Supplementary material for

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

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Supplementary material S1. List of ICP Forests Level II plots 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	
Finland	15_5	1997	2011		Picea abies	Lack	
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	
Finland	15_19	1999	2011		Picea abies	Lack	
Finland	15_20	1998	2011		Pinus sylvestris	Lack data	
Finland	15_21	2000	2010		Picea abies	Lack	

Finland	15_23	1998	2010		Picea abies	Lack	
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	

Supplementary material S2. 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).

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

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. 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 (Fig. 4 and 5).

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.

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.

Trends for 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: 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 multivariate statistical analyses to investigate the main factors explaining differences in DOC trends among the selected plots. Firstly, we used General Discriminant Analysis (GDA) (Raamsdonk et al., 2001) to determine the importance of soil solution and deposition variables in the separation of groups with different trend classes (P, N, NS) in DOC. We also accounted for the part of the variance due to the different soil layers (depth interval) as an independent categorical variable. Secondly, we applied Structural Equation Models (SEM) to test whether deposition variables had an effect (direct, indirect or total) on DOC trends 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 Supplementary Material S3). 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 GDA and SEM. 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 GDA that was performed using Statistica 6.0 (StatSoft, Inc. Tule, Oklahoma, USA) and the LMMs that were performed using SAS 9.3 (SAS institute, Inc., Cary, NC, USA).

Supplementary material S3. 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 Figure 6 and the Structural Equation Models (SEM) (Figure 11), as the number of cases available for each depth will be 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.

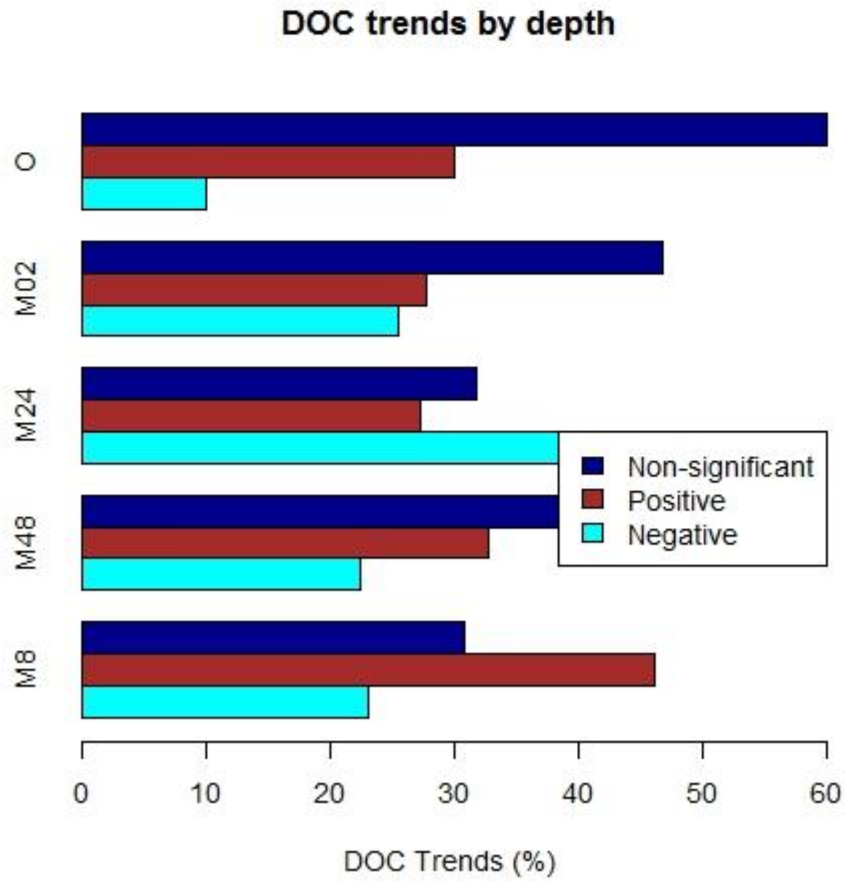

Figure S1. Percentage of non-significant, positive and negative trends per soil depth interval (O: organic layer, M02: mineral soil 0-20 cm, M24: mineral soil 20-40 cm, M48: mineral soil 40-80 cm, M8: mineral soil > 80 cm).

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 count with 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 (Figure S2).

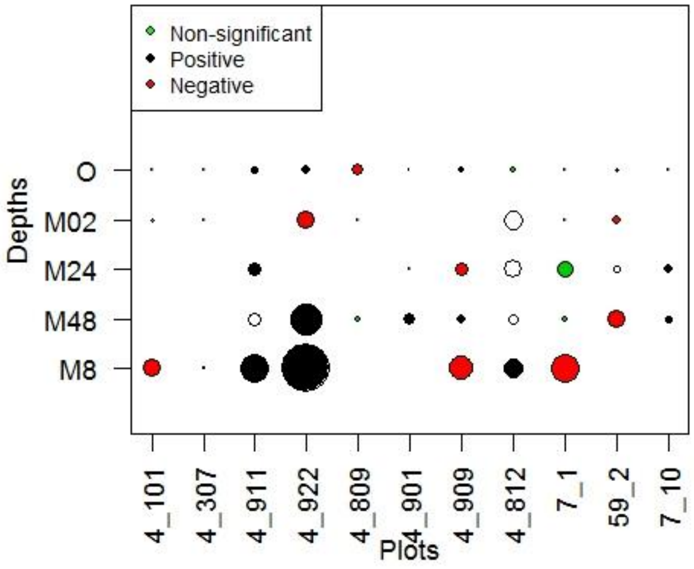

Figure S2. Direction of the trend (non-significant, positive and negative) per soil depth interval (O: organic layer, M02: mineral soil 0-20 cm, M24: mineral soil 20-40 cm, M48: mineral soil 40-80 cm, M8: mineral soil > 80 cm) for the 11 plots with DOC measured at least at 3 soil depth intervals including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

Supplementary material S4. Structural equation model with trends in SO_4^{2-} and NO_3^- deposition

The same structural equation models (SEM) represented in Figure 11 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}) (Figure S3). The SEMs for all the cases and for cases with low and medium N deposition are shown in Figure S3.

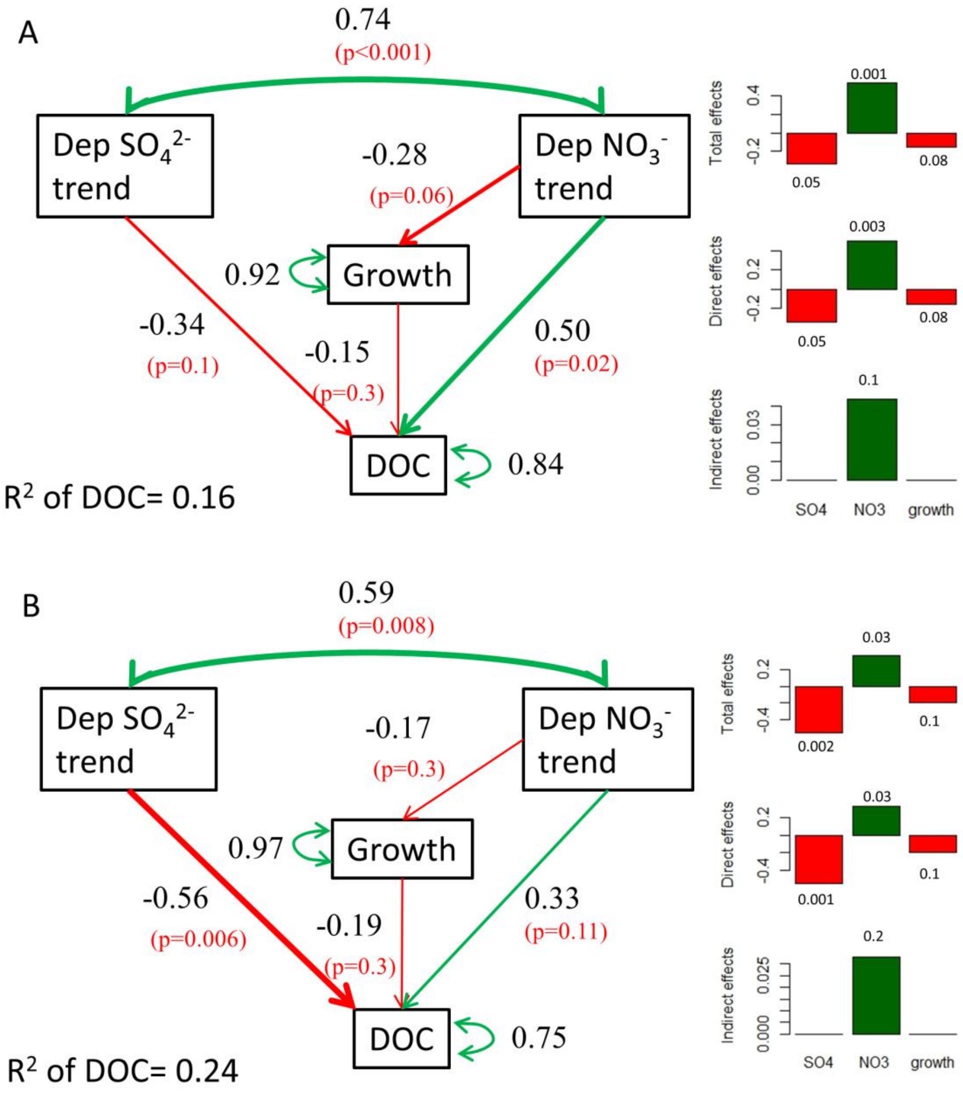

Figure S3. Diagram of the structural equation model (SEM) that best explains the maximum variance of the resulting trends of DOC concentrations in soil solution for: A) all the cases and B) cases with low or medium N deposition, with trends in SO_4^{2-} and NO_3^- deposition (% yr^{-1}) with direct effects 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.

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