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Figure S1

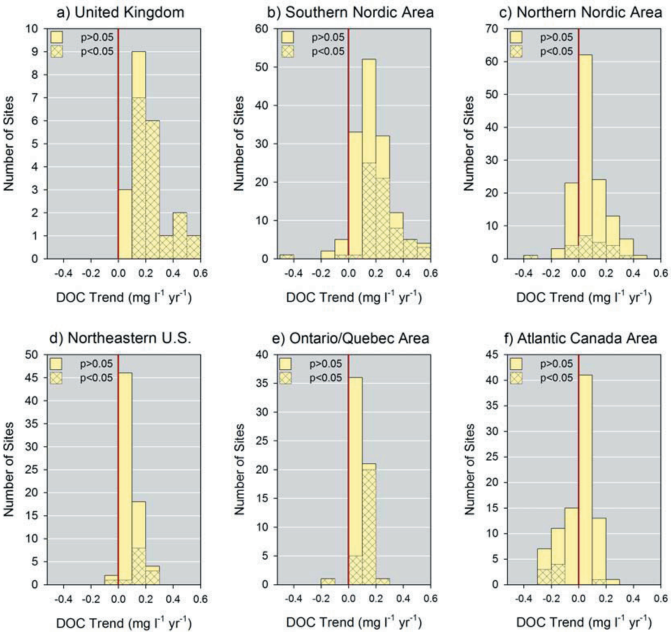

Figure S1: Histograms of trend magnitudes (and significance) for regions of Europe (upper panels) and North America (lower panels). Bars show total number of sites in each trend class; cross-hatched portions of bars represent trend slopes significant at $p < 0.05$. Trends in all regions except Atlantic Canada are dominated by positive slopes.

Figure S2

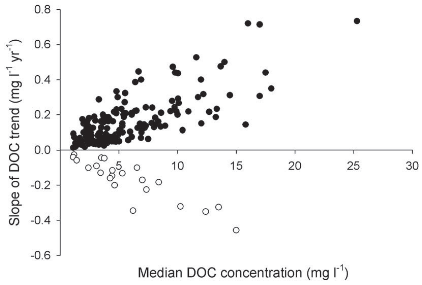

Figure S2: Relationship between DOC trend (mg l^{-1} yr^{-1}) and absolute DOC concentration (presented as the median at each site for the period 1990-2004). Positive and negative trends ($p< 0.05$) indicated by filled and empty circles respectively. Because DOC change is strongly related to absolute DOC concentration, we use % change in DOC (DOC trend/median concentration) in many of our analyses.

Figure S3

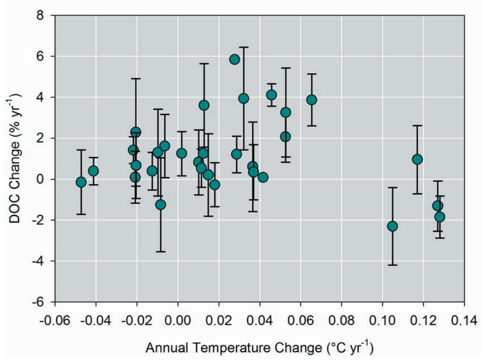

Figure S3: Relationship between relative DOC change at surface water monitoring sites and trends in annual air temperature (both for the period 1990-2004), determined from CRUTEM2 simulated air temperature anomaly data from the Climate Research Unit, University of East Anglia (http://www.cru.uea.ac.uk/cru/data/tem2/) compiled in a 5x5 degree grid. Multiple surface water monitoring sites are located in each temperature grid cell, resulting in a range of DOC trends for each estimated (grid-based) change in air temperature.

Table S1

Region	Period	Data Type	No. Sites	No. of significant trends (p<0.05) (-/+ = negative/positive slope)									
				ANNUAL		WINTER		SPRING		SUMMER		AUTUMN	
				-	+	-	+	-	+	-	+	-	+
New England, N.E. United States†	1990-2004	discharge	5	0	0	0	0	0	0	0	0	0	0
Adirondacks, N.E. United States†	1990-2004	discharge	4	0	0	0	0	0	0	0	0	0	0
N. Appalachians, N.E. United States†	1990-2004	discharge	6	0	0	1	0	0	1	0	2	0	0
Southern Norway‡	1970's-2004	discharge	3	0	0	0	2	1	0	1	0	1	0
Southern Finland*	1988-2003	discharge	3	0	0	0	0	0	0	0	0	1	0
United Kingdom$	1990-2004	precipitation	11	0	0	0	0	0	1	0	0	0	0
Sudbury, Ontario, Canada#	1990-2004	precipitation	1	0	0	0	0	0	0	0	0	0	0

† Data source: http://waterdata.usgs.gov/nwis. Sites are continuously gauged streams closest to monitoring sites used in DOC trend analysis;
‡ Data source: De Wit et al. (in press)[18]. Sites (periods): Birkenes (84-03) , Storgama (75-03), Langtjern (77-03).
* Data source: Vuorenmaa et al. (2006)[10]. Sites (periods): Rudback (91-03); Valkea-Kotinen (88-03); Pieni Hietjärvi (88-03).
$ Data source: UK Met Office, Historic Station data. http://www.metoffice.gov.uk/climate/uk/stationdata. Sites chosen to represent regions where monitoring sites used in the DOC trend analysis occur. Sites: Braemar, Stornaway, Paisley, Newton Rigg, Ringway, Aberporth, Shawbury, Valley, Armagh, St Mawgan, Eastbourne.
# Data source: Sudbury Airport Meteorological Station.

Table S1: Summary of hydrologic trends in regions characterised by large increases in dissolved organic carbon. Hydrologic data (either from gauged streams or precipitation stations) are available from the majority of regions where DOC has risen most sharply. Hydrologic changes can have profound effects on DOC export[3], at least in the short term. Increased flows may lead to runoff more dominated by chemical constituents accumulated from shallow soil layers (where much of the production of DOC occurs)[14]; moreover, changes in stream discharge in the absence of changing DOC flux can lead either to decreased DOC values (through dilution) or to increased DOC levels (through concentration)[16]. In this assessment significant trends ($p <0.05$; Mann-Kendall Test) are rare, and show no overall positive or negative tendency.

Table S2

Region	Number of 5 degree grid cells representing surface water sites	No. of grid cells showing significant trends ($p<0.05$) (-/+ = negative/positive slope)									
		ANNUAL		WINTER		SPRING		SUMMER		AUTUMN	
		-	+	-	+	-	+	-	+	-	+
Northern Nordic	12	0	0	2	0	0	0	0	2	0	1
Southern Nordic	9	0	0	0	0	0	0	0	0	0	0
UK	5	0	2	0	0	0	0	0	0	0	1
United States	2	0	0	0	0	0	0	0	0	0	0
Ontario/Quebec	3	0	0	0	0	0	0	0	0	0	1
Atlantic Canada	5	0	3	0	3	0	0	0	0	0	0

Table S2: Summary of air temperature trends (Mann-Kendall Test) by region determined from CRUTEM2 simulated air temperature anomaly data from the Climate Research Unit, University of East Anglia (http://www.cru.uea.ac.uk/cru/data/tem2/) compiled in a 5x5 degree grid. Increasing temperature, and its subsequent effects on organic matter decomposition rates, have been widely proposed as an explanation for increasing DOC[3,4,15]. However, CRUTEM2 data provided no indication of widespread increases in seasonal or annual air temperatures across most regions over the 1990-2004 period. Consistent upward temperature trends were more widespread in eastern North America, but were most common in parts of Atlantic Canada where there was no dominant DOC tendency.

Table S3

Region	Total number of sites in region	No. of sites showing significant trends ($p<0.05$) (-/+ = negative/positive slope)	
		-	+
Northern Nordic	134	42	0
Southern Nordic	149	32	10
UK	22	0	5
United States	70	18	1
Ontario/Quebec	59	8	4
Atlantic Canada	88	0	8

Table S3: Summary of trends (Mann-Kendall Test) in nitrate concentration by region for sites in this dataset. Findlay[6] hypothesised that increasing DOC in the Hudson River results from higher rates of soil microbial processing (e.g., decomposition) due to the accumulation of atmospherically deposited nitrogen. Assessment of trends in nitrate concentration are of restricted value in this study since, unlike sulphate and chloride, nitrate is not conservative. Consistent upward trends in concentration in regions with DOC increases, however, would provide an indication that increased nitrogen saturation was influencing DOC concentrations. Significant trends are uncommon and the majority of these are downward.

Table S4

	Effect of ΔxSO_4^{2-}	Effect of ΔCl^- + ΔmSO_4^{2-}	% of total anion effect from ΔxSO_4^{2-}
UK	1.24 (1.32)	0.94 (0.87)	56.9
Southern Nordic	2.28 (1.06)	0.32 (0.81)	87.7
Northern Nordic	0.81 (0.49)	0.08 (0.38)	91.0
USA	1.08 (0.47)	-0.03 (0.19)	97.3
Ontario/Quebec	1.54 (0.85)	0.002 (0.02)	99.9
Atlantic Canada	0.46 (0.26)	-0.75 (0.65)	38.01

Table S4.: Estimates of mean effects (standard deviation in parentheses) for each region of the change in anthropogenic sulphate (ΔxSO_4^{2-}) and the combined effects of the change in chloride (ΔCl^-) and marine sulphate (ΔmSO_4^{2-}) on the percentage change in DOC, according to the coefficients in the model presented in Table 1 (see Methods). The effect of ΔCl^- was determined from the coefficients for ΔCl^- and the interaction term (ΔCl^-xCaMg). In the final column the sign for the mean effect of marine anions (column 2) is ignored. The results show that, on average, ΔxSO_4^{2-} is responsible for >85% of the DOC trend in all regions other than the UK (where anthropogenic and marine effects were similar) and Atlantic Canada (where marine effects dominated and, uniquely, the mean DOC trend was negative).

Table S5

	ΔSO_4^{2-}	ΔCl^-	CaMg*	ΔCl^-xCaMg*
ΔCl^-	0.468			
CaMg*	-0.419	-0.076		
ΔCl^-xCaMg*	0.507	0.781	-0.288	
%ΔDOC	-0.487	-0.448	-0.052	-0.253

Table S5: Pearson correlation coefficients for variables in the final multiple regression model (see Methods).
ΔSO_4^{2-} = Change in SO_4^{2-} ($\mu eq\ l^{-1}\ yr^{-1}$); ΔCl^- = Change in Cl^- ($\mu eq\ l^{-1}\ yr^{-1}$); CaMg* = Median CaMg* ($\mu eq\ l^{-1}$); ΔCl^-xCaMg* = Interaction term, Median CaMg* ($\mu eq\ l^{-1}$) x Change in Cl^- ($\mu eq\ l^{-1}\ yr^{-1}$); %ΔDOC = percentage change in DOC .