Biogeosciences, 13, 5567–5585, 2016
www.biogeosciences.net/13/5567/2016/
doi:10.5194/bg-13-5567-2016

Biogeosciences
Open Access
EGU

Trends in soil solution dissolved organic carbon (DOC) concentrations across European forests

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Received: 9 December 2015 – Published in Biogeosciences Discuss.: 26 January 2016
Revised: 13 September 2016 – Accepted: 15 September 2016 – Published: 7 October 2016

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract. Dissolved organic carbon (DOC) in surface waters is connected to DOC in soil solution through hydrological pathways. Therefore, it is expected that long-term dynamics of DOC in surface waters reflect DOC trends in soil solution. However, a multitude of site studies have failed so far to establish consistent trends in soil solution DOC, whereas increasing concentrations in European surface waters over the past decades appear to be the norm, possibly as a result of recovery from acidification. The objectives of this study were therefore to understand the long-term trends of soil solution DOC from a large number of European forests (ICP Forests Level II plots) and determine their main physicochemical and biological controls. We applied trend analysis at two levels: (1) to the entire European dataset and (2) to the individual time series and related trends with plot characteristics, i.e., soil and vegetation properties, soil solution chemistry and atmospheric deposition loads. Analyses of the entire dataset showed an overall increasing trend in DOC concentrations in the organic layers, but, at individual plots and depths, there was no clear overall trend in soil solution DOC. The rate change in soil solution DOC ranged between -16.8 and $+23\,\%\,\mathrm{yr}^{-1}$ (median $= +0.4\,\%\,\mathrm{yr}^{-1}$) across Europe. The non-significant trends (40 %) outnumbered the increasing (35 %) and decreasing trends (25 %) across the 97 ICP Forests Level II sites. By means of multivariate statistics, we found increasing trends in DOC concentrations with increasing mean nitrate (NO_3^-) deposition and increasing trends in DOC concentrations with decreasing mean sulfate (SO_4^{2-}) deposition, with the magnitude of these relationships depending on plot deposition history. While the attribution of increasing trends in DOC to the reduction of SO_4^{2-} deposition could be confirmed in low to medium N deposition areas, in agreement with observations in surface waters, this was not the case in high N deposition areas. In conclusion, long-term trends of soil solution DOC reflected the interactions between controls acting at local (soil and vegetation properties) and regional (atmospheric deposition of SO_4^{2-} and inorganic N) scales.

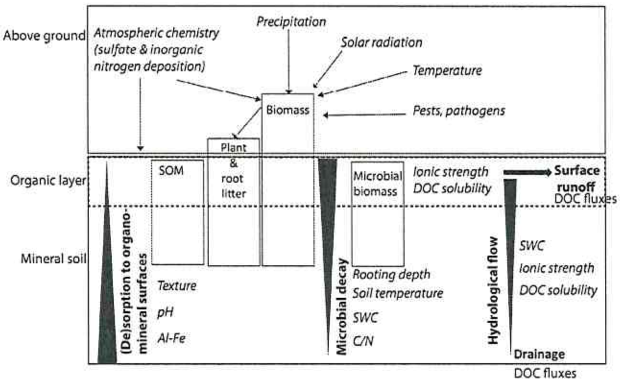


Figure 1. Schematic diagram illustrating the main sources (in boxes) of dissolved organic carbon (DOC) and the main processes (in bold) and factors (in italics) controlling DOC concentrations in soils.

1 Introduction

Dissolved organic carbon (DOC) in soil solution is the source of much of the terrestrially derived DOC in surface waters (Battin et al., 2009; Bianchi, 2011; Regnier et al., 2013). Soil solution DOC in forests is connected to streams through different hydrological pathways: DOC mobilized in the forest floor may be transported laterally at the interface of forest floor and mineral soil to surface waters or percolates into the mineral soil, where additional DOC can be mobilized and/or DOC is partly adsorbed on particle surfaces and mineralized thereafter (Fig. 1). From the mineral soil DOC may be leached either laterally or vertically via groundwater into surface waters (McDowell and Likens, 1988). Therefore, it could be expected that long-term dynamics of DOC in surface waters mirror those observed in ecosystem soil solutions.

Drivers related to climate change (temperature increase, precipitation change, atmospheric CO_2 increase), the decrease in acidifying deposition, or land use change and management may individually or jointly explain trends in surface water DOC concentrations (Evans et al., 2012; Freeman et al., 2004; Oulehle et al., 2011; Sarkkola et al., 2009; Worrall and Burt, 2004). Increasing air temperatures warm the soil, thus stimulating soil organic matter (SOM) decomposition through greater microbial activity (Davidson and Janssens, 2006; Hartley and Ineson, 2008; Kalbitz et al., 2000). Other drivers, such as increased atmospheric CO_2 and the accumulation of atmospherically deposited inorganic nitrogen, are thought to increase the sources of DOC by enhancing primary plant productivity (i.e., through stimulating root exudates or increased litterfall) (de Vries et al., 2014; Ferretti et al., 2014; Sucker and Krause, 2010). Changes in precipitation, land use and management (e.g. drainage of peatlands, changes in forest management or grazing systems) may alter the flux of DOC leaving the ecosystem, but no consistent trends in the hydrologic regime or land use changes have been detected in areas where increasing DOC trends have been observed (Monteith et al., 2007).

Recent focus has mainly been on decreasing acidifying deposition as an explanatory factor for DOC increases in surface waters in Europe and North America by means of decreasing ionic strength (de Wit et al., 2007; Hruška et al., 2009) and increasing the pH of soil solution, consequently increasing DOC solubility (Evans et al., 2005; Haaland et al., 2010; Monteith et al., 2007). Although the hypothesis of an increase in surface water DOC concentration due to a recovery from past acidification was confirmed in studies of soil solution DOC in the UK and northern Belgium (Sawicka et al., 2016; Vanguelova et al., 2010; Verstraeten et al., 2014), it is not consistent with trends in soil solution DOC concentrations reported from Finnish, Norwegian, and Swedish forests (Löfgren and Zetterberg, 2011; Ukonmaanaho et al., 2014; Wu et al., 2010). This inconsistency between soil solution DOC and stream DOC trends could suggest that DOC in surface water and soil solution responds differently to (changes in) environmental conditions in different regions (Akselsson et al., 2013; Clark et al., 2010; Löfgren et al., 2010). Alternatively, other factors such as tree species and soil type, may be co-drivers of organic matter dynamics and input, generation and retention of DOC in soils.

Trends of soil solution DOC vary among not only forests but often also within the same site (Borken et al., 2011; Löfgren et al., 2010). Forest characteristics such as tree species composition, soil fertility, texture or sorption capacity may affect the response of soil solution DOC to environmental controls, for instance, by controlling the rate of soil acidification through soil buffering and nutrient plant uptake processes (Vanguelova et al., 2010). Within a site, DOC variability with soil depth is typically caused by different intensity of DOC production, transformation, and sorption along the soil profile (Fig. 1). Positive temporal trends in soil solution DOC (increasing concentrations over time) have frequently been reported for the organic layers and shallow soils where production and decomposition processes control the DOC concentration (Löfgren and Zetterberg, 2011). However, no dominant trends are found for the mineral soil horizons, where physico-chemical processes, such as sorption, become more influential (Borken et al., 2011; Buckingham et al., 2008). Furthermore, previous studies have used different temporal and spatial scales which may have further added to the inconsistency in the DOC trends reported in the literature (Clark et al., 2010).

In this context, the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests, 2010) compiled a unique dataset containing data from more than 100 intensively monitored forest plots (Level II) which allow for regional trends in soil solution DOC of forests at a European scale to be unraveled, as well as for statistical analysis of the main controls behind these regional trends to be performed. Long-term measurements of soil solution DOC are available for these plots, along with information on aboveground biomass, soil properties, and atmospheric deposition of inorganic N and SO_4^{2-}, collected using a harmonized sampling protocol across Europe (Ferretti and Fischer, 2013). This dataset has previously been used to investigate the spatial variability of DOC in forests at European scale (Camino-Serrano et al., 2014), but an assessment of the temporal trends in soil solution DOC using this large dataset has not been attempted so far.

The main objective of this study is to understand the long-term temporal trends of DOC concentrations in soil solution measured at the ICP Forests Level II plots across Europe. Based on the increasing DOC trends in surface waters, we hypothesize that temporal trends in soil solution DOC will also be positive, but with trends varying locally depending on plot characteristics. We further investigated whether plot characteristics, specifically climate, inorganic N and SO_4^{2-} deposition loads, forest type, soil properties, and changes in soil solution chemistry can explain differences across sites in DOC trends.

2 Materials and methods

2.1 Data description

Soil solution chemistry has been monitored within the ICP Forests Programme since the 1990s on most Level II plots. The ICP Forests data were extracted from the pan-European Forest Monitoring Database (Granke, 2013). A list of the Level II plots used for this study can be found in the Supplement, Table S1. The methods for collection and analysis of soil solution used in the various countries (Switzerland: Graf Pannatier et al., 2011; Flanders, Belgium: Verstraeten et al., 2012; Finland: Lindroos et al., 2000; UK: Vanguelova et al., 2010, Denmark: Hansen et al., 2007) follow the ICP Forests manual (Nieminen, 2011). Generally, lysimeters were installed at several fixed depths starting at 0 cm, defined as the interface between the surface organic layer and underlying mineral soil. These depths are typically aligned with soil "organic layer", "mineral topsoil", "mineral subsoil", and "deeper mineral soil", but sampling depths vary among countries and even among plots within a country. Normally, zero-tension lysimeters were installed under the surface organic layer and tension lysimeters within the mineral soil. However, in some countries zero-tension lysimeters were also used within the mineral layers and in some tension lysimeters below the organic layer. Multiple collectors (replicates) were installed per plot and per depth to assess plots' spatial variability. However, in some countries, samples from these replicates were pooled before analyses or averaged prior to data transmission. The quality assurance and control procedures included the use of control charts for internal reference material to check long-term comparability within national laboratories as well as participation in periodic laboratory ring tests (e.g., Marchetto et al., 2011) to check the international comparability. Data were reported annually to the pan-European data center, checked for consistency and stored in the pan-European Forest Monitoring Database (Granke, 2013).

Soil water was usually collected fortnightly or monthly, although for some plots sampling periods with sufficient soil water for collection were scarce, especially in prolonged dry periods or in winter due to snow and ice. After collection, the samples were filtered through a 0.45 µm membrane filter, stored below 4 °C and then analyzed for DOC, together with other soil solution chemical properties (NO_3^-, Ca, Mg, NH_4^+, SO_4^{2-}, total dissolved Al, total dissolved Fe, pH, electrical conductivity). Information on the soil solution chemistry at the studied plots can be found in the Supplement (Tables S4–S11). The precision of DOC analysis differed among the laboratories. The coefficient of variation of repeatedly measured reference material was 3.7 % on average. The time span of soil solution time series used for this study ranged from 1991 to 2011, although coverage of this period varied from plot to plot (Table S1).

Soil properties; open field bulk deposition; and throughfall deposition of NO_3^-, NH_4^+, and SO_4^{2-} are measured at the same plots as well as stem volume increment. The atmospheric deposition of NO_3^-, NH_4^+ and SO_4^{2-} data covers the period 1999–2010 (Waldner et al., 2014). Stem volume growth was calculated by the ICP Forests network from diameter at breast height (DBH), live tree status, and tree height which were assessed for every tree ($DBH > 5$ cm) within a monitoring plot approximately every 5 years since the early 1990s. Tree stem volumes were derived from allometric relationships based on diameter and height measurements according to De Vries et al. (2003), accounting for species and regional differences. Stem volume growth (in cubic meters) between two consecutive inventories was calculated as the difference between stem volumes at the beginning and the end of one inventory period for living trees. Stem volume data were corrected for all trees that were lost during one inventory period, including thinning. Stem volume at the time of disappearance (assumed at half of the time of the inventory period) was estimated from functions relating stem volume of standing living trees at the end of the period vs. volume at the beginning of the period. The methods used for collection of these data can be found in the manuals of the ICP Forests Monitoring Programme (ICP Forests, 2010). The soil properties at the plots used for this study were derived from the ICP Forests aggregated soil database (AFSCDB.LII.2.1) (Cools and De Vos, 2014).

Since continuous precipitation measurements are not commonly available for the Level II plots, precipitation measurements for the location of the plots were extracted from the observational station data of the European Climate Assessment & Dataset (ECA&D) and the ENSEMBLES Observations (E-OBS) gridded dataset (Haylock et al., 2008). We used precipitation measurements extracted from the E-OBS gridded dataset to improve the temporal and spatial coverage and to reduce methodological differences of precipitation measurements across the plots. The E-OBS dataset contains daily values of precipitation and temperature from stations data gridded at 0.25° resolution. When E-OBS data were not available, they were gap-filled with ICP Forests precipitation values gained by deposition measurements where available.

2.2 Data preparation

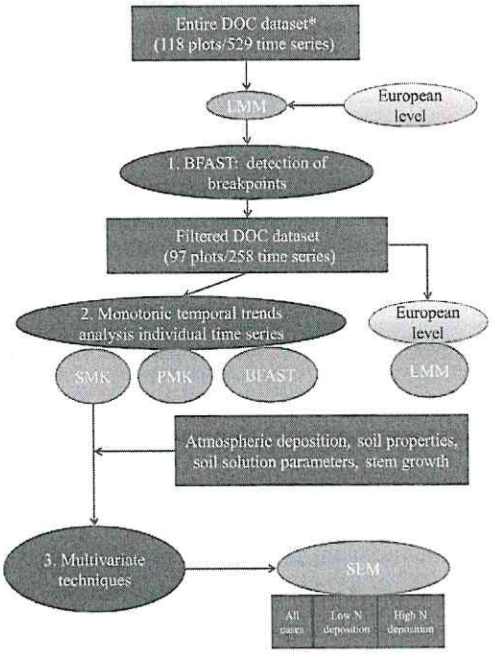
We extracted data from plots with time series covering more than 10 years and including more than 60 observations of soil solution DOC concentrations of individual or groups of collectors. Outliers, defined as ± 3 interquartile range of the 25 and 75 % quantiles of the time series, were removed from each time series to avoid the influence of a few extreme values in the long-term trend (Schwertman et al., 2004). Values under $1\,mg\,L^{-1}$, which is the detection limit for DOC in the ICP Level II plots, were replaced by $1\,mg\,L^{-1}$. After this filtering, 529 time series from 118 plots, spanning from Italy to Norway, were available for analysis. Soil solution, precipitation, and temperature were aggregated to monthly data by the median of the observations in each month and by the sum of daily values in the case of precipitation. Data of inorganic N (NH_4^+ and NO_3^-) and SO_4^{2-} throughfall and open field bulk deposition measured at the plots were interpolated to monthly data (Waldner et al., 2014).

The plots were classified according to their forest (broadleaved/coniferous-dominated) and soil type (World Reference Base (WRB), 2006), their stem growth (slow, $< 6\,m^3\,ha^{-1}\,yr^{-1}$; intermediate, $6–12\,m^3\,ha^{-1}\,yr^{-1}$; and fast, $> 12\,m^3\,ha^{-1}\,yr^{-1}$), and their soil solution pH (low, < 4.2; intermediate, 4.2–5; high, > 5). Plots were also classified based on mean throughfall inorganic N ($NO_3^- + NH_4^+$) deposition level, defined as high deposition (HD, $> 15\,kg\,N\,ha^{-1}\,yr^{-1}$), medium deposition (MD, $5–15\,kg\,N\,ha^{-1}\,yr^{-1}$), and low deposition (LD, $< 5\,kg\,N\,ha^{-1}\,yr^{-1}$), as well as mean throughfall SO_4^{2-} deposition level, defined as high deposition (HD, $> 6\,kg\,S\,ha^{-1}\,yr^{-1}$), and low deposition (LD, $< 6\,kg\,S\,ha^{-1}\,yr^{-1}$).

2.3 Statistical methods

Time series can typically be decomposed into random noise, seasonal, and trend components (Verbesselt et al., 2010). In this paper, we used methods to detect the actual trend (change in time) after removing the seasonal and random noise components. The sequence of methods applied is summarized in Fig. 2. The analysis of temporal trends in soil solution DOC concentrations was carried out at two levels: (1) the European level and (2) the plot level. While the first analysis allows an evaluation of the overall trend in soil solution DOC at a continental scale, the second analysis indicates whether the observed large-scale trends are occurring at local scales as well, and tests whether local trends in DOC can be attributed to certain driver variables.

Linear mixed-effects models (LMMs) were used to detect the temporal trends in soil solution DOC concentration at European scale (Fig. 2). For these models, the selected 529 time series were used. For the trend analysis of individual time series, however, we focused on the long-term trends in soil solution DOC at European forests that show monotonicity. Therefore, DOC time series were first analyzed using the Breaks For Additive Seasonal and Trend (BFAST) algorithm to detect the presence of breakpoints (Verbesselt et al., 2010; Vicca et al., 2016), with the time series showing breakpoints, i.e., not monotonic, being discarded (see "Description of the statistical methods" in the Supplement). In total, 258 mono-



* Time series > 10 years and > 60 obs.

Abbreviation	Model	Type of analysis
LMM	Linear mixed-effects models	Temporal trends
BFAST	Breaks For Additive Seasonal and Trend	Analysis of breakpoints in time series
SMK	Seasonal Mann-Kendall test	Monotonic temporal trends
PMK	Partial Mann-Kendall test	Monotonic temporal trends
SEM	Structural equation model	Multivariate analysis (direct/indirect effects)

Figure 2. Flow-diagram of the sequence of methods applied for analysis of temporal trends of soil solution DOC and their drivers.

tonic time series from 97 plots were used for our analysis after filtering (Fig. 2). Then, monotonic trend analyses were carried out from the filtered dataset using the seasonal Mann–Kendall (SMK) test for monthly DOC concentrations (Hirsch et al., 1982; Marchetto et al., 2013). Partial Mann–Kendall (PMK) tests were also used to test the influence of precipitation as a co-variable to detect whether the trend might be due to a DOC dilution/concentration effect (Libiseller and Grimvall, 2002). Sen (1968) slope values were calculated for SMK and PMK. Moreover, LMMs were performed again with the filtered dataset to compare results with and without time series showing breakpoints (Fig. 2).

For this study, five soil depth intervals were considered: the organic layer (0 cm), topsoil (0–20 cm), intermediate (20–40 cm), subsoil (40–80 cm) and deep subsoil (> 80 cm). The slopes of each time series were standardized by dividing them by the median DOC concentration over the sampling period (relative trend slope), aggregated to a unique plot–soil depth slope and classified by the direction of the trend as significantly positive, i.e., increasing DOC over time (P, $p < 0.05$); significantly negative, i.e., decreasing DOC over time (N, $p < 0.05$); and non-significant, i.e., no significant change in DOC over time (NS, $p \geq 0.05$). When there was more than one collector per depth interval, the median of the slopes was used when the direction of the trend (P, N, or NS) was similar. After aggregation per plot–depth combination, 191 trend slopes from 97 plots were available for analysis (Table S2). Trends for other soil solution parameters (NO_3^-, Ca^{2+}, Mg^{2+}, NH_4^+, SO_4^{2-}, total dissolved Al, total dissolved Fe, pH, electrical conductivity), precipitation and temperature were calculated using the same methodology as for DOC. Since the resulting standardized Sen slope in $\% \, yr^{-1}$ (relative trend slope) was used for all the statistical analyses, from here on we will use the general term "trend slope" in order to simplify.

Finally, structural equation models (SEMs) were performed to determine the capacity of the several factors (SO_4^{2-} and/or NO_3^- deposition, stem growth and soil solution chemistry) in explaining variability in the slope of DOC trends among the selected plots (Fig. 2). We evaluated the influence of both the annual mean ($kg \, ha^{-1} \, yr^{-1}$) and the trends ($\% \, yr^{-1}$) in deposition and soil solution parameters. All the statistical analyses were performed in R software version 3.1.2 (R Core Team, 2014) using the "rkt" (Marchetto et al., 2013), "bfast01" (de Jong et al., 2013) and "sem" (Fox et al., 2013) packages, except for the LMMs that were performed using SAS 9.3 (SAS institute, Inc., Cary, NC, USA). More detailed information on the statistical methods used can be found in the Supplement.

3 Results

3.1 Soil solution DOC trends at European scale

First, temporal trends in DOC were analyzed for all the European DOC data pooled together by means of LMMs to test for the presence of overall trends. A significantly increasing DOC trend ($p < 0.05$) in soil solution collected with zero-tension lysimeters in the organic layer was observed mainly under coniferous forest plots (Table 1). Similarly, a significantly increasing DOC trend ($p < 0.05$) in soil solution collected with tension lysimeters was found in deep mineral soil (> 80 cm) for all sites, mainly for coniferous forest sites (Table 1), but this trend is based on a limited number of plots which are not especially well distributed in Europe (75 % of German plots). By contrast, non-significant trends were found in the other mineral soil depth intervals (0–20, 20–40 and 40–80 cm) by means of the LMMs. When the same analysis was applied to the filtered European dataset, i.e., without the time series showing breakpoints, fewer significant trends were observed: only an overall positive trend ($p < 0.05$) was found for DOC in the organic layer using zero-tension lysimeters, again mainly under coniferous forest sites, but no statistically significant trends were found in the mineral soil (Table 1).

3.2 Soil solution DOC concentration trend analysis of individual time series

We applied the BFAST analysis to select the monotonic time series in order to ensure that the detected trends were not influenced by breakpoints in the time series. Time series with breakpoints represented more than 50 % of the total time series aggregated by soil depth interval (245 out of 436).

The individual trend analysis using the SMK test showed trend slopes of soil solution DOC concentration ranging from -16.8 to $+23\,\%\,\mathrm{yr}^{-1}$ (median $= +0.4\,\%\,\mathrm{yr}^{-1}$, interquartile range $= +4.3\,\%\,\mathrm{yr}^{-1}$). Among all the time series analyzed, the non-statistically significant trends (40 %, 104 time series) outnumbered the significantly positive trends (35 %, 91 time series) and significantly negative trends (24 %, 63 time series) (Table 1). Thus, there was no uniform trend in soil solution DOC in forests across a large part of Europe. Furthermore, the regional trend differences were inconsistent when looking at different soil depth intervals separately (Figs. 3 and 4), which made it difficult to draw firm conclusions about the spatial pattern of the trends in soil solution DOC concentrations in European forests.

The variability in trends was high, not only at continental scale but also at plot level (Fig. 5). We found consistent within-plot trends only for 50 out of the 97 sites. Moreover, some plots even showed different trends (P, N or NS) in DOC within the same depth interval, which was the case for 17 plot–depth combinations (16 in Germany and 1 in Norway), evidencing a high small-scale plot heterogeneity.

Trend directions (P, N or NS) often differed among depths. For instance, in the organic layer, we found mainly non-significant trends, and if a trend was detected, it was more often positive than negative, while positive trends were the most frequent in the subsoil (below 40 cm) (Table 1). Nevertheless, it is important to note that a statistical test of whether there was a real difference in DOC trends between depths was not possible as the set of plots differed between the different soil depth intervals. However, a visual comparison of trends for the few plots in which trends were evaluated for more than three soil depths showed that there was no apparent difference in DOC trends between soil depths (Figs. S1 and S2).

Finally, for virtually all plots, including precipitation as a co-variable in the PMK test gave the same result as the SMK test, which indicates that precipitation (through dilution or concentration effects) did not affect the DOC concentration trends. A dilution/concentration effect was only detected in four plots (Table S1).

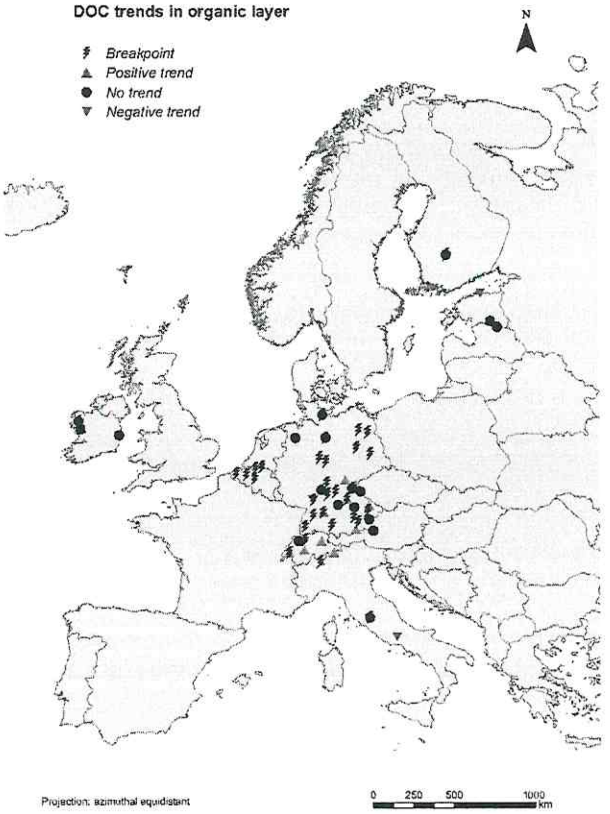


Figure 3. Directions of the temporal trends in soil solution DOC concentration in the organic layer at plot level. Trends were evaluated using the seasonal Mann–Kendall test. Data span from 1991 to 2011.

3.3 Factors explaining the soil solution DOC trends

3.3.1 Effects of vegetation, soil and climate

There was no direct effect of forest type (broadleaved vs. coniferous) on the direction of the statistically significant trends in soil solution DOC (Fig. 6a). Both positive and negative trends were equally found under broadleaved and coniferous forests (χ^2 $(1, n = 97) = 0.073$, $p = 0.8$). Increasing DOC trends, however, occurred more often under forests with a mean stem growth increment below $6\,\mathrm{m}^3\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$ over the study period, whereas decreasing DOC trends were more common in forests with a mean stem growth increment between 6 and $12\,\mathrm{m}^3\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$ (χ^2 $(2, n = 53) = 5.8$, $p = 0.05$) (Fig. 6b). Only six forests with a mean stem growth above $12\,\mathrm{m}^3\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$ were available for this study (five showing increasing DOC trends and one showing a decreasing DOC trend) and thus there is not enough information to draw conclusions about the relationship between stem growth and soil solution DOC trends for forests with very high stem growth ($> 12\,\mathrm{m}^3\,\mathrm{ha}^{-1}\,\mathrm{yr}^{-1}$).