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The importance of the relationship between scale and process in understanding long-term DOC dynamics

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ABSTRACT

Concentrations of dissolved organic carbon have increased in many, but not all, surface waters across acid impacted areas of Europe and North America over the last two decades. Over the last eight years several hypotheses have been put forward to explain these increases, but none are yet accepted universally. Research in this area appears to have reached a stalemate between those favouring declining atmospheric deposition, climate change or land management as the key driver of long-term DOC trends. While it is clear that many of these factors influence DOC dynamics in soil and stream waters, their effect varies over different temporal and spatial scales. We argue that regional differences in acid deposition loading may account for the apparent discrepancies between studies. DOC has shown strong monotonic increases in areas which have experienced strong downward trends in pollutant sulphur and/or seasalt deposition. Elsewhere climatic factors, that strongly influence seasonality, have also dominated inter-annual variability, and here long-term monotonic DOC trends are often difficult to detect. Furthermore, in areas receiving similar acid loadings, different catchment characteristics could have affected the site specific sensitivity to changes in acidity and therefore the magnitude of DOC release in response to changes in sulphur deposition. We suggest that confusion over these temporal and spatial scales of investigation has contributed unnecessarily to the disagreement over the main regional driver(s) of DOC trends, and that the data behind the majority of these studies is more compatible than is often conveyed.

1. Introduction

There have been widespread observations of increased dissolved organic carbon (DOC) concentrations in surface waters across parts of Europe and North America over the last two decades (Driscoll et al., 2003; Worrall et al., 2004; Evans et al., 2005; Skjelkvale et al., 2005). This has raised concerns about drinking water treatment and the production of carcinogenic byproducts (Gallard and von Gunten, 2002; Holden et al., 2007), and the further possibility that climate change is causing degradation of soil carbon stores (Freeman et al., 2001a; Bellamy et al., 2005). In both cases there is a common perception that DOC increases are likely to be environmentally detrimental, and increasingly land managers are seeking guidance from the scientific community with respect to practical methods to control or even reverse these trends.

Several hypotheses have been put forward to explain increasing DOC trends (Table 1). One hypothesized driver for increasing DOC trends is a long-term change in the chemistry of atmospheric deposition that has been recorded across many of these areas as a result of reductions in anthropogenic sulphur and, in some locations, seasalt deposition (Evans et al., 2006; Vuorenmaa et al., 2006; de Wit et al., 2007; Monteith et al., 2007; Dawson et al., 2009; Hruska et al., 2009; Oulehle and Hruska, 2009). However, others have rejected this hypothesis, arguing that DOC trends are more consistent with changes in rainfall, temperature and/or atmospheric carbon dioxide (CO_2) than declining atmospheric sulphur deposition (Worrall and Burt, 2007a; Eimers et al., 2008c; Lepisto et al., 2008; Sarkkola et al., 2009), building on earlier studies suggesting relationships between these drivers and increased DOC (Freeman et al., 2001a; Freeman et al., 2004; Hongve et al., 2004; Fenner et al., 2007). Some reject the deposition hypothesis outright as DOC concentrations have decreased in some areas where acid deposition has declined (Clair et al., 2008). Other drivers have also been suggested; these include changing nitrogen deposition (Findlay, 2005), solar radiation in boreal lakes (Hudson et al., 2003), and land management practices (Yallop and

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Table 1
Summary of published research about long-term trends in DOC concentrations. Sites are typically 'acid sensitive', with range of soils (peat, podzols and mineral soils) with forest and/or moorland vegetation cover. Countries are Canada (CA); Czech Republic (CzR); Finland (FI); Norway (NO); Sweden (SE); United Kingdom (UK); United States of America (USA). *Significant monotonic trend that is either increasing (+), decreasing (-) or has no significant trend (nt). Information not reported (nr). Acid deposition quantified in terms of sites with 'high' (H) or 'low' (L) deposition. Text typed in italics is information based on authors knowledge and not reported in the specific paper. Water body is classified as lake (L) or stream (S). Statistical methods are summarized as: Seasonal Kendall test and Sen slope (SKT); Mann-Kendall test and theil slope (MKT); correlation (C); linear regression (LR); multiple linear regression (MLR); mixed-effect model (MEM); process-based model (PM); artificial neural network (ANN); Student's T-Test (TT). Table rows are ordered in terms of disagreement, agreement or no mention of acid deposition hypothesis as driver of DOC trends. NB this is a summary of research and does not include all papers published on DOC trends.

Paper	Region	No. Site	DOC trend*			Driver of trend							Catchment			Monitoring			Statistical method
			+	nt	-											Time period		Sample frequency	
						Seasalt dep.	Acid dep.	Nitrogen enr.	Atmos. CO_2	Temperature	Preip./runoff	Management	Historic acid deposition	Area (km^2)	Lake/stream	Start	End		
Freeman et al. (2001a)	UK	22	20	2	0		✗			✓		✗	*H–L*	*0.5–16*	L/S	1988	2000	1–3 month	SKT
Hudson et al. (2003)	CA	9	nr	nr	nr		✗			✗	✓		nr	0.9–5.9	L	1978	1998	5–24/year	MLR
Hongve et al. (2004)	NO	24	24	0	0					✗	✓		*H*	0.1–9	L	1983	2001	>1 year	TT
Worrall et al. (2004)	UK	198	153	45	0		✗	✓		✓	✓	✓	H–L	0.04–2100	L/S	1961/	2000	nr	SKT
Striegl et al. (2005)	USA	1	0	0	1					✓			L	831400	S	1978	2003	6–8/year	ANCOVA
Worrall and Burt (2007b)	UK	315	216	44	55		✗		✓	✓	✓		H–L	nr	L/S	1962/	2002	1–4 weeks	SKT/ MLR
Clair et al. (2008)	CA	3	0	1	2		✗						L	17–297	S	1983/	2004	1 week	SKT
Eimers et al. (2008a)	CA	7	6	1	0		✗	✗		✗	✓		nr	0.1–1.9	S	1980	2001	1–2 weeks	MKT/MLR
Lepisto et al. (2008)	FI	1	0	1	0		✗			✓	✓	✗	L	3160	S	1962	2005	~3–1month	MKT
Sarkkola et al. (2009)	FI	8	7	1	0		✗			✓	✓		L	0.2–4.9	S	1979	2006	<1–4weeks	SKT/MEM
Hejzlar et al. (2003)	CzR	1		0	1		✓						H	438	S	1969	1983	1 day	SKT; MLR
			1				✓			✓	✓					1984	2000		
Findlay (2005)	USA	1	1	0	0		✓	✓		✗	✗	✗	H	21 000	S	1988	2003	1–4 weeks	LR
Evans et al. (2006)	UK	11	11	0	0	✓	✓			✓	✗		*H–L*	*0.5–16*	L	1988	2003	3 month	MLR
Vuorenmaa et al. (2006)	FI	13	10	3	0		✓				✗		nr	0.3–4.36	L	1987	2003	1 month	SKT/C/MLR
de Wit et al. (2007)	NO	3	1	2	0		✓			✗	✗		H	0.4–0.8	S	1985	2003	1 week	SKT/MLR
Monteith et al. (2007)	UK, SE, NO, FI, USA, CA	522	363	20	139	✓	✓			✗	✗		*H–L*	nr	L/S	1990	2004	nr	SKT/MLR
Erlandsson et al. (2008)	SE	28	nr	nr	nr	✗	✓			✗	✓		*H–L*	210–26800	S	1970	2004	1 month	MKT/MLR
Futter et al. (2008)	FI	1	1	0	0		✓			✓	✓		L	0.3	L/S	1992	2001	nr	PM/ANN/ MKT
Dawson et al. (2009)	UK	2	2	0	0		✓			✗	✗		*H–L*	nr	S	1986	2007	~1 week	MEM
Hruska et al. (2009)	CzR	2	2	0	0		✓			✗	✗		H	0.2–0.3	S	1993	2007	1 week	LR/ SKT
Oulehle and Hruska (2009)	CzR	11	9	2	0		✓			✗	✗	✗	H	8–74	L/S	1969	2006	~1 month	SKT/MLR