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

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ICP Forests Level II plots data used for the trend analysis

Table S1. List of ICP Forests Level II plots used for the trend analysis and their dominant forest species and resulting trend calculated using the Seasonal Mann-Kendall test (NS; non-significant, P: positive, N: negative). Rows in green correspond to the plots where at least one time series has been used for the individual trend analysis after filtering out the breakpoints. Rows in red correspond to the plots with measurements of DOC in soil solution that have not been used for the individual trend analysis because there was not enough data (Lack data) or breakpoints were detected (BP). Collector types are tension lysimeters (TL) or zero-tension lysimeters (ZTL).

Country	Code plot	Start year	End year	Collector type	Tree species	Trend	Dilution effect
France	1_6	1998	2011	TL	*Quercus robur*	NS	
France	1_17	1998	2011	TL	*Quercus petraea*	NS	
France	1_30	1998	2011	TL	*Quercus petraea*	N	
France	1_37	1998	2011	TL	*Picea abies*	NS	
France	1_41	1998	2011	TL	*Picea abies*	N	
France	1_46	1998	2011	TL	*Picea abies*	NS/N	
France	1_57	1998	2011	ZTL	*Fagus sylvatica*	P/NS	
France	1_63	1998	2011	TL	*Fagus sylvatica*	NS/N	
France	1_84	1998	2011	TL	*Pinus sylvestris*	N	
France	1_90	1998	2011	TL	*Abies alba*	NS/P	depth= -0.2, coll=1
France	1_93	1998	2011	TL	*Abies alba*	NS	
France	1_96	1998	2011	TL	*Abies alba*	P/NS	
France	1_98	1998	2011	TL	*Abies alba*	NS	
France	1_100	1998	2011	TL	*Abies alba*	NS	
Belgium	2_1	2000	2005		*Picea abies*	Lack data	
Belgium	2_8				*Quercus petraea*	Lack data	
Belgium	2_11	1999	2011	ZTL/TL	*Fagus sylvatica*	P	
Belgium	2_14	1999	2011	ZTL/TL	*Pinus nigra*	NS/P	
Belgium	2_15	1999	2011	ZTL/TL	*Pinus sylvestris*	NS/P	
Belgium	2_16	1999	2011	ZTL/TL	*Quercus robur*	NS	

Belgium	2_21	1999	2011	ZTL/TL	*Fagus sylvatica*	P	
Germany	4_101	1996	2011	TL	*Fagus sylvatica*	NS/N	
Germany	4_301	1997	2011	TL	*Fagus sylvatica*	NS	
Germany	4_302	1997	2011		*Picea abies*	BP	
Germany	4_303	1998	2011	TL	*Picea abies*	N	
Germany	4_304	1998	2011	TL	*Fagus sylvatica*	N	
Germany	4_305	1998	2011		*Picea abies*	BP	
Germany	4_306	1996	2011	TL	*Fagus sylvatica*	P	
Germany	4_307	1996	2011	TL	*Pinus sylvestris*	NS/P	depth=-2.5, coll=3
Germany	4_308	1993	2011	TL	*Quercus robur*	N	
Germany	4_502	1998	2011	TL	*Quercus robur*	N/NS	
Germany	4_503	1997	2011		*Fagus sylvatica*	BP	
Germany	4_506	1997	2011	TL	*Picea abies*	NS	
Germany	4_603	1998	2005		*Fagus sylvatica*	Lack data	
Germany	4_604	1998	2001		*Fagus sylvatica*	Lack data	
Germany	4_605	1998	2005		*Fagus sylvatica*	Lack data	
Germany	4_606	1996	2011	TL	*Fagus sylvatica*	NS	
Germany	4_607	1998	2010		*Fagus sylvatica*	Lack data	
Germany	4_701	1996	2011	TL	*Picea abies*	Weight_	
Germany	4_702	1996	2011	TL	*Picea abies*		
Germany	4_703	1996	2011	TL	*Fagus sylvatica*	NS/P	
Germany	4_704	1996	2011	TL	*Fagus sylvatica*	Weight_P	
Germany	4_705	1996	2011	TL	*Quercus petraea*	N/Weight_N	
Germany	4_706	1996	2011	TL	*Quercus robur*	P/Weight_P	
Germany	4_707	1996	2011	TL	*Pinus sylvestris*	P	
Germany	4_802	1997	2011	TL	*Picea abies*	N	
Germany	4_806	1997	2011	TL	*Picea abies*	P	
Germany	4_808	1997	2011	TL	*Picea abies*	N/NS	
Germany	4_809	1997	2010	TL	*Picea abies*	N/NS	

Germany	4_812	1997	2011	TL	*Picea abies*	P/N/Weight_N	
Germany	4_901	1996	2011	ZTL/TL	*Pinus sylvestris*	P/N	
Germany	4_902	1996	2011	ZTL/TL	*Picea abies*	NS	
Germany	4_903	1998	2011	ZTL/TL	*Fagus sylvatica*	P	
Germany	4_904	1996	2011	ZTL/TL	*Larix decidua*	NS	
Germany	4_905	1996	2011	ZTL/TL	*Pinus sylvestris*	P/NS	
Germany	4_906	1996	2011	ZTL/TL	*Picea abies*	NS/P	
Germany	4_907	1996	2006		*Fagus sylvatica*	Lack data/BP	
Germany	4_908	1996	2011	ZTL/TL	*Picea abies*	NS/N	
Germany	4_909	1996	2011	ZTL/TL	*Picea abies*	NS/Weight_P/P	depth=-1.2, coll=15
Germany	4_910	1996	2006		*Quercus robur*	Lack data/BP	
Germany	4_911	1996	2011	ZTL/TL	*Fagus sylvatica*	P/Weight_P	
Germany	4_912	1996	2006		*Pinus sylvestris*	Lack data/BP	
Germany	4_913	1996	2011	ZTL/TL	*Quercus petraea*	NS	
Germany	4_914	1996	2011	ZTL/TL	*Quercus petraea*	NS	
Germany	4_915	1996	2006		*Fagus sylvatica*	Lack data	
Germany	4_916	1996	2006		*Picea abies*	Lack data	
Germany	4_917	1996	2006		*Picea abies*	Lack data	
Germany	4_918	1996	2006		*Pinus sylvestris*	Lack data	
Germany	4_919	1996	2011	ZTL/TL	*Fagus sylvatica*	N/P/NS	
Germany	4_920	1998	2011	ZTL/TL	*Picea abies*	P	
Germany	4_921	1997	2011	ZTL/TL	*Quercus petraea*	P/Weight_P	
Germany	4_922	1997	2011	ZTL/TL	*Picea abies*	P/N	depth=-0.5, coll=6
Germany	4_1001	1998	2011	TL	*Quercus robur*	P/NS	
Germany	4_1201	2001	2007		*Pinus sylvestris*	Lack data	
Germany	4_1202	2001	2011	TL	*Pinus sylvestris*	NS	
Germany	4_1203	2000	2011		*Pinus sylvestris*	BP	

Germany	4_1204	2000	2011	TL	*Pinus sylvestris*	NS	
Germany	4_1205	2000	2011	TL	*Pinus sylvestris*	NS	
Germany	4_1206	2000	2007		*Pinus sylvestris*	Lack data	
Germany	4_1302	1998	2011	TL	*Fagus sylvatica*	N/P	
Germany	4_1303	1997	2011	TL	*Pinus sylvestris*	NS	
Germany	4_1401	1996	2012	TL	*Picea abies*	NS/P	
Germany	4_1402	1996	2012	TL	*Picea abies*	P	
Germany	4_1403	1996	2012	TL	*Picea abies*	NS/P	
Germany	4_1404	1996	2012	TL	*Picea abies*	NS/P	
Germany	4_1405	1996	2012	TL	*Pinus sylvestris*	NS	
Germany	4_1406	1996	2011	TL	*Quercus petraea*	P	
Germany	4_1501	1998	2011	TL	*Pinus sylvestris*	N/P	
Germany	4_1502	1998	2011	TL	*Pinus sylvestris*	N	
Germany	4_1605	2007	2011		*Picea abies*	Lack data	
Germany	4_1606	2007	2011		*Fagus sylvatica*	Lack data	
Germany	4_1607	2007	2011		*Pinus sylvestris*	Lack data	
Germany	4_1608				*Quercus petraea*	Lack data	
Germany	4_1609				*Abies alba*	Lack data	
Italy	5_1	1999	2011	ZTL	*Fagus sylvatica*	N	
Italy	5_9	1999	2011	ZTL	*Quercus cerris*	NS	
UK	6_512	2004	2011		*Quercus robur*	Lack data	
UK	6_517	2002	2010		*Quercus robur*	Lack data	
UK	6_715	2002	2011	TL	*Pinus sylvestris*	NS	
UK	6_716	2002	2009		*Pinus sylvestris*	Lack data	
UK	6_919	2004	2011		*Picea sichensis*	Lack data	
UK	6_920				*Picea sichensis*	Lack data	

UK	6_922	1997	2011	TL	*Picea sichensis*	P	
Ireland	7_1	1991	2000	ZTL/TL	*Picea sichensis*	P/NS	
Ireland	7_10	1991	2011	ZTL and others/ TL	*Picea sichensis*	NS/P	
Ireland	7_11	1991	2011	ZTL/TL	*Quercus petraea*	N/NS	
Denmark	8_11	1996	2011	TL	*Picea abies*	NS	
Denmark	8_34	1997	2011	TL	*Fagus sylvatica*	NS	
Denmark	8_74	2002	2012		*Fagus sylvatica*	Lack data/BP	
Denmark	8_85	2003	2011		*Quercus robur*	Lack data	
Greece	9_3					Lack data	
Greece	9_4					Lack data	
Sweden	13_1301	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_1403	1996	2006		*Picea abies*	Lack data	
Sweden	13_5201	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_5202	1996	2006		*Picea abies*	Lack data	
Sweden	13_5401	1996	2006		*Picea abies*	Lack data	
Sweden	13_5501	1996	2006		*Picea abies*	Lack data	
Sweden	13_5502	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_5601	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_5602	1996	2006		*Picea abies*	Lack data	
Sweden	13_5603	1996	2006		*Picea abies*	Lack data	
Sweden	13_5701	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_5702	1996	2006		*Picea abies*	Lack data	
Sweden	13_5703	1996	2006		*Picea abies*	Lack data	
Sweden	13_5801	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_6001	1996	2006		*Fagus sylvatica*	Lack data	

Sweden	13_6002	1996	2006		*Quercus robur*	Lack data	
Sweden	13_6003	1996	2006		*Picea abies*	Lack data	
Sweden	13_6102	1996	2006		*Fagus sylvatica*	Lack data	
Sweden	13_6103	1996	2006		*Picea abies*	Lack data	
Sweden	13_6301	2000	2006		*Fagus sylvatica*	Lack data	
Sweden	13_6302	1996	2006		*Picea abies*	Lack data	
Sweden	13_6401	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_6501	1996	2006		*Picea abies*	Lack data	
Sweden	13_6503	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_6507	1996	2006		*Picea abies*	Lack data	
Sweden	13_6601	1996	2006		*Picea abies*	Lack data	
Sweden	13_6702	1996	2006		*Picea abies*	Lack data	
Sweden	13_6703	1996	2006		*Picea abies*	Lack data	
Sweden	13_6802	1996	2006		*Picea abies*	Lack data	
Sweden	13_6803	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_6901	1996	2006		*Picea abies*	Lack data	
Sweden	13_7402	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_7404	1996	2006		*Picea abies*	Lack data	
Sweden	13_7501	1996	2006		*Pinus sylvestris*	Lack data	
Sweden	13_7502	1996	2006		*Picea abies*	Lack data	
Austria	14_9	1997	2010	TL	*Fagus sylvatica*	N	
Austria	14_16	2001	2010	TL	*Picea abies*	NS	
Finland	15_1	1998	2011		*Pinus sylvestris*	Lack data	
Finland	15_3	1998	2011		*Picea abies*	Lack data	
Finland	15_5	1997	2011		*Picea abies*	Lack data	
Finland	15_6	1997	2011		*Pinus sylvestris*	Lack data	
Finland	15_11	1997	2011	ZTL	*Picea abies*	NS	

Finland	15_16	1998	2011		*Pinus sylvestris*	Lack data	
Finland	15_17	1998	2011		*Picea abies*	Lack data	
Finland	15_19	1999	2011		*Picea abies*	Lack data	
Finland	15_20	1998	2011		*Pinus sylvestris*	Lack data	
Finland	15_21	2000	2010		*Picea abies*	Lack data	
Finland	15_23	1998	2010		*Picea abies*	Lack data	
Switzerland	50_2	1999	2012	ZTL/TL	*Picea abies*	P	
Switzerland	50_3	1999	2012	Mix collector type one	*Fagus sylvatica*	N/NS	
Switzerland	50_4	1999	2011	ZTL/TL	*Pinus cembra*	NS/P	
Switzerland	50_8	1999	2012	ZTL/TL	*Fagus sylvatica*	NS/P	
Switzerland	50_12	1999	2012	ZTL/TL	*Quercus cerris*	NS	
Switzerland	50_15	1999	2011	ZTL/TL	*Abies alba*	N	
Switzerland	50_16	1999	2012	Mix collector type one	*Fagus sylvatica*	N/P	
Norway	55_1	1996	2011	ZTL/TL	*Picea abies*	NS/N	
Norway	55_9	1996	2011	TL	*Picea abies*	P/Weight_P	
Norway	55_14	1996	2011	TL	*Picea abies*	N	
Norway	55_18	1999	2010	TL	*Pinus sylvestris*	P	
Norway	55_19	1998	2011	TL	*Picea abies*	N	
Czech Republic	58_521	2006	2011		*Picea abies*	Lack data	
Czech Republic	58_2015	2006	2011		*Fagus sylvatica*	Lack data	
Czech Republic	58_2361	2006	2011		*Quercus fruticosa*	Lack data	
Estonia	59_2	1999	2011	ZTL	*Pinus sylvestris*	NS/N	
Estonia	59_3	1999	2011	ZTL	*Pinus sylvestris*	NS	
Estonia	59_7	2002	2011	ZTL	*Pinus sylvestris*	NS	

Description of the statistical methods

1) Overall trend analysis at European scale

Linear mixed-effects models (LMM) were used to detect the temporal trends in soil solution DOC concentrations at the European scale. For these models, the complete ICP Forests Level II dataset was used. Because the dependent variable (DOC concentration) was usually not normally distributed, it was log-transformed to improve normality. Different models were built per depth and per collector type (tension or zero-tension lysimeters). For each model, the variable describing the temporal effect was the year, centered on the year 2000 (year-2000), which was considered as fixed effect. Also, month (1-12) was considered as fixed effect to account for seasonality. Two random factors describing the country ($ctry_{int}$) and plot ($plot_{int}$) effects and one random coefficient accounting for the between plot variation of the temporal effect ($plot_{slp}$) were considered in each LMM (Equation 1). The LMMs were further adjusted by stratification of data according to forest type in order to investigate possible differences in DOC trends between broadleaved and coniferous forests. The models were built following Jonard et al. (2015).

$$logDOC = \left[a + month + ctry_{int}\,(0,\sigma_{ci}^2) + plot_{int}(0,\sigma_{pi}^2)\right] + \left[b + plot_{slp}(0,\sigma_{ps}^2)\,\right] \cdot (year - 2000) + \varepsilon(0,\sigma^2) \quad (1)$$

where σ_{ci}^2, σ_{pi}^2, σ_{ps}^2 and σ^2 are the variances of the random factors 'country' and 'plot', of the random coefficient 'plot' and of the residual term (ε), respectively.

2) Trend analysis of individual time series

Temporal changes in terrestrial ecosystems can either be monotonic changes, or discontinuous with abrupt changes resulting in breakpoints (de Jong et al., 2013). Monotonicity of time series is generally assumed when analyzing DOC data for temporal trends (Filella and Rodriguez-Murillo, 2014). However, it is rarely statistically tested and, thus, potential abrupt changes in the time series may be overlooked. This issue becomes important in temporal trend analysis since a breakpoint may cause changes in the direction of the trend and could lead us, for example, to classify a time series as constant, when in reality we may have averaged out separate periods with significant changes (de Jong et al., 2013). On the other hand, breakpoints may erroneously induce the detection of a significant trend in long-term time series due to artifacts.

For these reasons, we focused on the investigation of the potential 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). When a breakpoint was detected in a time series, there were two possibilities: first, one of the segments (before or after the detected breakpoint) was longer than 9 years, and, in this case, only the longest segment was used for the subsequent analysis of monotonic trends; second, the breakpoint split the time series in two segments shorter than 9 years and then the time series was not used for the analysis of monotonic trends. We used a length threshold of 9 years, which is the minimum time series length recommended for long-term trend analysis (Libiseller and Grimvall, 2002; Waldner et al., 2014). In total, 258 time series from 97 plots were selected for analysis of monotonic trends (Table S2). No clear pattern could be observed in the distribution of time series of DOC with breakpoints, which appeared to occur randomly across the study plots (Figs. 3 and 4).

Monotonic trend analyses were carried out using the Seasonal Mann Kendall (SMK) test for monthly DOC concentrations (Hirsch et al., 1982; Marchetto et al., 2013). Partial Mann Kendall (PMK) test was also used to test the influence of monthly precipitation as a co-variable, i.e., to test if the trend detection might be due to a DOC dilution/concentration effect (Libiseller and Grimvall, 2002). For the SMK and PMK tests, the trend slopes were estimated following Sen (1968), as the median of all the slopes determined by all pairs of sample points. The SMK and PMK account for seasonality of the time series by computing the test on each of the seasons (in our case months) separately. The resulting slopes were also tested against the slopes calculated by BFAST. Finally, the individual slopes calculated according to Sen (1968) for each time series using the SMK or PMK method were standardized by dividing them by the median DOC concentration over the sampling period to avoid the influence of the magnitude of DOC concentration in the between-site comparison. The resulting standardized slopes (relative slopes) were used for the subsequent statistical analysis.

For this study, five 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 then aggregated to a unique slope per depth interval in each plot (hereafter called “plot-soil depth combination”) and classified by the direction of the trend as significantly positive (P, $p < 0.05$), significantly negative (N, $p < 0.05$) and not significant

(NS, $p \geq 0.05$). When there was more than one collector per depth class, the median of the slopes was used when the direction of the trend (P, N or NS) was similar. When the different trends at the same plot-soil depth combination were either P and NS, or N and NS, it was marked as “Weighted positive” and “Weighted negative”. The five plot-soil depth combinations for which the calculated slopes showed opposite trend directions were discarded. All aggregated trend slopes came from time series measured using the same collector type. After aggregation per plot-depth combinations, 191 trend slopes from 97 plots were available for analysis (Table S2).

Trends for soil solution parameters (NO_3^-, Ca, Mg, 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: individual time series were analyzed using the SMK test and the relative slopes were calculated and aggregated to plot-soil depth combinations.

Finally, we performed a multivariate statistical analysis to investigate the main factors explaining differences in DOC trends among the selected plots. We applied Structural Equation Models (SEM) to test whether deposition variables had an effect (direct, indirect or total) on the relative trends slopes of DOC through different pathways (Grace et al., 2010). For the SEMs, we assumed that there is no effect of soil depth on the DOC trends (see next section in Supplementary Material). We applied three SEM models: 1) for all the slopes in DOC, 2) only for the forests with low or medium total N deposition, and, 3) only for the forests with high total N deposition. For each case, we searched for the most parsimonious adequate model using the Akaike information criterion (AIC) and R^2. The significance level (*p* value) of the total, direct and indirect effects were calculated using the bootstrap (with 1200 repetitions) technique (Davison et al., 1986). Dependent variables were log-transformed to improve normality of the continuous variables and then standardized before performing the SEMs. 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).

Table S2. Summary of number of time series used in the study

	Entire dataset	Without breakpoints
All time series	1480 (173 plots)	--

Time series >60 observations and > 10 years	529	258
Aggregated plot-depth combinations	436	191
Plots	118	97

Depth effect on the individual trends in soil solution DOC

Trends in soil solution from different soil depth intervals were mixed for the Pearson's chi-squared test performed for Fig. 5 and the Structural Equation Models (SEM) (Fig. 6), as the number of cases available for each depth are insufficient to compute the statistics if we separate per soil depth interval. To check if the trends calculated at different depths were actually independent from the soil depth interval, we performed a Pearson's chi-squared test and found that the differences in trends among soil depth intervals were not statistically significant $\chi^2(8, N = 174) = 10.94$, $p = 0.21$) (Fig. S1). Therefore, we assumed that there is no difference in trends among soil depth layers and performed the subsequent statistical analysis mixing the trends from different soil depths.

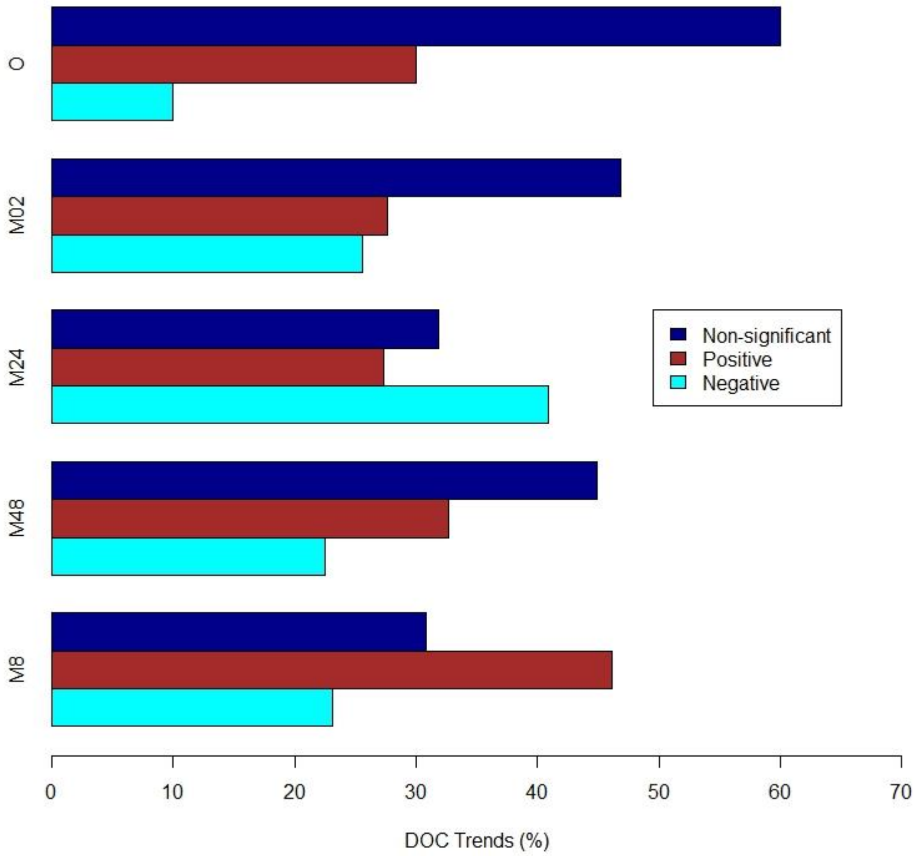

Figure S1. Percentage of non-significant, positive and negative trends per soil depth interval (O: organic layer, M02: mineral soil 0-20 cm, M24: mineral soil 20-40 cm, M48: mineral soil 40-80 cm, M8: mineral soil > 80 cm).

However, a real difference in DOC trends between soil depths may be obscured by the fact that datasets differ between different depths (not all the sites have DOC time series that could be analyzed for trends at all the soil depth intervals) and thus, we cannot rule out that there exists a difference in trends per soil depth. Although the number of sites with DOC trends analyzed at more than three soil depths (including the organic layer) is not enough to apply the same statistics for this subset, we visually compared the 11 sites with this information available and found that, at first sight, it was confirmed that there is no a real difference in trends between soil depth intervals (Fig. S2).

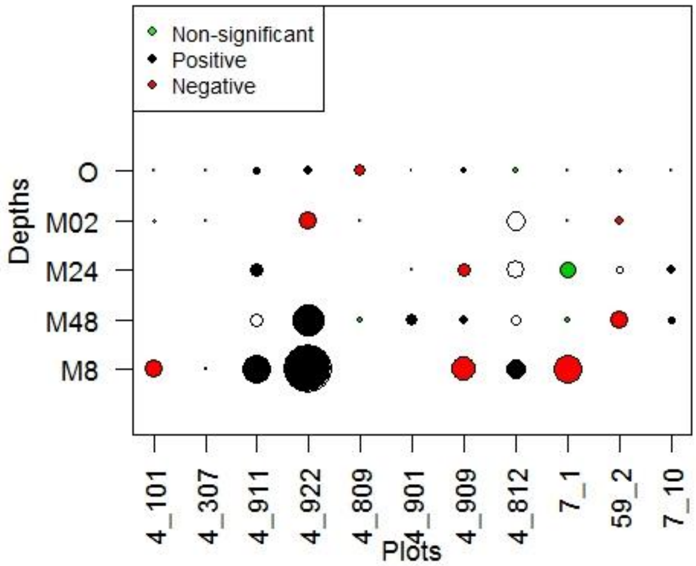

Figure S2. Direction of the trend (non-significant, positive and negative) per soil depth interval (O: organic layer, M02: mineral soil 0-20 cm, M24: mineral soil 20-40 cm, M48: mineral soil 40-80 cm, M8: mineral soil > 80 cm) for the 11 plots with DOC measured at least at 3 soil depth intervals including the organic layer. The size of the circle is proportional to the magnitude of the trend slope.

Structural equation model with trends in SO_4^{2-} and NO_3^- deposition

The same structural equation models (SEM) represented in Fig. 6 were performed using the trends in SO_4^{2-} and NO_3^- deposition (% yr^{-1}) instead of the mean values of SO_4^{2-} and NO_3^- throughfall deposition (kg ha^{-1} yr^{-1}). The SEMs for all the cases and for cases with low and medium inorganic N deposition are shown in Fig. S3.

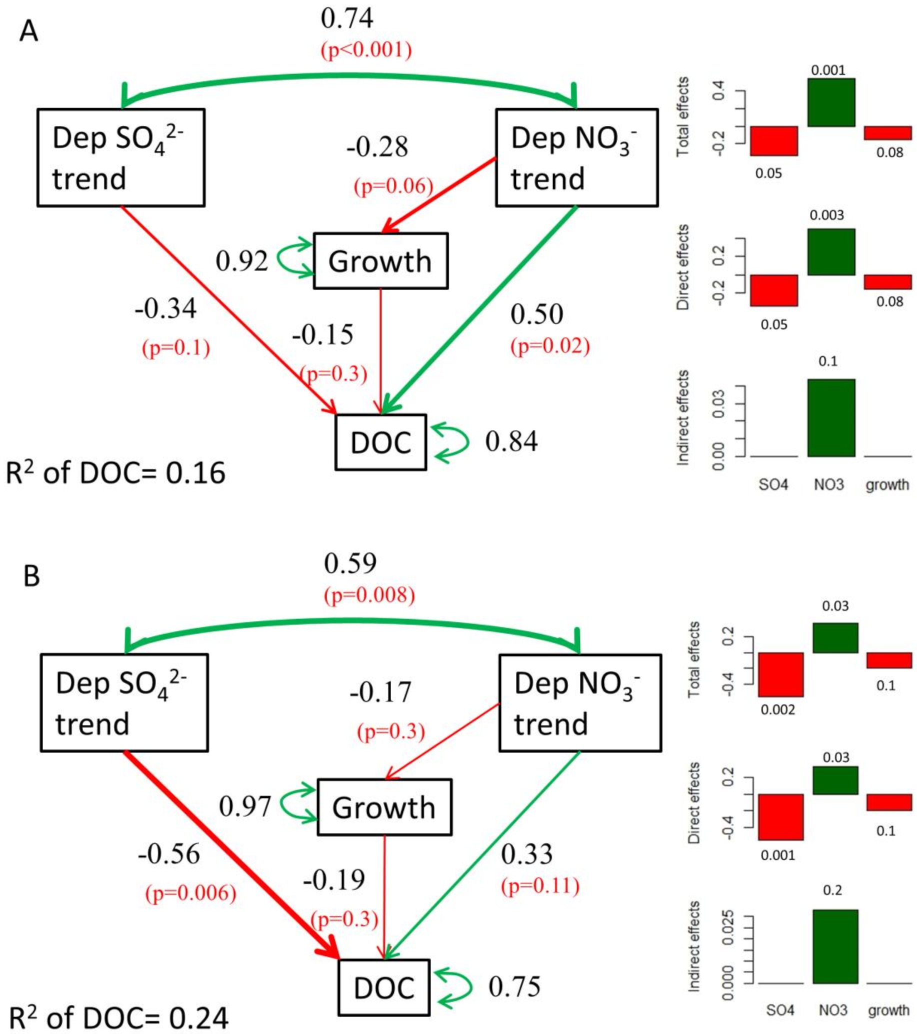

Figure S3. Diagram of the structural equation model (SEM) that best explains the maximum variance of the resulting trends of DOC concentrations in soil solution for: A) all the cases and B) cases with low or medium inorganic N deposition, with trends in SO_4^{2-} and NO_3^- deposition (% yr^{-1}) with direct effects and indirect effects through effects on mean annual stem volume increment (growth) in m^3 ha^{-1} yr^{-1}). P-values of the significance of the corresponding effect between brackets. Green arrows indicate positive effects and red arrows indicate negative effects.

Comparison of methods of individual trend analysis

We applied the BFAST analysis to select the monotonic time series in order to assure that the overall 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). In total, 191 plot-soil depth combinations from 97 plots were analyzed after filtering out the time series showing breakpoints and 94% of the analyzed plot-depth combinations showed consistent trends among replicates collected at the same depth. In contrast, when also considering the time series with breakpoints, the trends calculated for plot-depth combinations agreed only in 75% of the cases implying that the proportion of contradictory trends within plot-depth combinations increased from 6% in the dataset without breakpoints to 25% in the entire dataset (Fig. S4). For both datasets, the majority of the trends were not statistically significant (44% and 41%, for the dataset with and without breakpoints, respectively). In other words, filtering the time series for breakpoints reduced the within-plot variability, while most of the plots showed similar aggregated trends per plot-depth combinations. For this reason, the results discussed in this paper correspond only to the trends of monotonic (breakpoint filtered) time series of soil solution DOC concentrations.

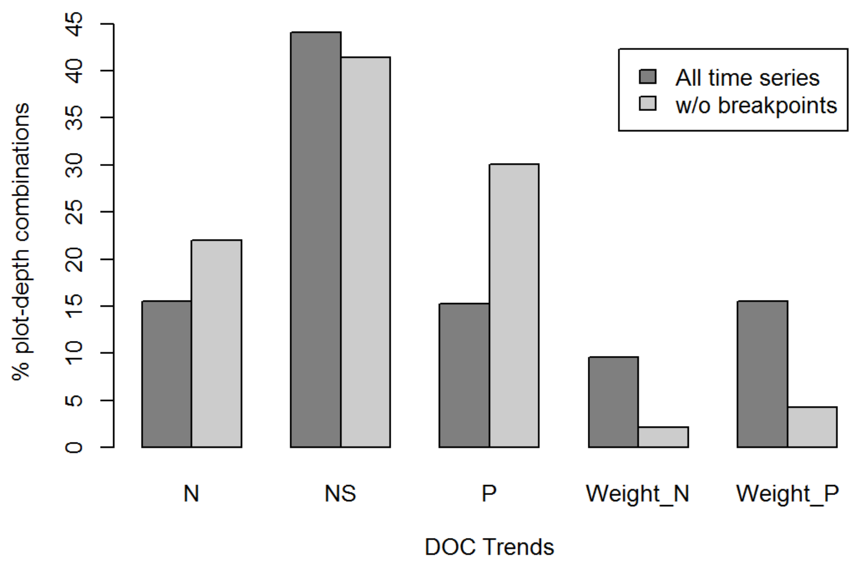

Figure S4. Percentage of plot-soil depth combinations for which negative (N), non-significant (NS), positive (P), negative and non-significant (Weight_N) and positive and non-significant (Weight_P) trends of DOC concentrations were found using SMK (seasonal Mann-Kendall) tests when 1) all the 436 time series were used, 2) only 191 time series without breakpoints (detected using the BFAST (*Breaks For Additive Seasonal and Trend*) analysis) were used.

There was a good agreement between results using the three methods: BFAST, SMK, and PMK. The direction and significance of the trend agreed for 84.5% of the time series analyzed. For the majority of the remaining time series for which the trends did not agree, BFAST did not detect a trend when SMK and PMK did, thus, the latter two methods seemed more sensitive for trend detection than BFAST. Trends computed with SMK and PMK agreed well. The direction of the trend for SMK and PMK only differed for the intermediate soil layer (20-40 cm), as a result of the two extra sites for which SMK tests were performed, but not the PMK, that showed a marked positive trend (1.1 and 2 % yr^{-1}). However, when using exactly the same set of sites, the trend did not differ between the two methods.

Implications of using standardized DOC slopes versus absolute DOC slopes.

The standardized (relative) slopes of DOC concentrations were used for the study of the factors affecting the soil solution DOC trends (Fig. 5 and 6). The main reason for this decision was that using the median DOC concentration as a reference (as we did with the standardization) allowed us to determine whether the absolute trend in DOC was quantitatively large or not from an ecological perspective, because the absolute trend slope will be highly dependent on the initial DOC concentrations of the site.

The absolute trend slopes show the real magnitude and significance of the trend, but do not allow for comparison among sites or horizons. Since the aim of this study is to test whether there is a general DOC trend and to compare sites across Europe, we decided that using the relative slope was more consistent.

Moreover, due to limitations of the statistical analysis, we worked with time series per "plot-soil depth combinations", which means that different soil layers were mixed in the statistical analysis. Again, the standardization of the slopes of DOC concentrations allowed us to compare trends among different soil horizons by removing the effect of the decreasing soil solution DOC concentrations with soil depth. Otherwise, using the absolute trends would introduce a bias when we try to explain the DOC trends in relation with other parameters, because the trend slope would be highly dependent on the actual DOC concentrations, which, in turn, are very variable, not only among sites, but also among soil depths.

The influence of the DOC concentration levels was checked before deciding to use the standardized slopes (Fig. S5). It seemed that there was no relationship between the DOC trend slopes (relative and absolute) and the median DOC concentrations, with positive and negative trends occurring at both low and high DOC concentrations and, thus, we decided that using the standardized slopes will not hide any effect of the median DOC concentrations on the direction of the DOC trends.

This decision, however, has a drawback: the strength of the trend is clearly influenced by the DOC concentration levels. The fact that we used the standardized slope of DOC implied that it may be identical for two sites with very different mean DOC concentrations. DOC concentration decreases with depth and is lower in the deep mineral soil than in the upper mineral soil (Table S3) and by standardizing the slope, the magnitude of the trend was exaggerated in lower soil layers where both the absolute slope of DOC and the median DOC concentration are low (Table S3). This issue is well illustrated in Fig. S5, that shows how the highest standardized slopes are usually at low DOC concentrations (mostly in mineral soil layers), while the highest absolute slopes are at higher DOC concentrations (mostly in organic and upper soil layers).

In other words, in quantitative terms DOC trends are much higher in the organic layer than in the mineral soils but, in relative terms, DOC is increasing in the same proportion (Table S3). Because the aim of this study is to explain the high heterogeneity of DOC trends found across Europe, instead of the quantification of the trends at local scale, the relative trends were discussed throughout the manuscript. Consequently, our results should be interpreted with caution, keeping in mind that the relations between DOC trends and explaining factors are discussed only from a relative point of view.

Nevertheless, the statistical analyses (LMM, SMK, PMK and BFAST) were done on the absolute value and the resulting Sen's slopes were then standardized. Thus, the fact that trends are expressed in relative terms has consequences on the interpretation of the results, but has no influence on the statistical test itself (carried out on the absolute values of DOC), that is, on the significance and direction of the trends.

Table S3. Comparison of median relative trend slope (rslope in % yr-1) and absolute trend slope (abs slope in mg L^{-1} yr^{-1}) of DOC concentrations in soil solution and their interquartile range using the Seasonal Mann-Kendall test (SMK). (O: organic layer, M02: mineral soil 0-20 cm, M24: mineral soil 20-40 cm, M48: mineral soil 40-80 cm, M8: mineral soil > 80 cm.)

Soil depth	rslope (% yr^{-1})	abs slope (mg L^{-1} yr^{-1})
O	1.18 (±3.37)	0.32 (±1.2)
M02	0.04 (±3.41)	0.008 (±0.52)
M24	0.61 (±8.62)	0.025 (±0.48)
M48	1.01 (±4.79)	0.013 (±0.22)
M8	1.18 (±9.39)	0.032 (±0.31)

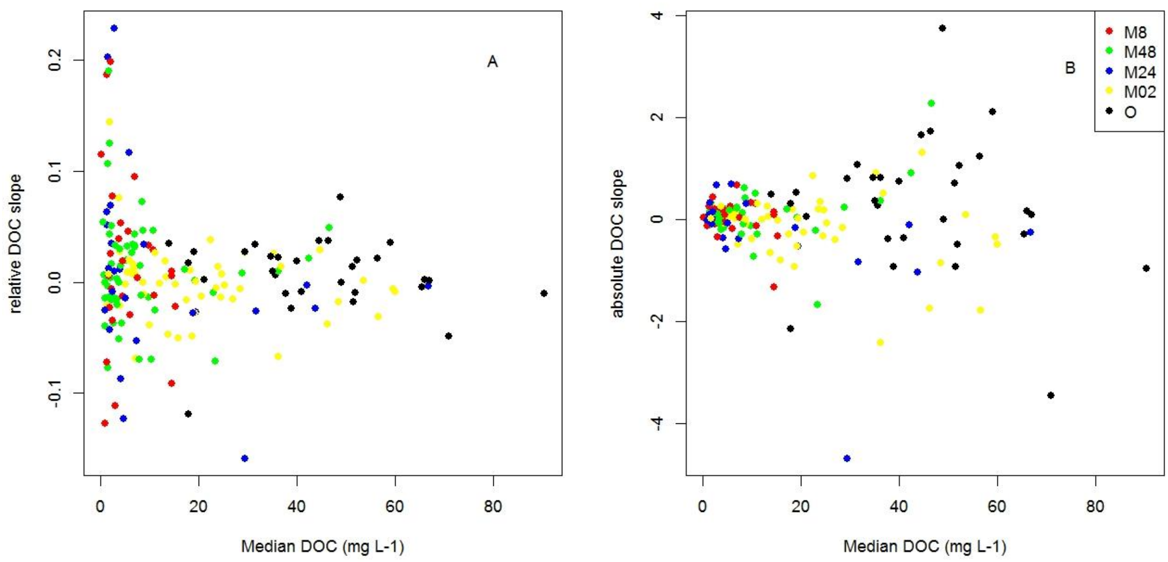

Figure S5. A) Standardized trends (relative DOC slope) versus median DOC concentrations. B) Absolute trends (absolute slope DOC) versus median DOC concentrations. The different colors represent the different soil layers.

Information on the soil solution chemistry at the studied ICP Forests Level II plots

Table S4. Median soil solution DOC concentrations (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median [DOC]	25% percentile	75% percentile	n	median [DOC]	25% percentile	75% percentile	n
Broadleaved									
TL	O	41.35	28.99	56.05	637	44.56	32.00	59.10	475
	M02	8.80	4.30	21.20	8397	8.68	4.50	23.50	3104
	M24	3.78	1.67	8.90	2584	3.19	1.85	4.76	928
	M48	2.60	1.10	6.40	10635	2.70	1.08	5.80	4634
	M8	2.60	1.17	6.53	4354	2.65	1.53	7.00	1797
ZTL	O	33.33	21.00	51.12	4057	30.88	18.01	51.10	1956
	M02	4.26	3.51	6.28	608	4.30	2.80	9.30	192
	M24	20.44	13.40	34.37	94				0
	M48	3.42	2.61	4.51	427	0.91	0.50	1.64	85
	M8	2.42	2.11	3.62	34				0
Coniferous									
TL	O	49.00	35.10	67.36	2496	50.90	38.20	65.40	693
	M02	15.70	7.09	31.15	10914	12.80	5.90	25.50	5813
	M24	5.72	2.40	16.50	5116	5.00	2.10	21.89	2476
	M48	4.44	2.30	11.40	13979	4.30	2.29	10.90	6431
	M8	3.70	1.60	7.91	5024	4.29	2.55	10.12	1597
ZTL	O	42.92	29.03	60.80	4079	44.60	30.18	60.80	2703
	M02	36.90	22.20	56.40	2781	36.00	24.00	53.00	253
	M24	16.34	8.76	31.59	645				0
	M48	44.00	17.40	62.35	227	13.70	10.30	36.25	251
	M8	4.14	3.28	4.81	84				0

Table S5. Median soil solution pH, 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS	WITHOUT BREAKPOINTS

		median pH	25% percentile	75% percentile	n	median pH	25% percentile	75% percentile	n
Broadleaved									
TL	O	3.9	3.8	4.1	636	3.90	3.80	4.10	518
	M02	4.5	4.2	5.2	8346	4.60	4.20	6.2	3322
	M24	6.3	4.9	7.1	2482	6.10	4.90	6.7	993
	M48	5.1	4.5	6.7	10496	5.10	4.40	6.5	5162
	M8	6.4	4.6	7.8	4228	4.50	4.30	6.46	2115
ZTL	O	5.30	4.40	6.30	4026	5.30	4.30	6.60	2025
	M02	6.15	5.00	7.6	608	5.00	4.80	5.75	227
	M24	4.70	4.50	5	93	0.00	0.00	0	0
	M48	8.30	8.20	8.4	426	5.20	5.10	5.3	108
	M8	8.20	8.00	8.3	34	0.00	0.00	0	0
Coniferous									
TL	O	4.00	3.80	4.40	2496	3.80	3.60	4.00	726
	M02	4.30	4.00	4.7	10634	4.30	4.00	4.7	6930
	M24	4.60	4.30	5	4739	4.60	4.30	4.8	2849
	M48	4.50	4.30	4.9	13596	4.50	4.20	4.9	7462
	M8	4.57	4.30	6.4	4837	4.48	4.29	4.7	1660
ZTL	O	4.02	3.80	4.60	4038	4.00	3.80	4.80	2839
	M02	4.40	4.10	4.9	2412	4.80	4.53	5.3	254
	M24	4.90	4.50	5.4	551	0.00	0.00	0	0
	M48	4.80	4.10	5.1	225	4.40	4.27	4.9	319
	M8	4.70	4.60	4.8	84	0.00	0.00	0	0

Table S6. Median soil solution conductivity (µS cm^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median COND	25% percentile	75% percentile	n	median COND	25% percentile	75% percentile	n
Broadleaved									
TL	O	128.00	93.50	189.50	631	140.00	103.00	212.50	507
	M02	60.00	42.25	99	7651	69.55	45.00	104	3066
	M24	86.00	47.00	180	1503	70.45	45.90	120	548
	M48	68.00	45.00	137	8538	70.00	48.58	145	4320
	M8	148.50	61.63	305.75	3006	133.00	59.00	210	1736

ZTL	O	71.00	48.00	110.00	2750	70.00	46.60	111.00	1489
	M02	63.35	34.00	86.775	608	28.20	19.10	51.05	227
	M24	44.00	28.00	56	93	0.00	0.00	0	0
	M48	282.00	254.00	318	425	19.30	16.38	25.325	108
	M8	485.50	446.50	539.75	34	0.00	0.00	0	0
Coniferous									
TL	O	77.00	56.00	124.00	2425	85.00	65.00	155.00	725
	M02	58.00	31.00	92	9222	61.00	33.00	105.5	5699
	M24	50.00	30.00	97	2954	56.00	31.00	111	1715
	M48	56.00	37.00	94	10270	56.00	37.20	99	6658
	M8	104.00	55.00	207.75	2850	120.50	66.00	259	1118
ZTL	O	65.30	45.00	104.00	2296	64.00	42.30	106.00	1537
	M02	39.20	25.00	59	2627	27.00	20.08	41.1	228
	M24	32.00	21.00	57.95	615	0.00	0.00	0	0
	M48	39.05	28.00	150.5	214	95.85	46.48	155.5	290
	M8	50.00	31.75	69.25	84	0.00	0.00	0	0

Table S7. Median soil solution Ca (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median [Ca]	25% percentile	75% percentile	n	median [Ca]	25% percentile	75% percentile	n
Broadleaved									
TL	O	4.18	1.83	7.85	633	5.369	3.193	9.204	515
	M02	2.12	0.80	5.3	8381	2.80	1.04	9.56525	3396
	M24	4.09	1.50	14.18	2555	3.69	0.92	9.005	999
	M48	2.31	0.70	9.385	10600	2.80	0.92	7.7	5204
	M8	5.68	1.50	41.7825	4322	2.80	0.51	13.75	2151
ZTL	O	4.10	2.05	7.06	4049	3.90	1.40	6.36	2030
	M02	8.33	1.67	13.59	608	1.23	0.75	2.425	227
	M24	2.35	1.25	3.296	94	0.00	0.00	0	0
	M48	58.86	51.26	67.485	419	0.72	0.58	1.06	108
	M8	73.75	60.78	92.8	34	0.00	0.00	0	0
Coniferous									

TL	O	3.36	1.47	6.39	2490	1.55	0.98	3.66	722
	M02	0.66	0.25	1.72	10890	1.00	0.36	2.45	6985
	M24	0.82	0.30	1.8665	5079	0.90	0.30	1.61	2901
	M48	0.82	0.32	2.07	13901	0.92	0.32	2.285	7511
	M8	2.10	0.49	10.6575	4986	1.97	0.53	8.285	1700
ZTL	O	1.50	0.72	2.80	4052	1.50	0.72	2.80	4052
	M02	1.13	0.53	2.14	2777	1.13	0.53	2.14	2777
	M24	1.20	0.62	2.31	644	1.20	0.62	2.31	644
	M48	3.00	1.81	3.895	227	3.00	1.81	3.895	227
	M8	0.76	0.47	1.1975	84	0.76	0.47	1.1975	84

Table S8. Median soil solution Mg (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median [Mg]	25% percentile	75% percentile	n	median [Mg]	25% percentile	75% percentile	n
Broadleaved									
TL	O	1.05	0.48	1.90	633	1.18	0.62	2.08	515
	M02	0.80	0.42	1.5	8382	0.86	0.51	1.46	3395
	M24	1.01	0.50	2.13	2563	1.18	0.62	2.295	999
	M48	0.95	0.37	2.0745	10611	1.02	0.46	2.19	5205
	M8	1.72	0.73	3.94	4323	1.29	0.51	2.88	2152
ZTL	O	1.06	0.61	1.80	4049	0.98	0.57	1.60	2029
	M02	0.70	0.28	1.05	608	0.32	0.21	0.545	227
	M24	0.63	0.30	0.808	94	0.00	0.00	0	0
	M48	0.63	0.50	0.785	419	0.29	0.24	0.33	108
	M8	3.76	3.18	4.01	34	0.00	0.00	0	0
Coniferous									
TL	O	0.72	0.33	1.24	2490	0.24	0.17	0.63	722
	M02	0.36	0.20	0.68	10899	0.47	0.28	0.84	6990
	M24	0.40	0.22	0.898	5081	0.40	0.22	0.83	2902
	M48	0.44	0.21	0.9	13910	0.55	0.31	1.1	7518
	M8	0.98	0.39	1.875	4990	0.93	0.50	2	1699
ZTL	O	0.40	0.20	0.76	4061	0.40	0.20	0.83	2789
	M02	0.37	0.20	0.616	2773	0.49	0.38	0.6375	262

	M24	0.44	0.25	0.927	644	0.00	0.00	0	0
	M48	0.76	0.49	3.725	227	0.55	0.35	0.91	321
	M8	0.85	0.37	1.3425	84	0.00	0.00	0	0

Table S9. Median soil solution S-SO_4^{2-} (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median [SO_4^{2-}]	25% percentile	75% percentile	n	median [SO_4^{2-}]	25% percentile	75% percentile	n
Broadleaved									
TL	O	2.50	1.30	4.17	592	3.20	1.63	4.58	476
	M02	2.00	1.33	3.3875	8383	1.93	1.19	3.3	3370
	M24	2.63	1.60	3.8	2556	2.70	1.98	3.565	1007
	M48	2.80	1.50	4.7	10571	3.10	1.90	5.5	5188
	M8	4.04	2.83	6.371	4323	5.05	3.10	9.2	2116
ZTL	O	1.01	0.60	1.70	4041	0.86	0.53	1.40	2029
	M02	0.75	0.52	1.21275	608	0.76	0.63	0.8785	227
	M24	2.05	1.02	3.15975	94	0.00	0.00	0	0
	M48	1.06	0.80	1.52	426	0.79	0.67	0.8625	108
	M8	10.38	9.15	11.855	34	0.00	0.00	0	0
Coniferous									
TL	O	1.27	0.67	2.30	2483	0.80	0.46	1.37	722
	M02	1.51	0.90	3	10885	1.94	1.08	3.608	7021
	M24	2.39	1.40	3.862	5086	2.25	1.40	3.558	2933
	M48	2.96	1.60	4.6	13941	2.90	1.70	4.63	7537
	M8	4.34	2.42	7.2	4977	5.46	3.13	9.30125	1672
ZTL	O	0.71	0.34	1.48	4064	0.67	0.31	1.38	2800
	M02	0.66	0.38	1.337	2776	0.57	0.42	0.77	261
	M24	1.74	0.77	4.5975	644	0.00	0.00	0	0
	M48	1.20	0.89	11.315	226	4.45	1.30	8.291	318
	M8	1.33	1.09	1.60325	84	0.00	0.00	0	0

Table S10. Median soil solution N-NO_3^- (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median $[NO_3^-]$	25% percentile	75% percentile	n	median $[NO_3^-]$	25% percentile	75% percentile	n
Broadleaved									
TL	O	3.74	1.46	9.29	617	4.88	1.94	11.04	518
	M02	0.56	0.04	2.5285	8123	0.91	0.24	2.6825	3372
	M24	0.50	0.02	3.23	2535	0.62	0.02	2.8615	991
	M48	0.26	0.02	1.659	10358	0.33	0.03	2.3	5165
	M8	0.40	0.05	5.0275	4218	0.73	0.13	6.1595	2002
ZTL	O	1.60	0.56	3.79	3975	1.03	0.21	2.60	1994
	M02	0.86	0.40	1.8725	608	0.70	0.30	1.6	227
	M24	0.47	0.14	0.87975	94	0.00	0.00	0	0
	M48	0.35	0.06	0.8	423	0.52	0.23	0.8525	108
	M8	0.02	0.02	0.022	34	0.00	0.00	0	0
Coniferous									
TL	O	1.14	0.16	4.19	2388	1.06	0.08	4.87	677
	M02	0.14	0.02	1.3	10431	0.27	0.02	1.87775	6940
	M24	0.17	0.02	1.267	4745	0.10	0.02	1.334	2844
	M48	0.10	0.02	1.2	13195	0.11	0.02	1.3	7194
	M8	0.27	0.02	1.0895	4971	0.37	0.06	1.2	1691
ZTL	O	0.56	0.13	1.74	4055	0.34	0.05	1.18	2777
	M02	0.02	0.02	0.06	2275	0.05	0.02	0.17	260
	M24	0.02	0.02	0.03	489	0.00	0.00	0	0
	M48	0.02	0.02	0.09875	226	0.65	0.03	7.988	321
	M8	2.54	0.50	4.6805	84	0.00	0.00	0	0

Table S11. Median soil solution Al (mg L^{-1}), 25% and 75% percentiles and number of observations (n) for the different forest types, soil depth intervals and collector types with the entire dataset (with breakpoints) and with the dataset without time series showing breakpoints (without breakpoints).

		WITH BREAKPOINTS				WITHOUT BREAKPOINTS			
		median [Al]	25% percentile	75% percentile	n	median [Al]	25% percentile	75% percentile	n
Broadleaved									

TL	O	0.38	0.17	0.76	574	0.30	0.15	0.76	490
	M02	0.81	0.39	1.62	7767	0.78	0.30	1.7	3107
	M24	0.05	0.02	0.387	2406	0.05	0.02	0.333	979
	M48	0.30	0.02	1.02	9871	0.30	0.02	1	4918
	M8	0.05	0.02	0.87	4180	0.91	0.17	2.79	2101
ZTL	O	0.17	0.06	0.32	3278	0.12	0.03	0.22	1536
	M02	0.14	0.02	0.45	577	0.22	0.14	0.35	222
	M24	0.37	0.22	0.48	94	0.00	0.00	0	0
	M48	0.02	0.02	0.04	378	0.14	0.09	0.21	107
	M8	0.02	0.02	0.02	30	0.00	0.00	0	0
Coniferous									
TL	O	1.14	0.74	1.79	2162	0.93	0.59	1.27	622
	M02	1.35	0.69	2.19	10398	1.44	0.72	2.44875	6514
	M24	0.92	0.36	2.2145	4871	0.90	0.38	2.391	2762
	M48	1.11	0.38	2.341	13454	0.96	0.32	2.2	7157
	M8	1.58	0.02	3.399	4857	2.63	1.01	5.475	1674
ZTL	O	0.24	0.12	0.49	3944	0.21	0.11	0.39	2704
	M02	0.87	0.44	1.48	2709	1.10	0.81	1.7	262
	M24	0.73	0.22	1.7235	611	0.00	0.00	0	0
	M48	2.01	1.20	7.015	210	2.95	1.90	5.568	303
	M8	1.62	1.01	2.3275	66	0.00	0.00	0	0

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