlations between total and colloidal element concentration; again, the correlation was strongest in the case of large NNPs and fine colloids ($r = 0.70$, $p < 0.001$ and $r = 0.72$, $p < 0.001$, respectively, Figure 2 and Figures S1–S3 in Supporting Information S1). In the case of Ca and Zn, only the concentrations of the fraction of medium-sized NNPs correlated with the total concentration ($r = 0.62$, $p < 0.001$ and $r = 0.60$, $p < 0.001$, respectively, Table S3 in Supporting Information S2), while there were weak or scattered relationships between total and colloidal element concentrations for Mg, P, and Si ($r < 0.5$). The concentration of these elements in the colloidal size range in all three fractions did not change systematically with total concentrations (Figure 2, Figures S1–S3 in Supporting Information S1, Table S3 in Supporting Information S2). The highest correlation between individual colloidal elements was found for Ca and Mg as well as for Al and Fe ($r = 0.80$, $p < 0.001$ and $r = 0.86$, $p < 0.001$, respectively; Table S4 in Supporting Information S2).

Based on PCA and Pearson's Correlation (Figure 3 and Table S4 in Supporting Information S2), we identified four distinct groups which showed similar behavior for the colloidal concentration: (a) Al, Si, Fe, Mn, and org C, (b) Mg and Ca, (c) P, and (d) Zn. The latter two did not group with other elements. As the elements within one group showed similar seasonal behavior, only one representative element per group will be shown in the following: (a) org C, (b) Ca, (c) P, and (d) Zn.

3.2. Seasonal Pattern of Nanoparticles and Colloids

3.2.1. Influence of Temperature, Precipitation and pH

Looking at all sites, the concentrations of most measured elements were only poorly related to temperature and precipitation. Magnesium and Ca concentrations showed a weak positive correlation with temperature ($r = 0.36$, $p < 0.001$ and $r = 0.33$, $p < 0.001$, respectively), while Zn was negatively correlated to temperature ($r = -0.53$, $p < 0.001$) (Table S4 in Supporting Information S2). Dividing the sites by pH classes revealed that acidic sites had significantly higher concentrations of Fe ($p < 0.05$), Mn ($p < 0.001$), and Zn ($p < 0.001$), while eutrophic sites showed significantly higher amounts of Ca ($p < 0.001$). The pH was not significantly related to the concentrations of Al, P, org C, Si, and Mg ($p > 0.05$). Differencing between acidic and eutrophic sites did not improve the correlation between colloidal concentrations and temperature and precipitation.

3.2.2. Influence of Soil on Seasonal Behavior of Nanoparticles and Colloids

Besides climatic factors and pH, the categorized soil groups had a significant effect on colloidal element concentration. The soil group had a significant impact on the colloidal concentration of Mg and Ca ($p < 0.05$), while

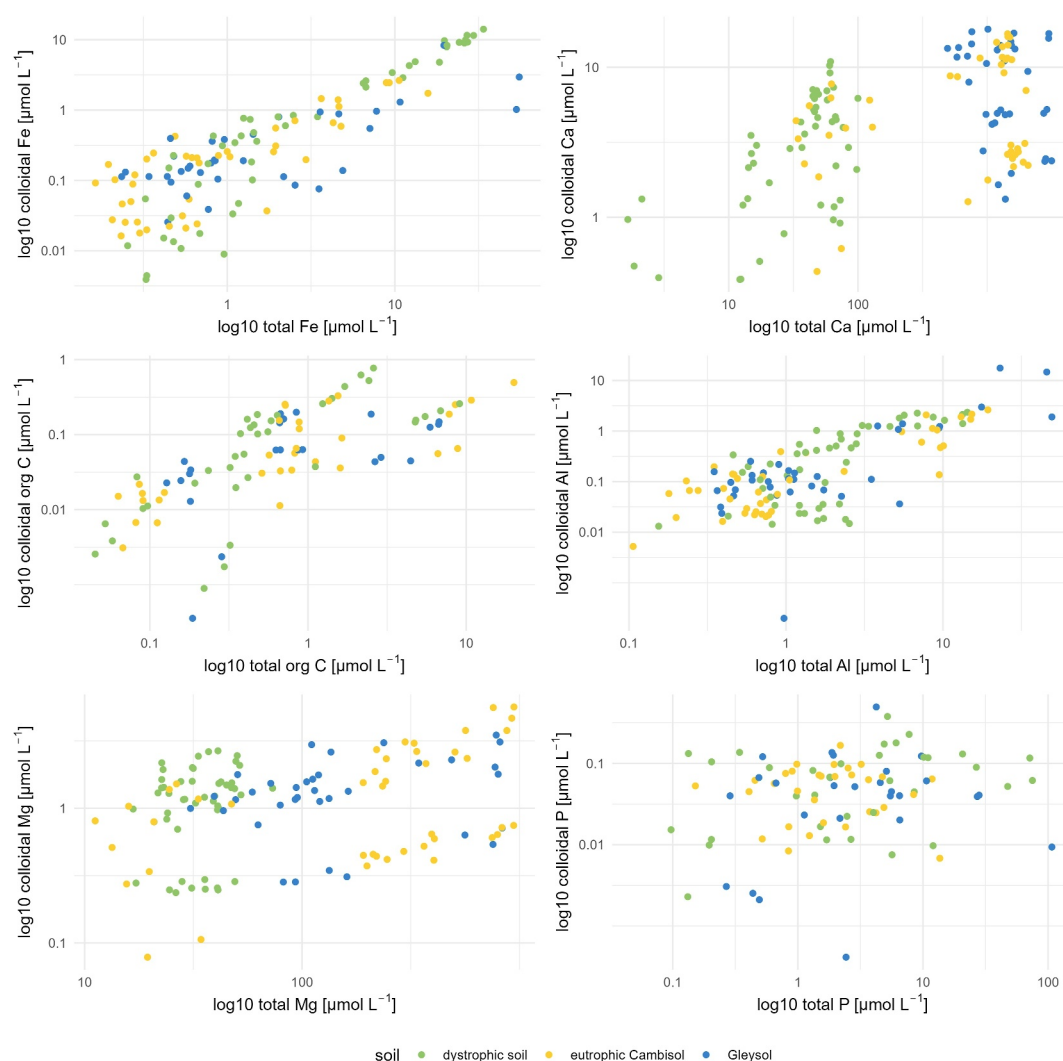


Figure 2. Relationships between colloidal element concentrations and total element concentrations for six selected elements, grouped by soil class (colors). The axes are shown on a logarithmic scale to illustrate concentration differences across several orders of magnitude.

there was a trend for org C, Al, Zn, Si, Fe, and Mn (ANOVA $p < 0.05$; GLMM $p > 0.05$). The categorized soil group had no significant effect on colloidal P concentration ($p > 0.05$).

The two-way ANOVA with repeated measures as well as the GLMM revealed that the interaction of season and categorized soil group also had a significant influence on the colloidal concentrations of Al, Fe, Mg, and Ca ($p < 0.05$) and there was a trend for Zn, Si, and org C (ANOVA $p < 0.05$, GLMM $p > 0.05$), while there was no influence on P and Mn ($p > 0.05$).

Thus, while the total colloidal element concentration differed between soils, the seasonal behavior of the colloidal concentrations also showed differences. Dystrophic soils showed a peak in the concentrations of org C (Figure 4) in spring with a decrease toward autumn and winter months ($p < 0.05$). Zn concentrations were significantly lower in spring than in the other three seasons ($p < 0.05$). Ca concentration of the dystrophic soils showed two peaks in April and August, and significant differences between summer and spring compared to autumn and winter ($p < 0.001$) (Figure 4).

Eutrophic Cambisols tended to have their peak element discharge in late spring and summer. Iron, Mn, and org C (as shown for org C; Figure 4) showed a trend of higher concentrations in summer months than in autumn (ANOVA $p < 0.05$). The same trend was observed for Si (Figure S10 in Supporting Information S1); here,

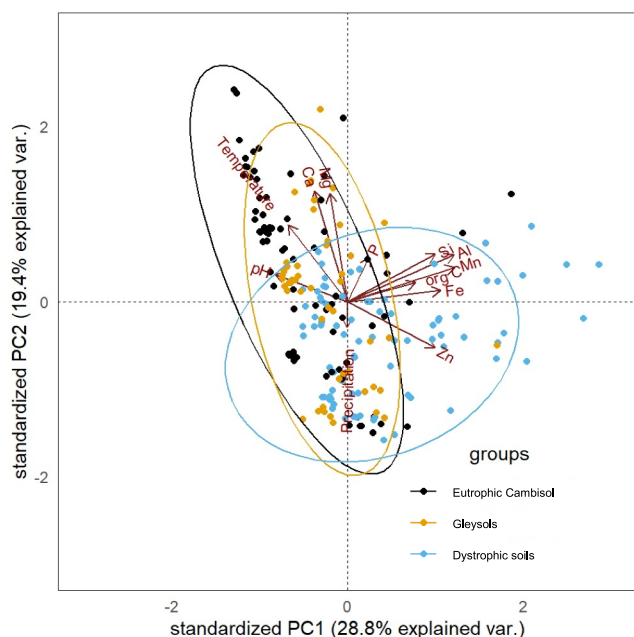


Figure 3. Principal component analysis conducted to show correlations between different elements.

concentrations tended to be higher in summer than in the three other seasons (ANOVA $p < 0.05$), while for Al the concentrations were significantly higher in summer than in the other three seasons ($p < 0.05$). For Ca, there was a stable element concentration from February to October, which decreased sharply in November with significantly less discharge in winter ($p < 0.05$). Zinc showed the opposite behavior to Ca, with significantly higher concentrations in winter compared to low concentrations in spring and summer ($p < 0.001$) (Figure 4).

In contrast to dystrophic soils and eutrophic Cambisols, Gleysols had single events of high element discharge with no clear seasonality. Organic C had peak concentrations in March, May and August, which were significantly higher than concentrations in January and February ($p < 0.01$) for all 3 months, while May and August concentrations were also significantly higher than concentrations in November and October. In the same months, Al showed peak concentrations that were significantly higher than the concentrations in September, October, and November ($p < 0.01$, Figure S3 in Supporting Information S1). For Ca, the Gleysols showed a similar pattern as the eutrophic Cambisols with a stable Ca concentration from spring to autumn and a significantly lower Ca discharge in the winter months ($p < 0.001$). Also, for Zn, the seasonal behavior was similar in Gleysols as for eutrophic Cambisols with significantly higher concentrations in winter ($p < 0.01$) (Figure 4).

Further, the categorized soil group affected the correlation between elements within the colloidal fraction. For dystrophic soils, Mn was not correlated with Al, Si, and org C (Table S5 in Supporting Information S2). For eutrophic soils, Si showed only a correlation to Zn (Table S6 in Supporting Information S2). For Gleysols, by contrast, all elements were correlated with Fe except for Zn. Further, P, which was usually not correlated with other elements (except to Mn for eutrophic Cambisols), showed significant correlations with Al and Si (common in clay minerals) as well as with org C and Ca (Table S7 in Supporting Information S2).

When testing the relation between colloidal concentrations in stream waters for single elements with monthly temperature and precipitation, new significant correlations appeared (Tables S8–S10 in Supporting Information S2): Calcium, Fe, and Mn showed a negative correlation to precipitation for dystrophic soils (Table S8 in Supporting Information S2), while for all three elements, the opposite trend was observed for eutrophic soils (Table S9 in Supporting Information S2). There, org C was negatively correlated with temperature but positively correlated with precipitation ($r = -0.44$ and 0.59 , respectively, Table S9 in Supporting Information S2). Also, Ca was negatively correlated to precipitation in eutrophic soils ($r = -0.67$; Table S9 in Supporting Information S2). In addition, in eutrophic soils Ca was positively correlated with temperature and the same correlation was observed for Mg (Table S9 in Supporting Information S2). By comparison, the Gleysol sites exhibit the lowest number of correlations. Here, only Mg showed the same trend as for eutrophic soils, and Zn was negatively correlated to temperature, again similar to eutrophic soils (Tables S9 and S10 in Supporting Information S2).

4. Discussion

4.1. Contribution of Nanoparticles and Colloids to the Element Cycling in European Forested Streams

Previous studies have shown the importance of nanoparticles and colloids as constituents of the dissolved phase (Baken et al., 2016; Gottselig et al., 2014; Gottselig, Amelung, et al., 2017; Gottselig, Nischwitz, et al., 2017; Missong, Holzmann, et al., 2018). In the present study, nanoparticles and colloids <300 nm accounted for up to 94% (Zn), 96% (Si), and 100% (Al, Fe, Ca, Mn, and P) of total element concentrations, corroborating results of previous studies for soils and stream waters (Gottselig, Nischwitz, et al., 2017; Hill & Aplin, 2001; Jarvie et al., 2012). Different elements showed close correlations with total amounts as a function of the size fractions, such as Fe and Al as well as organic C and Mn in the large NNP and fine colloids, possibly indicating the presence of oxides and organo-mineral complexes in this colloidal size fraction likely dominated their total discharge, with additional preference to organic C (Gottselig, Amelung, et al., 2017). As Figure 2 and Figure S4 in Supporting Information S1 show that the largest colloid fraction did not consistently contain higher absolute concentrations of, for example, Al

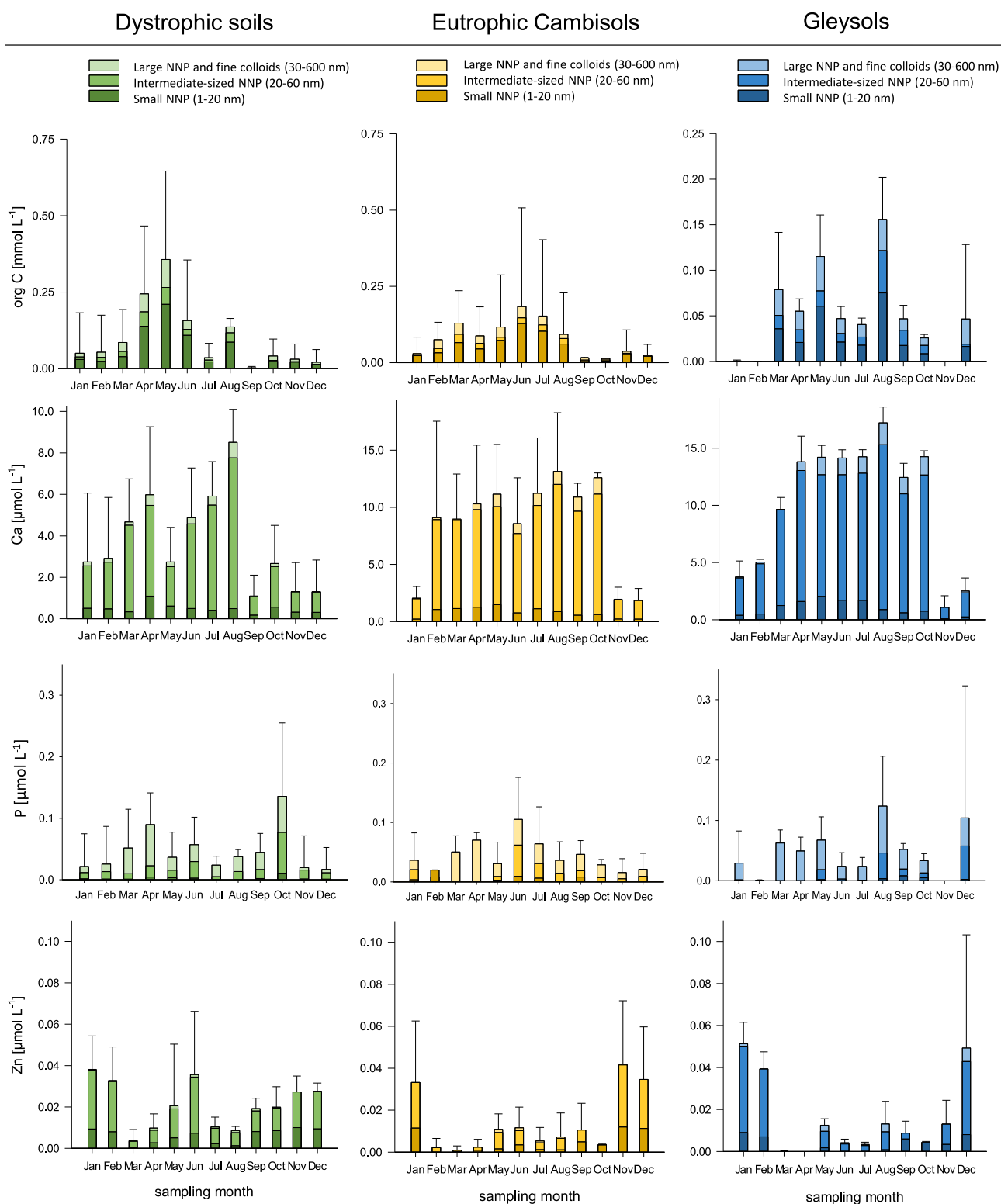


Figure 4. Monthly concentrations of elements grouped by three different categorized soil groups. Error bars represent the cumulative error from the three size fractions of nanoparticles (NNP) and colloids. Please note the different scaling of the y-axis.

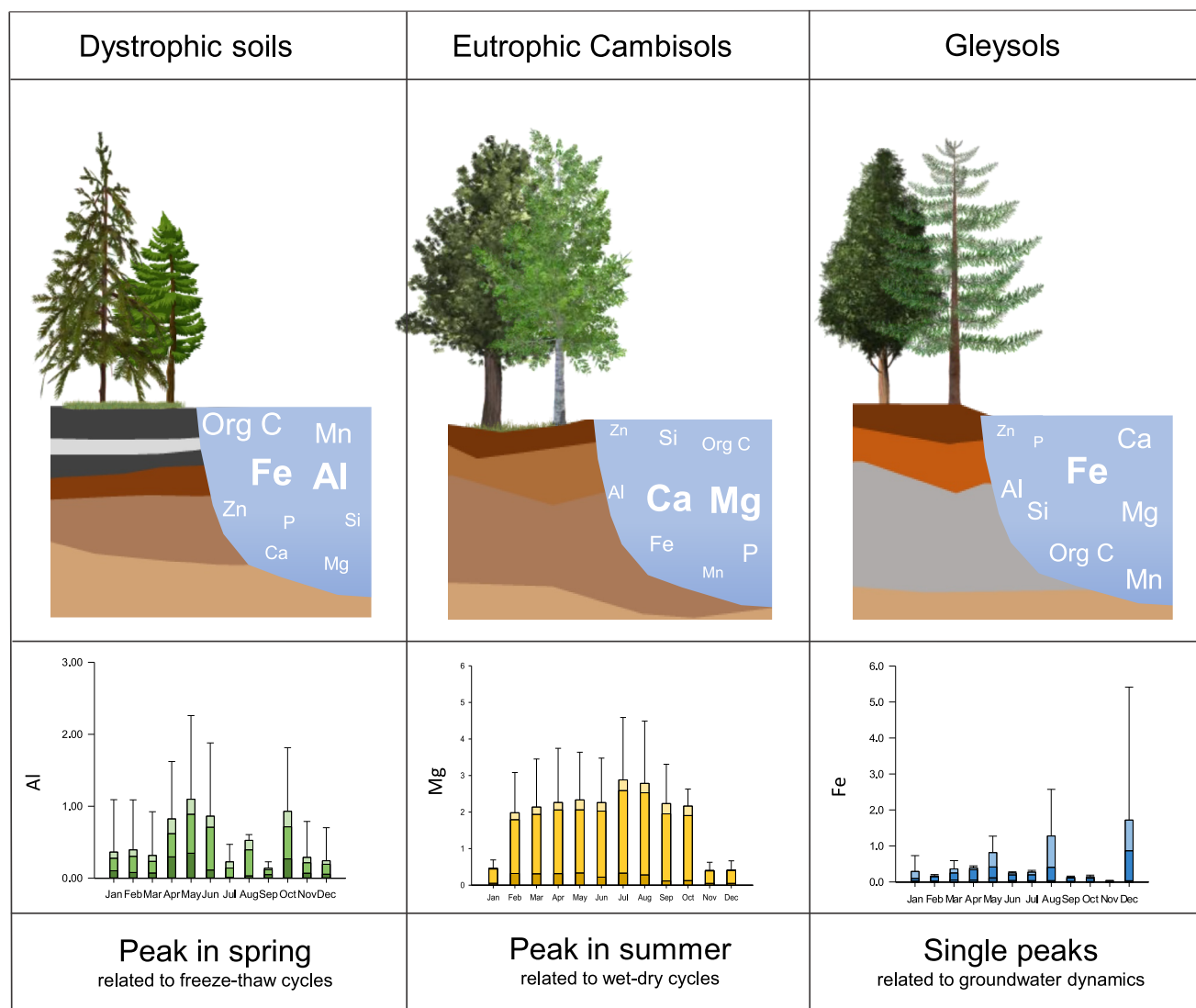


Figure 5. Conceptual model of seasonal colloidal discharge of the three different soil groups. Font size and thickness indicate the dominant element in the stream water. The stacked bar charts show the monthly colloidal discharge of one dominant element per soil group as an example.

and Fe than the first fraction. It is likely that it is not only particle size or mass that play a decisive role in element binding but also the composition and chemical properties of the colloids. In contrast, the total concentrations of P showed less clear relations to individual P fractions, likely reflecting the variable bonding forms of P to, for example, oxides under acidic conditions or the Ca-bearing materials under elevated pH values.

We found that colloidal discharge showed a seasonal pattern for terrestrial soils, whereas Gleysols showed season-independent events with high colloidal discharge (Figure 5). More research is now needed to support or falsify these results for a larger number of sampling sites per soil group; the number was limited in our study, particularly for eutrophic soils and Gleysols, with two sites sampled each. Additionally, the monthly sampling intervals may not have captured short-term (i.e., daily to weekly) hydrological and temperature fluctuations (Siebers et al., 2023) and high rainfall events (Burger et al., 2021). Despite these limitations, our data strongly support earlier hints on the influence of soil horizons, pH and parent material on colloidal export (Gottselig, Amelung, et al., 2017; Missong, Bol, et al., 2018; Missong, et al., 2025). We conclude that the concentration and contribution of colloids in stream waters differ not only between seasons but also within soil groups.

Gottselig, Amelung, et al. (2017) performed a one-time sampling from the streams studied here. The authors reported positive relationships between total and colloidal concentrations of Fe, Al, and org C but scattered correlations for P, Si, and Ca. These trends were confirmed here, suggesting a ubiquitous coupling or decoupling of the respective element cycles, respectively. However, Gottselig, Amelung, et al. (2017) also found a scattered relationship between total and colloidal element concentrations for Mn, while we identified a positive correlation ($r = 0.65$, $p < 0.001$) (Table S1 in Supporting Information S2). We also found lower proportions of colloidal-bound elements, which differed by a factor of 1.5–2.4 relative to the study of Gottselig, Amelung, et al. (2017), except for Ca and Si. For P, the difference was a factor of 10 (see also Gu et al., 2020). Gu et al. (2020) sampled stream water during the wet season, which might have led to elevated colloidal concentrations (Oliveira et al., 2019). Hence, the timing of sampling seems to have a significant effect on element concentrations, presumably reflecting a season-dependent release from soil.

4.2. Controlling Factors for Nanoparticle and Colloidal Abundance in Stream Water

As the release of colloids from soils depends on temperature and precipitation (Gu et al., 2018; C. Jiang et al., 2013), we assumed that the presence of colloids in stream water also depends on weather conditions. Kjaergaard, Moldrup, et al. (2004), Kjaergaard, Poulsen, et al. (2004) found that colloids are increasingly released from soils under wet conditions when elements can be leached from the soil. Thus, elevated precipitation can promote the release of colloids from soils (Oliveira et al., 2019), whereas dry conditions may favor cementation and particle binding (Gu et al., 2018). In the present study, however, there was no significant correlation between colloidal element concentrations and monthly precipitation. Also, temperature had a minor effect on the prevalence of colloids in stream water; only weak positive correlations with MAT were found for Mg and Ca (explaining up to 13% of data variability) and a negative correlation for Zn, explaining up to 28% of data variability. Consequently, we assume that additional factors overlay the effect of precipitation and temperature on colloidal release into forest streams.

As the speciation of elements is pH-dependent, we assumed that the pH, as an ecosystem property, plays an important role in the binding of different elements to colloids (see also Gottselig, Amelung, et al., 2017). We could partly support this assumption as colloidal concentrations of Fe, Mn, and Zn were significantly higher for acidic sites, while significantly higher concentrations of Ca were found for eutrophic sites. However, the majority of acidic sites are located in the north. These northern forests are located in cooler climates with acidic soils covered by organic layers, which may promote the release of soil elements by organic complexation reactions. The southern sites were dominated by calcareous soils (Gottselig, Amelung, et al., 2017). However, we did not find a clear north–south trend for Si, Mn, and P. Therefore, we concur with Gottselig, Amelung, et al. (2017) that pH was rather a surrogate for other covariates affecting colloid release and transport, such as soil group, parent material and climatic conditions.

Soil colloids are assumed to be a major source of colloids in stream water, because the composition of stream water colloids is often similar to that of adjacent soils (Gu et al., 2018; Missong, Holzmann, et al., 2018). The prevalence of such soil colloids is highly variable and differs between different soils, and even between soil horizons (Huang et al., 2016; Missong, Holzmann, et al., 2018; Séquaris et al., 2013) depending on their physical properties (e.g., moisture and temperature) and chemical properties (such as pH and ionic strength) (C. Jiang et al., 2013; Klitzke and Lang, 2009; Tsao et al., 2011). For instance, soils with high OM contents promote colloid dispersion and transport due to increased surface charge and the formation of organo-mineral complexes (Missong, Bol, et al., 2018). Clay-rich soils may enhance colloid retention and stabilization due to aggregation (Braun et al., 2025), while acidic, sandy soils may produce fewer aggregates. Additionally, the presence of macropores can enhance the leaching of colloids also in fine-textured soils (Julich et al., 2016). It seemed reasonable to assume that the elemental composition of stream colloids will also vary when these streams have different adjacent soils. Our results confirm this assumption, as the elemental composition of stream colloids differed significantly for streams surrounded by different soils, each showing a specific seasonality of elements.

4.3. Elemental Composition and Seasonality of Colloids Affected by Soil

We identified the categorized soil group as the main controlling factor for the elemental composition in stream water, as these mainly reflect the respective soil properties. For example, streams of eutrophic soils had significantly more Ca colloids than those in acidic sites, where soils were decalcified and hence Ca-poor. In such acidic

sites, dystrophic soils in forests are characterized by a low pH, enrichment in sesquioxides and an organic surface layer (Konrad et al., 2021). There, Fe, Mn, Al, and org C play an important role in colloid formation and are particularly enriched in stream water, especially from the organic layer and also from the sesquioxide-enriched subsoil (Liang et al., 2010; Wang et al., 2020). Accordingly, higher colloidal concentrations of Al and Fe are found in streams adjacent to acidic soils. Interestingly, the streams near Gleysols also show higher concentrations of Ca colloids than acidic sites. Here, the parent material, which contains calcareous components (both the flysch (Alpthal) and the loess (Frankenforst) parent material), presumably delivers Ca colloids to the stream (Gottselig et al., 2020). In streams near these Gleysols, the clustering of elements even mirrored the element cycling of the respective soils, as all elements except Zn correlated with the redox-sensitive element Fe. In such groundwater-affected soils, Fe oxides, which are an important sorbent for elements, are dissolved when reducing conditions occur in the groundwater-affected subsoil, which promotes the release of both Fe and oxide-associated elements, such as colloidal C. Our data are in line with laboratory studies with basaltic soils, where the reduction of Fe oxides increased the C mobilization by the formation of C colloids (Buettner et al., 2014). Similarly, in paddy soils under anoxic conditions, an up to five-fold increased dispersal of water-extractable colloids was observed (Said-Pullicino et al., 2021), and proportions of colloidal Fe increased with increased duration of management (X. Jiang et al., 2023), which underlines the important role of Fe oxides and anoxic conditions for colloidal release from soils. In summary, our data provide strong evidence that the adjacent soils are the major source of colloids in stream water. With soil groups varying across Europe, stream water colloid composition also varies as a function of soil colloid composition.

The seasonality of elemental discharge was soil-specific. In streams near dystrophic soils, many elements had significantly higher concentrations in spring than in the other seasons, as illustrated for org C (Figure 4). In concordance, Gottselig et al. (2020) and Yesavage et al. (2012) also found peaks of colloidal Fe export in spring. It is noteworthy that two of the three sites with dystrophic soils were located in Northern Europe and experienced snow melt and high water tables in spring. Burger et al. (2021) had already noted that snowmelt affects the colloidal load of streams, reporting that colloidal org C export from a dystrophic Cambisol was related to snowmelt rather than to precipitation. Furthermore, Krickov et al. (2019) found that the occurrence of terrestrial colloids in Siberian rivers was largest in spring after snowmelt and permafrost thaw. Even if one site with dystrophic soil (Lourizela) did not have snow, this site had its highest precipitation rates in April (Table S2 in Supporting Information S2) and likely experienced similar mobilization by high precipitation. Also, thawing soils can favor rapid nutrient release as freeze-thaw cycles disrupt soil aggregates (Siebers et al., 2018), while reduced plant uptake in late winter facilitates their export to streams (Decker et al., 2003). Furthermore, the low electrolyte concentrations in the melt water favor the expansion of the electrical coupler layer, which promotes the dispersion of colloidal particles (Amelung et al., 2018). In summer, by contrast, significantly less colloidal org C was found in the streams, which is probably a result of enhanced microbial activity and decomposition of organic matter.

The enhanced microbial activity and resulting decomposition of SOM under warmer temperatures also results in the release of its adsorbed elements (Kirschbaum, 2006). For instance, Ca^{2+} cations showed a positive correlation with temperature, as mentioned above, and were elevated from spring to autumn in all sampled sites (Figures 2 and 3). Therefore, particularly in the northern sites, Ca likely originates from the organic layer of the forest soils (Wang et al., 2020). In the southern sites, it is likely that elevated chemical weathering rates with elevated temperatures also contributed to the formation of Ca colloids (Zhang et al., 2021).

In streams adjacent to eutrophic Cambisols, most elements showed a positive correlation with monthly precipitation, together with the highest colloid concentrations during summer. When elevated temperatures enhance the release of elements (Eiriksdottir et al., 2011; Kirschbaum, 2006; Zimmermann et al., 2009), less but heavy rainfall in summer (Christensen & Christensen, 2004; Lind et al., 2016) facilitates their leaching and runoff via surface flow, which was particularly shown for org C (Gottselig et al., 2020). In fine-textured soils, repeated drying can lead to the formation of macropores due to shrinkage. These macropores promote preferential flow during re-wetting, which enhances colloid mobilization and export (Julich et al., 2016). Furthermore, repeated re-wetting for dried soil leads to disaggregation (Gu et al., 2018) and a flush of nutrient release by elevated microbial activity (Franzuebbers et al., 1994; Gordon et al., 2008; Missong et al., 2025). Hence, the summer peaks of selected elements such as Ca resulted from a combination of both enhanced organic matter decomposition and climate-induced nutrient flushes (Gottselig et al., 2020; Yesavage et al., 2012). However, some elements, like Zn, did not peak in summer and autumn but in the winter months. As Zn is an important and often deficient micronutrient (Noulas et al., 2018), most of the Zn released during the summer months is presumably taken up

during the growing season and consequently does not reach the streams. In contrast, with higher soil moisture supply in winter, acquisition by plants is at a minimum, while Zn, which is not taken up by plants, is likely released into soil solution by redox reactions.

Unlike the other soils, there was no seasonal pattern in colloidal element concentration for the Gleysols, just single events. Gleysols have more frequent redox cycles because they are highly influenced by changes in groundwater (Amelung et al., 2018). Groundwater was recently shown to be a main determinant of colloid composition and concentration in stream water (Gottselig et al., 2020), so it is likely that, in Gleysols, this effect is accelerated. Besides, Gleysols are also poor in soil structure (Amelung et al., 2018) and hence are more prone to riverbank erosion. Riverbank erosion enhances the movement of colloids from soil to stream and is indicated by high levels of Si and Al in the third fraction, as they indicate the occurrence of phyllosilicates in the stream (Gottselig, Amelung, et al., 2017). Consequently, the groundwater dependency and the soil structure of Gleysols strongly influenced the seasonal behavior of colloids in the stream water and differentiated the colloidal composition in the streams from that of other soils.

5. Conclusion

Natural nanoparticles and colloids play important roles in element cycling in forested ecosystems. We aimed to assess the seasonal behavior of colloids in stream water and the controlling factors and showed that natural nanoparticles and colloids bind up to 90% of elements in forest stream waters across Europe. Our data identified pH as an important driver, as the total element concentrations differed between acidic and eutrophic sites with acidic sites having significantly higher colloidal concentrations of Fe, Mn, and Zn and significantly lower Ca concentrations. Importantly, we found that colloidal export varies substantially between soil groups, improving our understanding of elemental transport at the catchment scale. This finding emphasizes the importance of considering soil properties when analyzing the export of elements in streams, particularly in colloidal size ranges. Future studies should expand on this work by exploring shorter-term hydrological variability, including additional soil groups and regions, in order to better capture the full complexity of colloid-mediated transport in forest headwater catchments.

Data Availability Statement

ICP-MS and OCD data and R scripts for the two-way ANOVA with repeated measures as well as the generalized linear mixed model are archived on Zenodo (Schimmel et al., 2025) [Data set and Software]. [10.5281/zenodo.14639137](https://doi.org/10.5281/zenodo.14639137).

References

- Amelung, W., Blume, H.-P., Fleige, H., Horn, R., Kandeler, E., Kögel-Knabner, I., et al. (2018). Scheffer/Schachtschabel Lehrbuch der Bodenkunde, 17, überarbeitete und ergänzte Auflage. In *Lehrbuch*. Springer Spektrum. <https://doi.org/10.1007/978-3-662-55871-3>
- Baalousha, M., Lead, J. R., von der Kammer, F., & Hofmann, T. (2009). Natural colloids and nanoparticles in aquatic and terrestrial environments. In J. R. Lead & E. Smith (Eds.), *Environmental and human health impacts of nanotechnology* (pp. 109–161). John Wiley & Sons, Ltd. <https://doi.org/10.1002/9781444307504.ch4>
- Baken, S., Regelink, I. C., Comans, R. N. J., Smolders, E., & Koopmans, G. F. (2016). Iron-rich colloids as carriers of phosphorus in streams: A field-flow fractionation study. *Water Research*, 99, 83–90. <https://doi.org/10.1016/j.watres.2016.04.060>
- Bates, D., Mächler, M., Bolker, B., & Walker, S. (2015). Fitting linear mixed-effects models using lme4. *Journal of Statistical Software*, 67(1). <https://doi.org/10.18637/jss.v067.i01>
- Binkley, D., Ice, G. G., Kaye, J., & Williams, C. A. (2004). Nitrogen and phosphorus concentrations in forest streams of the United States. *Journal of the American Water Resources Association*, 40(5), 1277–1291. <https://doi.org/10.1111/j.1752-1688.2004.tb01586.x>
- Braun, M., Juraschek, L., Siebers, N., & Kruse, J. (2025). Natural colloids in soil: Effect of storage and extraction conditions on colloid amount and composition. *Journal of Plant Nutrition and Soil Science*, 202400341. <https://doi.org/10.1002/jpln.202400341>
- Buettner, S. W., Kramer, M. G., Chadwick, O. A., & Thompson, A. (2014). Mobilization of colloidal carbon during iron reduction in basaltic soils. *Geoderma*, 221–222, 139–145. <https://doi.org/10.1016/j.geoderma.2014.01.012>
- Burger, D. J., Vogel, J., Kooijman, A. M., Bol, R., de Rijke, E., Schoorl, J., et al. (2021). Colloidal catchment response to snowmelt and precipitation events differs in a forested headwater catchment. *Vadose Zone Journal*, 20(3). <https://doi.org/10.1002/vzj2.20126>
- Christensen, O. B., & Christensen, J. H. (2004). Intensification of extreme European summer precipitation in a warmer climate. *Global and Planetary Change*, 44(1–4), 107–117. <https://doi.org/10.1016/j.gloplacha.2004.06.013>
- Conant, R. T., Ryan, M. G., Ågren, G. I., Birge, H. E., Davidson, E. A., Eliasson, P. E., et al. (2011). Temperature and soil organic matter decomposition rates—Synthesis of current knowledge and a way forward. *Global Change Biology*, 17(11), 3392–3404. <https://doi.org/10.1111/j.1365-2486.2011.02496.x>
- Decker, K. L. M., Wang, D., Waite, C., & Scherbatskoy, T. (2003). Snow removal and ambient air temperature effects on forest soil temperatures in Northern Vermont. *Soil Science Society of America Journal*, 67(4), 1234–1242. <https://doi.org/10.2136/sssaj2003.1234>

Acknowledgments

This study was funded by the German Research Foundation (DFG, Grant GO 2899/1-1). We gratefully thank all site operators performing the sampling and organizing the sample transport. We thank Manfred Stähli and the staff of the Eidg. Forschungsanstalt WSL for the sampling of the Alpthal sampling sites. We also thank Sónia Gouveia for providing statistical support. The Krycklan infrastructure is funded by the National Research Council via the Swedish Infrastructure for Ecosystem Science (SITES, Grant 2017-00635). Open Access funding enabled and organized by Projekt DEAL.

- Eiriksdottir, E. S., Gislason, S. R., & Oelkers, E. H. (2011). Does runoff or temperature control chemical weathering rates? *Applied Geochemistry*, 26, S346–S349. <https://doi.org/10.1016/j.apgeochem.2011.03.056>
- Espinosa, M., Turner, B. L., & Haygarth, P. M. (1999). Preconcentration and separation of trace phosphorus compounds in soil leachate. *Journal of Environmental Quality*, 28(5), 1497–1504. <https://doi.org/10.2134/jeq1999.00472425002800050015x>
- Fox, J., & Weisberg, S. (2019). An R companion to applied regression, third edition.
- Franzluebbers, K., Weaver, R. W., Juo, A. S. R., & Franzluebbers, A. J. (1994). Carbon and nitrogen mineralization from cowpea plants part decomposing in moist and in repeatedly dried and wetted soil. *Soil Biology and Biochemistry*, 26(10), 1379–1387. [https://doi.org/10.1016/0038-0717\(94\)90221-6](https://doi.org/10.1016/0038-0717(94)90221-6)
- Gordon, H., Haygarth, P. M., & Bardgett, R. D. (2008). Drying and rewetting effects on soil microbial community composition and nutrient leaching. *Soil Biology and Biochemistry*, 40(2), 302–311. <https://doi.org/10.1016/j.soilbio.2007.08.008>
- Gottselig, N., Amelung, W., Kirchner, J. W., Bol, R., Eugster, W., Granger, S. J., et al. (2017). Elemental composition of natural nanoparticles and fine colloids in European forest stream waters and their role as phosphorus carriers: Colloids in European forest streams. *Global Biogeochemical Cycles*, 31(10), 1592–1607. <https://doi.org/10.1002/2017GB005657>
- Gottselig, N., Bol, R., Nischwitz, V., Vereecken, H., Amelung, W., & Klumpp, E. (2014). Distribution of phosphorus-containing fine colloids and nanoparticles in stream water of a forest catchment. *Vadose Zone Journal*, 13(7), 1–11. <https://doi.org/10.2136/vzj2014.01.0005>
- Gottselig, N., Nischwitz, V., Meyn, T., Amelung, W., Bol, R., Halle, C., et al. (2017). Phosphorus binding to nanoparticles and colloids in forest stream waters. *Vadose Zone Journal*, 16(3), 1–12. <https://doi.org/10.2136/vzj2016.07.0064>
- Gottselig, N., Sohr, J., Uhlig, D., Nischwitz, V., Weiler, M., & Amelung, W. (2020). Groundwater controls on colloidal transport in forest stream waters. *Science of the Total Environment*, 717, 134638. <https://doi.org/10.1016/j.scitotenv.2019.134638>
- Gu, S., Gruau, G., Dupas, R., & Jeanneau, L. (2020). Evidence of colloids as important phosphorus carriers in natural soil and stream waters in an agricultural catchment. *Journal of Environmental Quality*, 49(4), 921–932. <https://doi.org/10.1002/jeq2.20090>
- Gu, S., Gruau, G., Malique, F., Dupas, R., Petitjean, P., & Gascuel-Oudou, C. (2018). Drying/rewetting cycles stimulate release of colloidal-bound phosphorus in riparian soils. *Geoderma*, 321, 32–41. <https://doi.org/10.1016/j.geoderma.2018.01.015>
- Hill, D. M., & Aplin, A. C. (2001). Role of colloids and fine particles in the transport of metals in rivers draining carbonate and silicate terrains. *Limnology & Oceanography*, 46(2), 331–344. <https://doi.org/10.4319/lo.2001.46.2.0331>
- Huang, L.-M., Zhang, X.-H., Shao, M.-A., Rossiter, D., & Zhang, G.-L. (2016). Pedogenesis significantly decreases the stability of water-dispersible soil colloids in a humid tropical region. *Geoderma*, 274, 45–53. <https://doi.org/10.1016/j.geoderma.2016.04.005>
- Jarvie, H. P., Neal, C., Rowland, A. P., Neal, M., Morris, P. N., Lead, J. R., et al. (2012). Role of riverine colloids in macronutrient and metal partitioning and transport, along an upland–lowland land-use continuum, under low-flow conditions. *Science of the Total Environment*, 434, 171–185. <https://doi.org/10.1016/j.scitotenv.2011.11.061>
- Jiang, C., Séquaris, J.-M., Vereecken, H., & Klumpp, E. (2013). Diffusion-controlled mobilization of water-dispersible colloids from three German silt loam topsoils: Effect of temperature: Diffusion-controlled mobilization of soil colloids. *European Journal of Soil Science*, 64, 777–786. <https://doi.org/10.1111/ejss.12086>
- Jiang, X., Bol, R., Nischwitz, V., Siebers, N., Willbold, S., Vereecken, H., et al. (2015). Phosphorus containing water dispersible nanoparticles in arable soil. *Journal of Environmental Quality*, 44(6), 1772–1781. <https://doi.org/10.2134/jeq2015.02.0085>
- Jiang, X., Wulf, A., Bol, R., & Klumpp, E. (2023). Phosphorus content in water extractable soil colloids over a 2000 years chronosequence of paddy-rice management in the Yangtze River Delta, China. *Geoderma*, 430, 116296. <https://doi.org/10.1016/j.geoderma.2022.116296>
- Julich, D., Julich, S., & Feger, K.-H. (2016). Phosphorus in preferential flow pathways of forest soils in Germany. *Forests*, 8(1), 19. <https://doi.org/10.3390/f8010019>
- Kirschbaum, M. (2006). The temperature dependence of organic-matter decomposition—Still a topic of debate. *Soil Biology and Biochemistry*, 38(9), 2510–2518. <https://doi.org/10.1016/j.soilbio.2006.01.030>
- Kjaergaard, C., Moldrup, P., de Jonge, L. W., & Jacobsen, O. H. (2004). Colloid mobilization and transport in undisturbed soil columns. II. The role of colloid dispersibility and preferential flow. *Vadose Zone Journal*, 3(2), 424–433. <https://doi.org/10.2136/vzj2004.0424>
- Kjaergaard, C., Poulsen, T. G., Moldrup, P., & de Jonge, L. W. (2004). Colloid mobilization and transport in undisturbed soil columns. I. Pore structure characterization and tritium transport. *Vadose Zone Journal*, 3(2), 413–423. <https://doi.org/10.2136/vzj2004.0413>
- Klitzke, S., & Lang, F. (2009). Mobilization of soluble and dispersible lead, arsenic, and antimony in a polluted, organic-rich soil—Effects of pH increase and counterion valency. *Journal of Environmental Quality*, 38(3), 933–939. <https://doi.org/10.2134/jeq2008.0239>
- Konrad, A., Billiy, B., Regenbogen, P., Bol, R., Lang, F., Klumpp, E., & Siemens, J. (2021). Forest soil colloids enhance delivery of phosphorus into a diffusive gradient in thin films (DGT) sink. *Frontiers in Forests and Global Change*, 3, 577364. <https://doi.org/10.3389/ffgc.2020.577364>
- Krickov, I. V., Pokrovsky, O. S., Manasypov, R. M., Lim, A. G., Shirokova, L. S., & Viers, J. (2019). Colloidal transport of carbon and metals by western Siberian rivers during different seasons across a permafrost gradient. *Geochimica et Cosmochimica Acta*, 265, 221–241. <https://doi.org/10.1016/j.gca.2019.08.041>
- Kuznetsova, A., Brockhoff, P. B., & Christensen, R. H. B. (2017). lmerTest package: Tests in linear mixed effects models. *Journal of Statistical Software*, 82(13). <https://doi.org/10.18637/jss.v082.i13>
- Laudon, H., Hasselquist, E. M., Peichl, M., Lindgren, K., Sponseller, R., Lidman, F., et al. (2021). Northern landscapes in transition: Evidence, approach and ways forward using the Krycklan Catchment Study. *Hydrological Processes*, 35(4), e14170. <https://doi.org/10.1002/hyp.14170>
- Lenth, R. (2023). _emmeans: Estimated marginal means, aka least-squares means. *R package version*, 1(8.5).
- Liang, X., Liu, J., Chen, Y., Li, H., Ye, Y., Nie, Z., et al. (2010). Effect of pH on the release of soil colloidal phosphorus. *Journal of Soils and Sediments*, 10(8), 1548–1556. <https://doi.org/10.1007/s11368-010-0275-6>
- Lind, P., Lindstedt, D., Kjellström, E., & Jones, C. (2016). Spatial and temporal characteristics of summer precipitation over central Europe in a suite of high-resolution climate models. *Journal of Climate*, 29, 3501–3518. <https://doi.org/10.1175/JCLI-D-15-0463.1>
- Missong, A., Bol, R., Nischwitz, V., Krüger, J., Lang, F., Siemens, J., & Klumpp, E. (2018). Phosphorus in water dispersible-colloids of forest soil profiles. *Plant and Soil*, 427(1–2), 71–86. <https://doi.org/10.1007/s11104-017-3430-7>
- Missong, A., Fetzer, J., Bol, R., Hagedorn, F., Winkler, P., Wang, L., et al. (2025). Leaching of colloidal phosphorus from organic layers and mineral topsoils of German beech forests in response to drought and nutrient inputs. *Journal of Soil Science and Plant Nutrition*, 25(1), 562–575. <https://doi.org/10.1007/s42729-024-02152-7>
- Missong, A., Holzmann, S., Bol, R., Nischwitz, V., Puhlmann, H., v. Wilpert, K., et al. (2018). Leaching of natural colloids from forest topsoils and their relevance for phosphorus mobility. *Science of the Total Environment*, 634, 305–315. <https://doi.org/10.1016/j.scitotenv.2018.03.265>
- Moradi, G., Bol, R., Trbojevic, L., Missong, A., Mörchen, R., Fuentes, B., et al. (2020). Contrasting depth distribution of colloid-associated phosphorus in the active and abandoned sections of an alluvial fan in a hyper-arid region of the Atacama Desert. *Global and Planetary Change*, 185, 103090. <https://doi.org/10.1016/j.gloplacha.2019.103090>

- Noulas, C., Tziouvalekas, M., & Karyotis, T. (2018). Zinc in soils, water and food crops. *Journal of Trace Elements in Medicine & Biology*, 49, 252–260. <https://doi.org/10.1016/j.jtemb.2018.02.009>
- Oliveira, M. L. S., Saikia, B. K., da Boit, K., Pinto, D., Tutikian, B. F., & Silva, L. F. O. (2019). River dynamics and nanoparticles formation: A comprehensive study on the nanoparticle geochemistry of suspended sediments in the Magdalena river, Caribbean industrial area. *Journal of Cleaner Production*, 213, 819–824. <https://doi.org/10.1016/j.jclepro.2018.12.230>
- R Core Team. (2022). *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. Retrieved from <https://www.R-project.org/>
- Regelink, I. C., Stoof, C. R., Rousseva, S., Weng, L., Lair, G. J., Kram, P., et al. (2015). Linkages between aggregate formation, porosity and soil chemical properties. *Geoderma*, 247–248, 24–37. <https://doi.org/10.1016/j.geoderma.2015.01.022>
- Said-Pullicino, D., Giannetta, B., Demeglio, B., Missong, A., Gottselig, N., Romani, M., et al. (2021). Redox-driven changes in water-dispersible colloids and their role in carbon cycling in hydromorphic soils. *Geoderma*, 385, 114894. <https://doi.org/10.1016/j.geoderma.2020.114894>
- Schimmel, H., Amelung, W., & Braun, M. (2025). Data: Natural nanoparticles and colloids in forested streams across Europe: Seasonal patterns and impact of soil groups [Dataset and software]. *Zenodo*. <https://doi.org/10.5281/ZENODO.14639137>
- Séguaris, J.-M., Klumpp, E., & Vereecken, H. (2013). Colloidal properties and potential release of water-dispersible colloids in an agricultural soil depth profile. *Geoderma*, 193–194, 94–101. <https://doi.org/10.1016/j.geoderma.2012.10.014>
- Siebers, N., Abdelrahman, H., Krause, L., & Amelung, W. (2018). Bias in aggregate geometry and properties after disintegration and drying procedures. *Geoderma*, 313, 163–171. <https://doi.org/10.1016/j.geoderma.2017.10.028>
- Siebers, N., Kruse, J., Jia, Y., Lennartz, B., & Koch, S. (2023). Loss of subsurface particulate and truly dissolved phosphorus during various flow conditions along a tile drain–ditch–brook continuum. *Science of the Total Environment*, 866, 161439. <https://doi.org/10.1016/j.scitotenv.2023.161439>
- Tsao, T. M., Chen, Y. M., & Wang, M. K. (2011). Origin, separation and identification of environmental nanoparticles: A review. *Journal of Environmental Monitoring*, 13(5), 1156. <https://doi.org/10.1039/c1em10013k>
- Wang, L., Missong, A., Amelung, W., Willbold, S., Prietzel, J., & Klumpp, E. (2020). Dissolved and colloidal phosphorus affect P cycling in calcareous forest soils. *Geoderma*, 375, 114507. <https://doi.org/10.1016/j.geoderma.2020.114507>
- Wickham, H., Averick, M., Bryan, J., Chang, W., McGowan, L., François, R., et al. (2019). Welcome to the tidyverse. *Journal of Open Source Software*, 4(43), 1686. <https://doi.org/10.21105/joss.01686>
- Wilkinson, K. J. & Lead, J. R. (Eds.) (2006). *Environmental colloids and particles: Behaviour, separation and characterisation*. John Wiley & Sons, Ltd. <https://doi.org/10.1002/9780470024539>
- Yesavage, T., Fantle, M. S., Vervoort, J., Mathur, R., Jin, L., Liermann, L. J., & Brantley, S. L. (2012). Fe cycling in the shale hills critical zone observatory, Pennsylvania: An analysis of biogeochemical weathering and Fe isotope fractionation. *Geochimica et Cosmochimica Acta*, 99, 18–38. <https://doi.org/10.1016/j.gca.2012.09.029>
- Zhang, Y., Yu, S., He, S., Sun, P., Wu, F., Liu, Z., et al. (2021). New estimate of chemical weathering rate in Xijiang River Basin based on multi-model. *Scientific Reports*, 11(1), 5728. <https://doi.org/10.1038/s41598-021-84602-1>
- Zimmermann, M., Meir, P., Bird, M. I., Malhi, Y., & Ccahuana, A. J. Q. (2009). Climate dependence of heterotrophic soil respiration from a soil-translocation experiment along a 3000 m tropical forest altitudinal gradient. *European Journal of Soil Science*, 60(6), 895–906. <https://doi.org/10.1111/j.1365-2389.2009.01175.x>